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# CODISPOSAL OF DIMINIMUS LEVELS OF LOW-LEVEL RADIOACTIVE WASTE AND SANITARY WASTE

By

E.S.K. Chian S.B. Ghosh B. Kahn M. Giabbai F.G. Pohland

Supported by

U.S. Department of Energy Idaho Operations Office Idaho Falls, Idaho 83407

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February 1986

# **GEORGIA INSTITUTE OF TECHNOLOGY**

A UNIT OF THE UNIVERSITY SYSTEM OF GEORGIA SCHOOL OF CIVIL ENGINEERING ATLANTA, GEORGIA 30332



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\*Principal Investigator Georgia Institute of Technology Atlanta, Georgia 30332

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#### SUMMARY

Factors Affecting the Mobility of Selected Radionuclides Codisposed with Municipal Refuse Within Landfills

Disposal of Low-Level Radioactive Waste (LLRW) has been accomplished by shallow land burial at licensed commercial sites since 1962. At some of the sites, however, subsurface migration of radionuclides, caused by leaching due to infiltration of groundwater, surface-water runoff, and incidental rainfall, have been reported. In addition, the gradual depletion of available volume for LLRW disposal at operational licensed commercial sites, due to site closure and increased generation of LLRW, has become a concern, since it may result in an increase in the volume requirements for storage of LLRW. Furthermore, the restrictions imposed on LLRW disposal, from January 1, 1986, by the Low-Level Radioactive Waste Policy Act of 1980 (PL96-573) may contribute to an additional increase in storage requirements for LLRW, since most of the states or the regional compacts (comprised of member states) currently do not have an operational site for LLRW disposal, and one of the provisions in this Act allows each state or a regional compact the authority to exclude LLRW generated outside its borders. In order to reduce the volume requirements for LLRW disposal, one of the suggested alternatives to shallow land burial of LLRW was the regulated release of low-activity wastes to a municipal sanitary

landfill. This alternative, namely, codisposal of selected radionuclides with municipal refuse within landfills, has been evaluated in this study.

The containment of the radionuclides, that are codisposed with municipal refuse, within a landfill during the landfill stabilization phase and the post-closure phase would allow the radionuclides to decay and minimize the possibility of impairment to the environmental setting around the landfill. In view of containment of LLRW codisposed with municipal refuse, leachate recycle would be the preferred mode of leachate management, since it would allow the LLRW to undergo their natural decay within the landfill. Also, the landfill should be lined and incorporate in its design the following structural elements: ' leachate and gas collection systems, a leak detection system, and provisions for leachate recycle.

The major objectives of this study were to evaluate the behavior of selected radionuclides (Co-58, Sr-85, and Ce-141) codisposed with municipal refuse within pilot-scale landfills during landfill stabilization, and to assess the potential for solubilization of Co-58, from cobalt sulfide precipitates, due to formation of complexes with humic substances present in leachate. A separate confirmation of such behavior was provided by the results of companion laboratory-scale studies on investigation of the potential of humic substances present in leachate to mobilize Co-58 from cobalt sulfide precipitate.

Two pilot-scale landfills, 3.05-m square and 4.28-m deep, were utilized to evaluate the factors affecting the mobility of selected radionuclides (Co-58, Sr-85, and Ce-141) codisposed with municipal refuse during landfill stabilization. These landfills were reinforced concrete lysimeters, lined with a 0.76-mm (30-mil) high density polyethylene liner, and had provisions for leachate collection and recirculation. The municipal refuse placed within the cells was spiked with Co-58, Sr-85, and Ce-141 at levels of 0.91 mCi, 1.09 mCi, and 5.78 mCi, respectively. The resulting loading levels, based on dry weight of refuse, for Co-58, Sr-85, and Ce-141 were 1.6 x  $10^{-7}$  Ci/kg, 2.0 x  $10^{-7}$  Ci/kg, and 1.06 x  $10^{-6}$  Ci/kg, respectively. Water was added to the landfills to simulate infiltration due to incidental rainfall in Atlanta, GA during the investigation, and leachate recycle was practiced in one of the landfills. During the study, the extent to which radionuclides and organics leached from both landfills was recorded. Analyses on leachate samples included detection of Co-58, Sr-85, and Ce-141 along with the indicator parameters, namely, pH, ORP, conductivity, total alkalinity, COD, BOD5, TOC, TIC, volatile fatty acids (acetic, propionic, isobutyric, butyric, and valeric), sulfide, chloride, iron, manganese, nickel, zinc, cobalt, and strontium.

Based on evaluation of the data on radionuclides and indicator parameters, and the known progression of events that occur within a landfill undergoing stabilization, the following major findings complement the state-of-the-art knowledge on codisposal of LLRW with municipal refuse within landfills:

i. The selected radionuclides, Co-58, Sr-85, and Ce-141, can be codisposed with municipal refuse at the loading levels utilized in this study.

ii. The immobilization of Co-58 and Sr-85 present in leachate was

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due to precipitation of these radionuclides as cobalt sulfide and strontium carbonate, while the immobilization of Ce-141 may be attributed to adsorption of the radionuclide onto the refuse.

- iii. The maximum levels of Co-58, Sr-85, and Ce-141, expressed as a percentage of initial loading levels, present in the leachate were 12.7%, 15.7% and less than 0.03%, respectively. The activity levels corresponding to these maximum levels of Co-58, Sr-85, and Ce-141 in leachate were lower than the maximum permissible concentrations of these radionuclides in water above natural background.
- iv. Humic substances, which constitute the major fraction of TOC of leachate after the degradation of readily degradable organics (final maturation phase), solubilize Co-58 from cobalt sulfide precipitate due to the formation of soluble complexes.
- v. Leachate recycle was the preferred mode of leachate management, since it allowed for containment of radionuclides within the landfill due to minimization of removal of leachate from the landfill. This allowed the radionuclides to undergo their natural decay within the landfill.

Thus, the radionuclides, Co-58, Sr-85, and Ce-141 can be safely codisposed with municipal refuse within landfills at the loading levels utilized in this study.

Further studies on the behavior of other radionuclides, which constitute LLRW viz., P-32, Cr-51, Se-75, Mo-99, I-131, Pu-238, and

Pu-240, within landfills during landfill stabilization is recommended, since the data obtained from such studies along with the data obtained from this study will form a data base on the behavior of LLRW within landfills, which in turn could be used to predict the behavior of LLRW with landfills.

#### CHAPTER I

#### INTRODUCTION

#### 1.1 Problem Description

Disposal of Low-Level Radioactive Waste (LLRW) has been accomplished by shallow land burial at licensed commercial sites since 1962. However, at some sites, the current practice of LLRW disposal has resulted in subsurface migration of radionuclides caused by leaching due to infiltration of groundwater, surface-water runoff, and incidental rainfall (1-15, 18). Also, the gradual depletion of the available volume for LLRW disposal at the licensed commercial sites due to site closure, increased generation of LLRW, and volume allotment programs at sites, has become a concern since it may lead to an increase in the storage requirements for LLRW (15-17).

The Low-Level Radioactive Waste Policy Act of 1980 (PL96-573) imposes new restrictions on LLRW disposal. According to the provisions in this Act, effective January 1, 1986, each state or regional compact (comprised of member states) is responsible for assuring adequate disposal of LLRW generated within its borders. Also, this Act allows the states forming a regional compact, to designate a state with a LLRW disposal site as the host state within the regional compact, and the authority to exclude LLRW from outside their region. Thus, these restrictions imposed upon LLRW disposal by the Act will lead to the development of LLRW disposal sites within a state or a regional compact, which currently does not have an operating site. Furthermore, these conditions of the Act may lead to an increase in the storage requirements for LLRW, since most of the states or regional compacts at present do not have an operating site (17).

In order to reduce the volume requirements for LLRW disposal, the Nuclear Regulatory Commission (NRC) has identified a need to develop a policy on regulating deminimus levels of radioactivity. Suggested alternatives to shallow land burial of low-activity wastes, under an exception to 10 CFR Part 20 included the regulated release of low-activity waste to uncontrolled disposal at a municipal sanitary landfill (19, 20).

The efficacy of codisposal of LLRW and municipal refuse hinges on the successful containment of LLRW within the landfill, during both the landfill stabilization phase and the post-closure phase. In view of total containment of LLRW, the design of landfills that receive municipal refuse and LLRW should incorporate provisions for leachate containment and recycle, since leachate recycle would contain LLRW within the leachate and the landfill environment until precipitants (e.g., sulfide, carbonates) are formed during landfill stabilization; these precipitants would precipitate LLRW within the landfills (21, 22, 23, 59, 64, 109). The precipitation of LLRW by potential-precipitant species, viz., sulfides, carbonates, and hydroxides, is counteracted by potential complexation of LLRW by humic substances present in leachate; this may lead to mobilization of precipitated LLRW. Thus. termination of leachate recycle during the final stages of landfill stabilization, namely, final maturation phase, would reduce the

potential mobilization of precipitated LLRW.

Codisposal of LLRW and municipal refuse within laboratory-scale lysimeters was investigated by Chang (21). Co-58, which was one of the radionuclides utilized to simulate LLRW, exhibited an increase in activity from 0.045 nCi/L to 0.1 nCi/L in leachate during methane fermentation phase of landfill stabilization. Chang (21) speculated that the increase in Co-58 in leachate was probably due to formation of soluble complexes of cobalt with humic substances present in leachate.

#### 1.2 Objectives of the Research

Codisposal of selected radionuclides and municipal refuse within two pilot-scale landfills was investigated and the major objectives of the investigation were:

- i) to evaluate the behavior of Co-58, Sr-85, and Ce-141
   codisposed with municipal refuse within the landfills
   during landfill stabilization; and,
- ii) to assess the potential of solubilization of Co-58 from precipitated cobalt sulfide by humic substances present in landfill leachates.

Since the containment of LLRW was a major determinant in the probable success of codisposal of LLRW with municipal refuse, both pilot-scale landfills were lined with 0.76-mm polyethylene liners (HDA Gundline, Gundle Lining Systems, Inc., Houston, TX) and designed for leachate collection and leak detection. One of the landfills was equipped with provisions for leachate recycle, since leachate recycle would accelerate landfill stabilization (22) and contain LLRW within the

leachate and the landfill environment until precipitants (e.g., sulfide, carbonates) are formed during landfill stabilization (23, 59).

# 1.3 Scope of the Research

An assessment of the technical feasibility of codisposal of selected radionuclides with municipal refuse within landfills would aid in evaluating this disposal technique as an alternative to shallow land burial for disposal of very low-activity wastes. The selected radionuclides, Co-58, Sr-85, and Ce-141, were codisposed with municipal refuse within two pilot-scale landfills and the extent to which these radionuclides along with the organics leached from the landfills was recorded. In addition, the potential for migration of these radionuclides through remobilization due to complexation with ligands, viz., humic substances, present in leachate was examined.

#### CHAPTER II

#### LITERATURE REVIEW

#### 2.1 Low-Level Radioactive Waste (LLRW)

Low-Level Radioactive Waste (LLRW) is defined as waste that contains less than 10 nCi (nanocurie) of long-lived  $\alpha$ -radiation per gram and has Y-radiation low enough to require minimal biological shielding and remote handling, or that average less than 35.7 curies of activity per m<sup>3</sup> (one curie of activity per cubic foot) of material or less than 200 mrem (millirem) per hour (26, 27). LLRW contains disposable filter cartridges, filter sludges, ion exchange resins, evaporator concentrates, contaminated clothing, and discarded tools (28).

# 2.1.1 Sources and Quantities of LLRW

Low-Level Radioactive Wastes are produced from fuel cycle, institutional research and development activities, medical and biological applications, and defense programs (27, 29). The fuel cycle accounted for 43% of LLRW produced annually from non-defense sources; the other sources (institutional, medical, and biological) for 47% to 49% of LLRW generated annually from non-defense sources (17, 29).

Commercial nuclear power plants are the largest generators of liquid LLRW from the nuclear fuel cycle. These wastes are classified as either boiling water reactor (BWR) wastes or pressurized water reactor (PWR) wastes. The important difference between these wastes is the absence of borates in the BWR wastes. The BWR and PWR wastes are collected and segregated into one of the eight categories, namely, high purity waste, medium purity waste, chemical waste, detergent waste, spent resin, filter precoat, secondary system wastes, and miscellaneous waste. The sources for generation of these types of wastes are summarized in Table 1.

Recent estimates have indicated that total volume of LLRW generated from nuclear fuel cycle was 2.7 x  $10^5 \text{ m}^3$  (30, 31); of this volume, 80% was produced at light water reactor (LWR) power plants and 20% resulted from fabrication of fuel. While the volume of LLRW produced varied from plant to plant, 4.5 x  $10^{-5} \text{ m}^3$  of LLRW were produced per megawatt of power generated at LWR plants (32).

The total volume of LLRW generated from other sources was 1.8 x  $10^5 \text{ m}^3$ . Anderson <u>et al</u>. (30) reported in 1978 that medical and biological sources constituted 80% of the LLRW (2.2 x  $10^4 \text{ m}^3$ ) produced annually. Industry and medical facilities produced 47% of the total LLRW generated annually in Pennsylvania (17). The total LLRW produced from fuel cycle and other sources is referred to as commercial LLRW.

#### 2.1.2 Characteristics of LLRW

The LLRW generated from the fuel cycle are classified into two categories, namely, wet wastes and dry wastes. The physical and chemical characteristics and the radiological properties of wet wastes generated from BWR, PWR, and Decontamination/Decommissioning Operations are presented in Tables 2 to 7. Since the precise composition of most of these wastes varied from plant to plant, the values listed in Tables 2 to 7 are typical values.

Waste Category	Source
High purity waste	Equipment drainage (BWR)
Medium purity waste	Miscellaneous leakage and floor drainage (BWR)
Chemical waste	Ion exchanger regenerations, solutions and laboratory drains (BWR, PWR)
Detergent waste	Drainage from laundry machines and personnel decontamination stations (BWR, PWR)
Spent resin	Water slurry of depleted bead resin from ion exchangers (BWR, PWR)
Filter precoat	Water slurry of powdered resin, diatomaceous earth, etc., from precoat- type filters (BWR, PWR)
Secondary system wastes	Turbine building floor and equipment drainage, steam generator blowdown, condensate polisher regeneration solutions, etc. These wastes normally contain little or no radioactivity (PWR).
Miscellaneous waste	Equipment and floor drainage and miscellaneous leakage

Table 1. Sources for Generation of Various Categories of Boiling Water Reactor (BWR) and Pressurized Water Reactor (PWR) Wastes (29)

Characteristic		Waste Category							
		High Purity Waste	Medium Purity Waste	Chemical Waste <sup>a</sup>	Detergent Waste	Bead Waste	Powdered Resin Filter Precoat	Diatomaceous Earth Filter Precoat	
Temperature	(°C)	21 to 65	21	77	21 to 65	21	21	21	
pH		6 to 8	6 to 8	6 to 8	7 to 9	-	-	-	
Conductivity	(µmho/cm)	<10	10 to 1,000	>10,000	-	-	-	-	
Density	(kg/m <sup>3</sup> )	1,000	1,000	1,180	1,000	-	-	-	
Activity <sup>b</sup>	(µCi/mL)	(0.88)(A+M)	(0.001)(A+M)	(40)(A+M)	See Table 4	1 to 10	0.1 to 500	0.05	
Constituents:									
Water <sup>C</sup>	(wt %)	>99	>99	75	99	50	50	40	
011	(ppm)	trace amts.	<1	-	-	-	-	-	
Detergent Suspended	(wt \$)	-	-	-	1	-	-	-	
Solids Sodium	(ppm)	<10	10 - 100	1,000	1,000	-	5	10	
Sulfate Sodium	(wt %)	-	-	22	-	-	-	-	
Chloride	(wt %)	-	-	2	-	-	5	-	
Bead Resin <sup>d</sup> Powdered	(wt %)	-	-	-	-	50	-	-	
Resin <sup>e</sup> Diatomaceous	(wt \$)	-	-	-	-	-	40	-	
Earth	(wt %)	-	-	-	-	-	-	40	

Table 2. Typical Characteristics of Boiling Water Reactor (BWR) Wet Wastes (29)

<sup>a</sup>Characteristics of chemical waste are after concentration by evaporation to solubility-limit of solution.

<sup>b</sup>For values of A and M, see Table 3.

<sup>C</sup>For bead resin and filter precoat type waste, water content listed is interstitial water only. <sup>d</sup>Bead resin is a variable mix of anion and cation resin beads.

ePowdered resin is a variable mix of powdered anion and cation resin beads.

œ

Nuclide	Reactor Coolant Fission Products A (µCi/mL)	Nuclide	Reactor Coolant Activation Products M (µCi/mL)
Sr-89 Sr-90 Zr-95 Nb-95 Mo-99 Ru-103 Ru-106 Ag-110m Te-129m Te-132 I-131 Cs-134 Cs-136 Cs-137 Ba-140 Ce-141 Ce-144 Pr-143 Np-239	2.7 x $10^{-3}$ 2.0 x $10^{-4}$ 3.5 x $10^{-5}$ 3.6 x $10^{-5}$ 2.0 x $10^{-2}$ 1.7 x $10^{-5}$ 2.2 x $10^{-6}$ 6.0 x $10^{-5}$ 3.4 x $10^{-5}$ 4.3 x $10^{-2}$ 1.2 x $10^{-2}$ 1.4 x $10^{-4}$ 9.2 x $10^{-5}$ 2.1 x $10^{-4}$ 7.8 x $10^{-3}$ 3.4 x $10^{-5}$ 3.0 x $10^{-5}$ 3.3 x $10^{-5}$ 2.1 x $10^{-1}$	P-32 Cr-51 Mn-54 Co-58 Co-60 Fe-59 Zn-65	$2.0 \times 10^{-5}$ 5.0 × 10 <sup>-4</sup> 4.0 × 10 <sup>-5</sup> 5.0 × 10 <sup>-3</sup> 5.0 × 10 <sup>-4</sup> 8.0 × 10 <sup>-5</sup> 2.0 × 10 <sup>-6</sup>

Table 3. Typical Concentrations of Expected Radionuclides in Reactor Coolant for Boiling Water Reactors (BWRs) (29)

#### Notes:

- Only isotopes with half lives > 24 hr are listed.
   Activity in main steam is 10<sup>-3</sup> times activity in reactor coolant, except that, for halogen, activity in main steam is 10<sup>-2</sup> times activity in reactor coolant.

Nu	uclide	Activity (μCi/mL)
Mr	n-58	$1.6 \times 10^{-6}$
Cc	5-58	6.4 x 10 <sup>-6</sup>
Cc	<b>b</b> -60	1.4 x 10 <sup>-5</sup>
Zr	-95	$2.3 \times 10^{-6}$
Ru	-103	$2.3 \times 10^{-7}$
Ru	106	$3.9 \times 10^{-6}$
Ag	g-110m	7.1 x 10 <sup>-7</sup>
I-	-131	$9.7 \times 10^{-7}$
Ca	3-134	2.1 x 10 <sup>-5</sup>
Cs	3-137	$3.9 \times 10^{-5}$
Ce	e-144	$8.0 \times 10^{-6}$

Table 4. Typical Concentrations of Expected Radionuclides in Laundry Wastes from Boiling Water Reactor (BWR) and Pressurized Water Reactor (PWR) (29)

Note: Only isotopes with half-lives > 24 hr are listed.

Characteristic		Wast	Waste Category				
		Miscellaneous Chemical Waste <sup>a</sup>	Secondary Side Condensate Polisher Regenerative Waste <sup>a</sup>				
Temperature	(°C)	77	77				
рН		2.5 to 4.0	2.5 to 4.0				
Boric Acid	(wt %)	12	-				
Activity <sup>b</sup>	(µCi/mL)	(0.2)(R)	(4x10 <sup>4</sup> )(S)				
Sodium Sulfate	(wt %)	-	14.9				
Ammonium Sulfate	(wt %)	-	9.6				
Sodium Chloride	(wt %)	-	2.0				

### Table 5. Typical Characteristics of Pressurized Water Reactor (PWR) Wet Wastes (29)

<sup>a</sup>Characteristics of miscellaneous chemical waste and secondary side condensate polisher regenerative waste are after concentration by evaporation to solubility limit of solution.

bSee Table 6 for R and S.

#### Note:

Characteristics of Detergent Wastes, Bead Resin, and Filter Precoat Backwash are the same as those of BWR (See Table 2).

Nuclide	Reactor Coolant Fission and Activation R (µCi/mL)	Secondary Side Fission and Activation S (µCi/mL)
Cr-51 Mn-54 Fe-55 Co-58 Fe-59 Co-60 Sr-89 Sr-90 Y-91 I-131 Rb-86 Cs-134 Cs-136 Cs-137	1.7 x $10^{-3}$ 2.7 x $10^{-4}$ 1.4 x $10^{-3}$ 1.4 x $10^{-2}$ 8.8 x $10^{-4}$ 1.8 x $10^{-3}$ 3.1 x $10^{-4}$ 8.8 x $10^{-6}$ 5.6 x $10^{-5}$ 2.4 x $10^{-1}$ 7.7 x $10^{-5}$ 2.2 x $10^{-2}$ 1.6 x $10^{-2}$	2.2 x $10^{-7}$ 5.8 x $10^{-8}$ 1.8 x $10^{-7}$ 1.9 x $10^{-6}$ 1.4 x $10^{-7}$ 2.4 x $10^{-7}$ 5.4 x $10^{-8}$ 1.3 x $10^{-9}$ 8.1 x $10^{-9}$ 3.6 x $10^{-5}$ 1.2 x $10^{-8}$ 3.5 x $10^{-6}$ 1.8 x $10^{-6}$ 2.4 x $10^{-6}$

Table 6.	Typical Concentrations of Expected Radionuclides
	in Reactor Coolant and Secondary Side for
	Pressurized Water Reactors (PWRs) (29)

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Note: Only isotopes with half-lives > 24 hr are listed.

Characteristic 		Component Decontamination Waste <sup>a</sup>	System Decontamination Waste <sup>b</sup>	Building Decontamination Waste <sup>C</sup> 21	
		21	21		
pH		4	4	5	
Conductivity	(µmho/cm)	) >10 <sup>4</sup>	>104	-	
Dow NS-1d	(wt %)	7	7	-	
Water	(wt %)	93	93	88	
Iron Oxide	(ppm)	600 tO 900	1200	-	
Co-60	(ppm)	1.4 to 1.6	-	-	
Radiac Wash <sup>e</sup>	(wt %)	-	-	5	
Crud	(wt %)	-	-	5	
Oil	(wt %)	-	-	2	
Activity:					
I-131	$(\mu Ci/mL)$	-	-	0.6	
Ce-134	$(\mu Ci/mL)$	-	-	0.4	
Ce-136	$(\mu Ci/mL)$	-	-	0.2	
Ce-137	$(\mu Ci/mL)$	-	-	1.4	
La-140	(µCi/mL)	-	-	5.2	
Ba-140	$(\mu Ci/mL)$	-	-	3.6	
Total Activit	y (µCi/mL)	1.2 to 2.2 <sup>f</sup>	0.48	-	

# Table 7. Characteristics of Typical Decontamination Waste Solutions (29)

<sup>a</sup>Data for component decontamination is based on decontamination of regeneration heat exchangers at Peach Bottom Units 2 and 3.

<sup>b</sup>Data for system decontamination is based on test loop set-up for decontamination of Dresden Unit 1 reactor collant system.

<sup>C</sup>Data for building decontamination is based on decontamination of auxiliary building at Three Mile Island Unit 2.

dDow NS-1 is a proprietary mixture of EDTA, inorganic salts, and organic inhibitors.

<sup>e</sup>Radiac Wash is a mixture of EDTA, mild detergent and other proprietory additives.

fTotal activity for component decontamination is based on 50/50
volume % mixture of decontamination solution and rinse water.
gTotal activity for component decontamination is based on 30/70
volume % mixture of decontamination solution and rinse water.

# 2.1.3 Disposal of LLRW

Disposal of LLRW, by tradition, has been accomplished by shallow land burial. While other disposal methods including intermediate depth burial, disposal in engineered facilities, disposal in drilled holes, deep-well injection, disposal in hydrofractured strata, disposal in cavities, and sealed disposal have been both proposed and reviewed, shallow land burial has been the most utilized method for disposal of LLRW (33).

The total volume of commercial LLRW buried within the United States has increased rapidly since 1962; currently, more than 5.66 x  $10^4$  m<sup>3</sup> of commercial LLRW are buried annually at licensed commercial sites (34). Defense operations have generated three times more LLRW than commercial operations. The volume of LLRW generated annually due to defense operations has leveled at 2.8 x  $10^3$  m<sup>3</sup>. The LLRW generated from defense operations are buried at 17 government-owned sites which are managed by the U.S. Department of Energy (DOE).

The shallow land burial of LLRW generated from commercial operations has been accomplished at six shallow-land burial sites, namely, Beatty, Nevada; Maxey Flats, Kentucky; West Valley, New York; Richland, Washington; Sheffield, Illinois; and Barnwell, South Carolina. Of these sites, Maxey Flats and West Valley sites have been closed due to migration of radionuclides caused by water management problems. The site at Sheffield, Illinois experienced lengthy delay with license renewal. Thus, only three sites at Beatty, Nevada; Richland, Washington; and Barnwell, South Carolina have been receiving LLRW for disposal (18). In addition, the site at Barnwell, South Carolina has instituted a volume allotment program for receiving LLRW for disposal; this has caused a three-fold increase in transportation cost incurred by some Midwestern utilities due to their LLRW being shipped to Richland, Washington and Beatty, Nevada.

#### 2.1.4 Problems Associated with Shallow-Land Burial of LLRW

The major problems identified at licensed-commercial sites for shallow-land burial are the migration of radionuclides at some sites and the gradual depletion of available volume for LLRW disposal due to site closure and increased generation of LLRW (1, 5, 6-16, 18, 35-51).

The problems associated with shallow-land burial are further compounded by the restrictions that are imposed on LLRW disposal by the Low-Level Radioactive Waste Act of 1980 (PL 96-573) from January 1986. This Act requires each state to be responsible for assuring adequate disposal for LLRW generated within its borders. Also, this Act allows states that have agreed to form a regional compact, the authority to exclude LLRW from outside their region. Six regional compacts of states, namely, Northwest, Central, Midwest, Southeast, Northeast, and Rocky Mountain have been formed. The status of low-level regional compacts is presented in Figure 1. These compacts have won legislative approval from their represented states. However, Congress has not approved any of the compacts (18).

Although the Nuclear Regulatory Commission (NRC) does not view storage as an alternative to disposal, one of the options for compacts without an operating site would be to provide interim storage capacity for their LLRW while developing disposal capacity. NRC has estimated that in the final environmental impact statement (FEIS) for 10 CFR 61

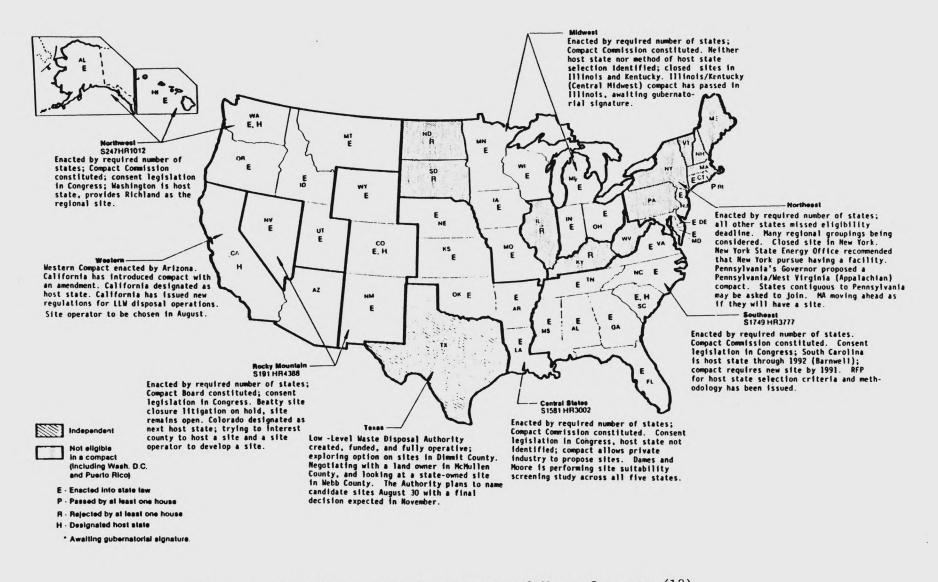


Figure 1. The Status of Regional Low-Level Waste Compacts (18).

licensing rule for land disposal of LLRW, site selection and characterization would take up to two years, and the licensing process would take one to two years. Thus, depending on the characteristics of the site and the complexity of the NRC review, the process could take two to four years which would obviously be extended in the event of a protracted hearing (18). Consequently, the interim storage of LLRW within regional compacts without an operating site may be inevitable; this would lead to an increase in the volume of LLRW to be disposed at a later date.

2.1.4.1 <u>Suggested Remedial Actions for Reduction in Volume of</u> <u>LLRW</u>. In order to reduce the volume requirement for LLRW disposal, the NRC has identified a need to develop a policy on regulating deminimus levels of radioactivity. Among the suggested alternatives to shallow land burial, under an exclusion in 10 CFR 20, was the regulated release of low-activity (deminimus levels) waste to uncontrolled disposal at a municipal sanitary landfill. The disposal of dry active waste (DAW) below deminimus levels within a landfill is one of the strategies being considered by utilities in regional compacts without an operating site (19, 20). Thus, codisposal of deminimus levels of LLRW and municipal refuse within landfills is being regarded as a viable strategy for reduction in the volume requirement for LLRW disposal.

#### 2.2.1 Controlled Landfill

Sanitary landfills have become the most common method of municipal refuse disposal in the United States, not only because of tradition, but also because of their economic advantage over other methods of solid waste management. Based on the type of solid waste it

receives, a landfill can be identified as one of the three types of landfills: "Sanitary Landfill" intended for municipal refuse disposal, "Secure Landfill" intended for hazardous waste disposal, and "Controlled Landfill" intended for codisposal of municipal refuse and hazardous waste. A leachate and gas collection/treatment strategy forms the basis of a "Controlled Landfill" since the potential migration of either leachate or gas from the landfill would result in environmental impairment (52).

#### 2.2.2 Municipal Refuse

The sources of municipal refuse are largely dependent on location and may be classified as residential, commercial, industrial, and agricultural. The type of municipal refuse produced from these sources usually include food wastes, rubbish, ash and residues, demolition and construction debris, treatment plant sludges, agricultural wastes, and hazardous wastes. Wastes originating from residential and commercial activities constitute municipal refuse. The composition of municipal refuse is presented in Table 8; of the various constituents, food and garden wastes represent the readily biodegradable fraction. The projected municipal refuse production in 1990, at 3.5 percent annual compound growth, will be 230 million metric tons (53).

#### 2.2.3 Landfill Characterization

A "controlled landfill" is usually adjoined by a natural environmental setting and the potential impairment of this setting is inextricably linked with the potential migration of the transport phases, namely, leachate and gas, from the landfill. A proper

Component	Tchobanoglous, <u>et al</u> ., (1977)	Myer, <u>et al</u> ., (1979)	Chain, <u>et al</u> ., (1977)	U.S. EPA (1968)	Walsh and Kinman	Solid Waste Management	
	(53)*	(54)*	(55)*	(56)*	(1981) (57)*	(1972) (58)*	
	_						
Food Wastes	5.76	0.94	14.4	11.1	3.6	6.05	
Garden Wastes	6:14	0.41	3.1	6.9	10.7	6.76	
Paper	48.14	44.79	36.5	48.6	42.6	47.5	
Cardboard	4.86	_	_	-	-	-	
Plastics	3.71	-	2.8	-	-	0.86	
Rubber	0.64	9.03	-	-	8.7	0.75	
Leather	0.51	-	-	-	-	0.34	
Textiles	2.30	3.08	0.7	0.7	-	0.68	
Wood	2.04	0.49	1.8	2.1	2.6	2.42	
Glass	9.98	7.61	6.8	8.3	12.2	7.41	
Tin Cans	7.42	-	-	_	_	-	
Non-Ferrous Metal		10.82	2.7	11.1	12.0	9.78	
Ferrous Metals	2.43	_	11.8	11.1	-	-	
Dirt, Ashes, and							
Brick	4.82	-	14.9	8.3	3.2	16.20	

Table 8. Co	omposition of	Municipal	Refuse	Produced	in	the	United	States	(Percent	Dry	Weight)
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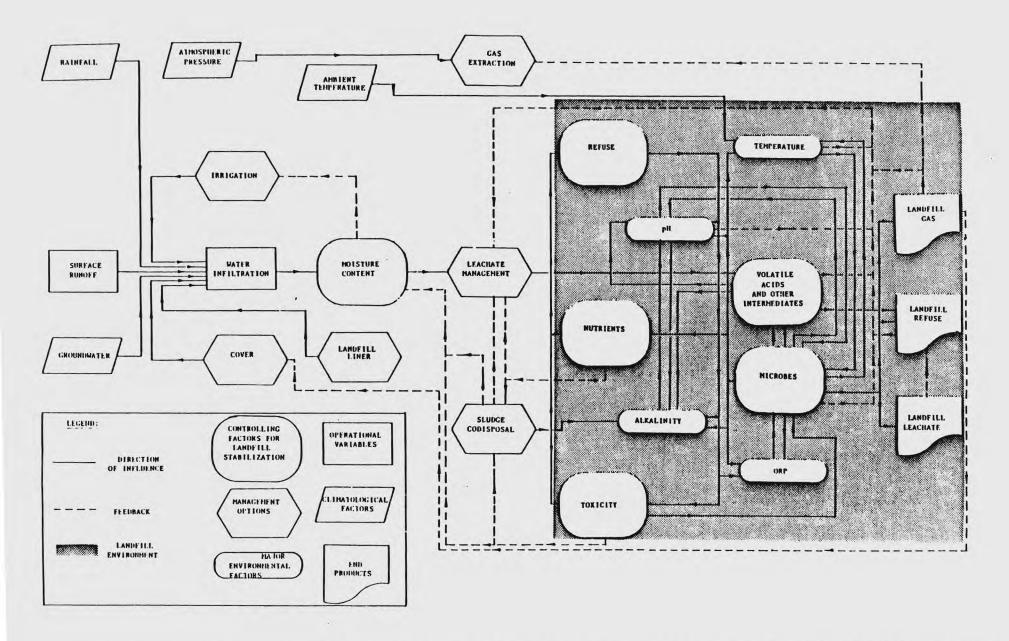
\*Indicates reference number

understanding of the processes contributing to landfill stabilization is important, especially when the landfill is not "secured", since the variations in leachate and gas during landfill stabilization indicate the setting's vulnerability to change. A better understanding of landfill behavior and development of more efficient design, operation, and control procedures for "controlled landfills" have resulted from investigations on codisposal of municipal refuse and hazardous wastes within "controlled landfills" (23, 54, 59, 64).

All candidate landfill sites should be evaluated initially in terms of climatological events and their impacts on local hydrogeology. The factors affecting landfill stabilization are presented in Figures 2 and 3. As shown in Figure 2, the rainfall-derived infiltration constitutes the principal source of moisture for generation of leachate during the life of a landfill; thus, a reliable determination of areal rainfall and its opportunity to enter, persist within, and exit the landfill environment are of prime importance. Moreover, rainfallderived infiltration will be affected by the landfill management strategies employed at the site, i.e., cover, lining, codisposal, irrigation/leachate recycle, and gas extraction procedures (60). The interrelationships between differenet parameters, within a landfill environment are shown in Figure 3.

#### 2.2.4 Landfill Stabilization

Landfills receiving municipal refuse along with varying quantities of hazardous wastes proceed through a series of stabilization phases; the importance and the longevity of each phase are determined by the existing climatological conditions, operational



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Figure 2. Factors and Management Techniques Influencing Leachate and Gas Production During Landfill Stabilization (60).

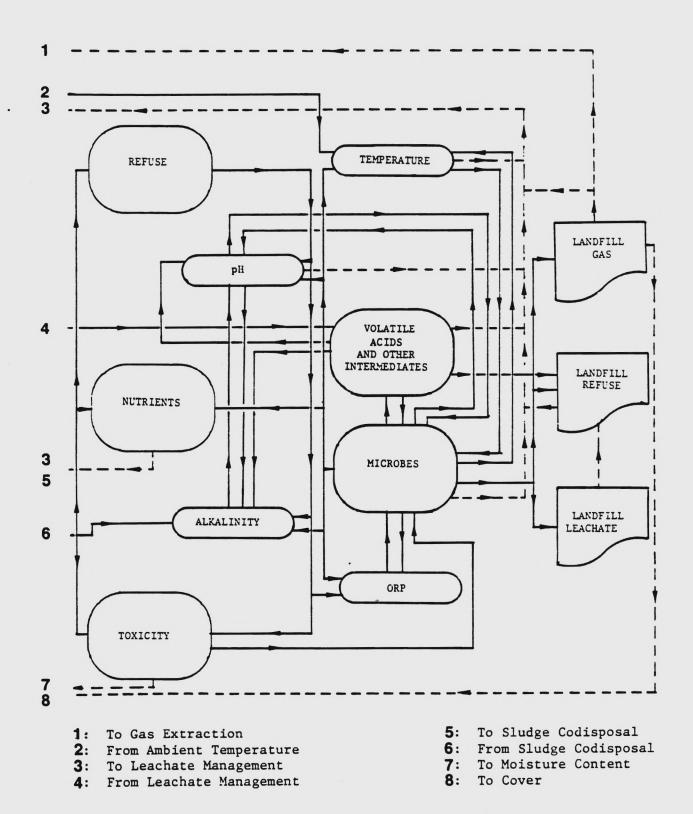


Figure 3. Interrelationships Between Different Parameters Within a Landfill Environment (60).

variables, management options and control factors operative or being applied externally or internally (52, 60). The concept of sequential behavior can be ascertained from results of studies on landfill stabilization (21, 23, 52, 59, 60). The choice of analysis of certain environmental parameters is governed by the fact that a landfill during its active life is analogous to a batch anaerobic digestor with limited inputs or outputs and has a functional retention time in years instead of days. Thus, the existence of discrete and sequential phases of refuse conversion and stabilization within a landfill receiving municipal refuse can be illustrated in terms of principal events and parametric changes by the following five stabilization phases: Initial Adjustment Phase, Transition Phase, Acid Formation Phase, Methane Fermentation Phase, and Final Maturation Phase (52, 60).

Phase I: Initial Adjustment--

- Initial placement of waste and preliminary accumulation of moisture occur.
- Initial subsidence and closure of each landfill area.
- Changes in environmental parameters are first detected to reflect the onset of stabilization processes, these processes trend in a logical fashion.

Phase II: Transition--

- "Field capacity" is exceeded and leachate is formed.
- A transition from initial aerobic to anaerobic microbial stabilization occurs.
- The primary electron acceptor shifts from oxygen to nitrates and sulfates with the displacement of oxygen by

carbon dioxide in the gas.

- A trend toward reducing conditions is established.
- Measurable intermediates such as the volatile fatty acids first appear in the leachate.

Phase III: Acid Formation--

- Intermediary volatile fatty acids become predominant with the continuing hydrolysis and fermentation of waste and leachate constituents.
- A precipitious decrease in pH occurs with a concomitant mobilization and possible complexation of metal species.
- Nutrients such as nitrogen and phosphorus are released and utilized in support of the growth of biomass comensurate with the prevailing substrate conversion rates.
- Hydrogen may be detected and affect the nature and type of intermediary products being formed.

Phase IV: Methane Formation--

- Intermediary products appearing during the acid formation phases are converted to methane and excess carbon dioxide.
- The pH returns from a buffer level controlled by the volatile fatty acids to one characteristic of the bicarbonate buffering system.
- Oxidation-reduction potentials are at their lowest values.
- Nutrients continue to be consumed.
- Complexation and precipitation of metal species proceed.
- Leachate organic strength is dramatically decreased in

correspondence with increases in gas production.

Phase V: Final Maturation--

- Relative dormancy following active biological stabilization of the readily available organic constituents in the waste and leachate.
- Nutrients may be limiting.
- Measurable gas production all but ceases.
- Natural environmental conditions become reinstated.
- Oxygen and oxidized species may slowly reappear with a corresponding increase in oxidation-reduction potential.
- More microbially resistant organic materials may slowly be converted with the possible production of humic-like substances capable of complexing with and re-mobilizing heavy metals.

All of the major events selected to describe and separate these landfill stabilization phases are encountered at one time or another in landfills containing municipal refuse, provided that the associated microbially mediated processes have been augmented by a sufficiency of moisture and nutrients, and are not being exposed to the inhibited influences of toxic materials (52, 60).

Since the landfill stabilization phases usually overlap during the active life of a landfill, they are often viewed in a collective fashion that trends to obscure their discrete and sequential existence; this limits the recognition and the understanding of the progress of events that result in maturation of stabilization processes. The time required for the maturation of stabilization processes may be described in terms of landfill "age". In fact, a landfill, during its active life, does not have a single "age" but rather a collection of landfill "ages" with different "ages" being associated with microenvironments within the landfill complex due to their respective progress towards stabilization (60). Moreover, the rate of progress towards stabilization depends upon the physical, chemical, and microbial conditions within the microenvironments. For example, high compaction may restrict the movement of moisture and nutrients throughout the mass of municipal refuse, acidic conditions established during acid formation phase may prevent the onset of active methane fermentation, and the presence of toxic substances may induce microbial inhibition within microenvironments of the landfill complex (52, 60).

# 2.2.5 Indicator Parameters Descriptive of Landfill Stabilization Phases

There are certain indicator parameters or indices capable of being used to detect and describe the presence, the intensity, and the longevity of each phase of landfill stabilization (60). Many of these parameters apply to the analysis of leachate; thus, they are important after leachate production has commenced. In addition, their applications and interrelationships within an overall landfill perspective are reflected by the nature of analyses, namely, physical, chemical, or biological. For example, pH and ORP are physical parameters that indicate acid-base and oxidation-reduction conditions, respectively. COD and BOD<sub>5</sub> are chemical and biological parameters, respectively, but are indicative of relative biodegradibility of the leachate produced from the landfill. Other critical parameters, that are used to identify and assess the intensity and the longevity of each solubilization

phase, include alkalinity (buffer capacity), conductivity (ionic strength/activity of ions), heavy metals (potential inhibition of microbial-mediated processes), individual and total volatile acids (breakdown of polymers to monomers, availability of substrate to methane formers), and chloride (dilution, mixing, and washout of organic and inorganic constituents present in leachate).

Ranges in intensity or concentration of indicator parameters will vary throughout the stabilization phases due to the principal function of each stabilization phase and dilution resulting from continuous infiltration of moisture into the landfill. Table 9 provides a compilation of data on the general ranges of intensity and concentration of indicator parameters throughout the landfill stabilization phases (25). An examination of these data indicates some overlap in the intensity or concentration of indicator parameters, particularly during the acid formation and the methane fermentation phases.

The processes contributing to landfill stabilization are most pronounced during the acid formation and the methane fermentation phases. The combined duration of these phases is reduced with leachate recycle due to accelerated stabilization (21-23, 59, 64, 109). During the contracted time interval, however, the intensity and the concentration of indicator parameters are magnified. Thus, the time span during which environmental impairment of the natural setting encompassing the landfill can occur is reduced with leachate recycle.

#### 2.2.6 Metal Solubility in Landfill Leachates

The solubility of heavy metals in landfill leachates is an important determinant in the performance of an anaerobic biological

	PIIASES OF STABILIZATION				
PARAMETER	TRANSITION	ACID FORMATION	METHANE FERMENTATION	FINAL MATURATION	REPORTED RANGE
Biochemical Oxygen Demand (BOD <sub>5</sub> ), mg/l	Low to high (100-10,900); Low to high biodegrad- ability	Moderate to high (1000-57,700); Moderate to high blodegrad- ability	High to low (3400-600); High to low biodegrad- ability	Low (4-120); Low blodegradability	4-57,700
Chemical Oxygen Demand (COD), mg/1	Low to high (480-18,600); Low to high oxidation potential	Moderate to high (1500-71,700); Noderate to high oxidation potential	High to low (9760-580); High to low oxidation potential	Low (31-900); Low oxidation potential	31-71,700
Total Organic Carbon (TOC), mg/l	Low to moderate (100- 3000); Low to moderate organic carbon content	Low to high (500-27,700); Low to high organic carbon conlent	Moderate to low (2230- 300); Moderate to low organic carbon content	Low (70-260); Low organic carbon content	70-27,700
Total Volatile Acids (TVA), mg/l Acetic	Absent to low (0-500)	Low to high (3000-18,800); Volatile acld formation	lligh to low (4000-250); Volatile acid conversion to methane	Absent to low (0-50)	0-18,800
BOD <sub>5</sub> /COD	Low to high (0.23-0.87); Low to high substrate biodegradability	Moderate to high (0.4-0.8); Hoderate to high substrate biodegradability	Moderate to low (0.64- 0.17); Moderate to low substrate biodegrad- ability	Low (0.02-0.13); Low substrate biodegradability	0.02-0.87
COD/TOC	High (4.3-4.8); Low oxidation state of organics	Moderate to high (2.1-3.4); Hoderate to low oxidation state of organics	Moderate to low (3.0- 2.0); Hoderate to high oxidation state of of organics	Low (0.4-2.0); Iligh oxidation state of organics	0.44-4.8
Total Kjeldahl Nitrogen (TKN), mg/l	Moderate to high (180-860)	Low to high (14-1970); Leach- ing and assimilation of nitrogenous compounds	Low (25-82); Assimila- tion of nitrogenous compounds	Low to moderate (7-490)	7-1970
Ammonia Nitrogen (NH <sub>3</sub> -N), mg/1	Moderate (120-225)	Low to high (2–1030); Pro- duction and biological up- take	lligh to low (430–6); Blological uptake	High to low (430-6); Biolog- ical uptake and release	2-1030

Table 9. Leachate Concentrations and General Significance During Landfill Stabilization (25).

Table 9. Continued...

	PHASES OF STABILIZATION				
PARAMETER	TRANSITION	ACID FORMATION	METHANE FERMENTATION	FINAL MATURATION	REPORTED RANGE
Nitrate Nitfogen (NO3 -N), mg/l	Low to high (0.1-51); Oxidation of annuonia	Moderate to low (19-<0.05); Nitrate reduction	Absent	Low (0.5-0.6)	<0.05-51
NH <sub>3</sub> -N/TKN	Low to high (0.1-0.91)	Low to high (0.02-0.98); Production and utilization	Low to high (0.1-0.84); Production and utiliza- tion	Moderate to high (U.54-0.97); Bio- logical uptake and release	0.02-0.98
Total Phosphate (PO <sub>4</sub> <sup>Ξ</sup> -P), mg/l	Low (0.6-1.7)	High to low (120–0.16); Bio- logical assimilation and complexation	Moderate to low (14- 0.7); Biological assimilation and com- plexation	Low to moderate (0.16-14)	0.16-120
Total Coliform, <u>CFU</u> 100m1	Moderate to low $(10^5 - 10^0)$	Moderate to low (10 <sup>5</sup> -10 <sup>0</sup> )	Absent	Absent	10 <sup>0</sup> -10 <sup>5</sup>
Fecal Coliform, CFU	Moderate to low (10 <sup>5</sup> -10 <sup>0</sup> )	Moderate to low (10 <sup>5</sup> -10 <sup>0</sup> )	Absent	Absent	10 <sup>0</sup> -10 <sup>5</sup>
Fecal Streptococci, <u>CFU</u> 100ml	Moderate to low (10 <sup>6</sup> -10 <sup>0</sup> )	Moderate to low (10 <sup>6</sup> -10 <sup>0</sup> )	Absent	Absent	10 <sup>0</sup> -10 <sup>6</sup>
Viruses, PFU/100ml	-	Absent	Absent	Absent	-
pH	Neutral (6-7)	Neutral to low (7.7-4.7); Volatile acid formation and inhibition	Heutral to high (6.3- 8.8); Volatile acid conversion to methane	Neutral to high (7.1-8.8); Absence of volatile acids	4.7-8.8
Oxidation-Reduction Potential (ORP), mV E <sub>c</sub>	High (+40); Incipient acrobic conditions	High to low (+80 to -240); Facultative and anaerobic conditions	Low (-73 to -240); Anaerobic methane fermentation	High (+97 to +163); High oxidation state of organics	-240 to +163

Table 9. Continued...

	PHASES OF STABILIZATION				
PARAMETER	TRANSITION	ACID FORMATION	METHANE FERMENTATION	FINAL MATURATION	REPORTED RANGE
Conductivity, µmhos/cm	Low (2450-3310); Anion and cation leaching	Low to high (1600–17,100); Volatile acid formation, anion and cation leaching and precipitation	Moderate to low (7700- 2900); Conversion of volatile acids to methane, metal precipitation	Low (1400-4500)	1400- 17,100
Solids (TS), mg/l	Low (2450-2960); Anion and cation leaching	Low to high (4120-55,300); Volatile acid formation, anion and cation leaching	Moderate (6410-2090); Con- version of volatile acids to methane, metal pre- cipitation and filtration	Low (1460-4640)	1460- 55,300
Alkalinity, mg/1 as CaCO <sub>3</sub>	Low (200-2050)	High to low (9650-140); Volatile acid formation and neutralization	Low to moderate (760- 5050); Volatile acid con- version to methane	Low to moderate (200-3520)	140-9650
Chloride (Cl <sup>-</sup> ), mg/l	Low to high (30-5000); Leaching and washout	Low to high (30-5000); Leach- ing and washout	Low to high (30-5000); Leaching and washout	low to high (30-5000)	30-5000
Sulfate (SO <sub>4</sub> <sup>≠</sup> ), mg/l	Low to moderate (10- 458); Leaching and oxidation of sulfur compounds	High to low (3240–10); re- duction of sulfate	Absent	Low (5-40)	5-3240
Sulfide (S <sup>®</sup> ), mg/l	Absent	High to low (818–1.6); Re- duction of sulfate and metal precipitation	Low (0.9); Metal pre- clpitation	Absent to low (0-0.9)	0.9-818
Cadmium (Cd), mg/1	Low (<0.005-0.01)	Low to moderate (<0.005-0.39); Inhibition and precipitation	Low (<0.005-0.1); Pre- cipitation	Low (0.004)	<0.005- 0.39
Calcium (Ca), mg/l	Low (190-490)	Low to high (70-3900); Leach- ing and precipitation	Moderate to low (490- 76); Precipitation	Low (76-254)	70 - 3900

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Table 9. Continued...

		PHASES OF STABILIZATION			
PARAHETER	TRANSITION	ACID FORMATION	METHANE FERMENTATION	FINAL MATURATION	REPORTED RANGE
Chromium (Cr), mg/l	Low (0.023-0.28)	High to low (18-0.06); Inhibi- tion and precipitation	Low (0.05); Precipita- tion	Low (0.05)	0.02-18
Copper (Cu), mg/l	Low (0.085-0.39)	High to low (2.2-0.005); Leach- ing and precipitation	Low (0.03-0.18); Pre- cipitation	Low (0.02-0.56)	0.005-2.2
lron (Fe), mg/l	Low to moderate (68-312)	High to low (2200-90), Leach- ing and precipitation	Moderate to low (336- 115); Precipitation	Low (4-20)	4-2200
Lead (Pb), mg/1	Low (0.001-0.004)	High to low (1.44-0.01); Inhibi- tion and precipitation	Low (0.01-0.1); Precipitation	Low (0.01-0.1)	0.001-1.44
Magnesium (Mg), mg/l	Low (66-96)	Low to high (3-1140); Leaching	Moderate to low (505-81)	Low (81-190)	3-1140
Manganese (Mn), mg/1	Low (0.6)	High to low (41–0.6); Leaching and precipitation	Low (0.6); Precipitation	Low (0.6)	0.6-41
Nickel (Ni), mg/l	Low to moderate (0.02- 1.55)	High to low (79-0.03); Inhibi- tion and precipitation	Low (0.1–1.0); Precipi- tation /	Low (0.07)	0.02-79
Potassium (K), mg/l	Low to high (35-2300)	Low to high (35-2300); Leach- ing and washout	Low to high (35-2300); Leaching and washout	Low to high (35-2300)	35-2300
Sodium (Na), mg/1	Low to high (20-7600)	Low to high (20-7600); Leach- ing and washout	Low to high (20-7600); Leaching and washout	Low to high (20-7600)	20-7600
Zinc (Zn), mg/1	Low to moderate (0.06-21)	High to Low (220–0.65); Inhibi- tion and precipitation	Low (0.4-6); Precipi- tation	Low (0.4)	0.06-220

system since the degree of toxicity of a metal to microorganisms, that contributes to microbial inhibition, is dependent on the amount of metal available to penetrate its cell wall (56). From the point of view of codisposal of LLRW and municipal refuse within controlled landfills, the solubility of LLRW in leachate is important since the presence of LLRW in the liquid transport phase (leachate) within a landfill increases the potential of LLRW migration in the event of a liner failure. The factors controlling metal solubility in leachates include the presence of potential precipitant species such as hydroxide, carbonate, and sulfide, the presence of complexing ligands (humic substances) that will tend to enhance metal solubility, and pH and ORP, due to their impact on the dissociation of species.

Sulfide, even at low concentrations, is a powerful precipitant and will precipitate most heavy metals, with the exception of chromium (59), as metal sulfides. Sulfide is produced under reducing conditions by microbially-mediated reductions of sulfates and other sulfur containing inorganic compounds (62). Sulfide in solution dissociates forming three species:  $H_2S$ ,  $HS^-$ , and  $S^{2-}$  and the distribution of these species is pH dependent. Equilibrium concentrations of soluble sulfide up to 200 mg/L as  $S^{2-}$  did not exert any significant toxic effects on anaerobic treatment (62, 63).

The distribution of carbonate species  $(H_2CO_3, HCO_3, and CO_3^{2-})$ in leachate is pH dependent. Carbonates and hydroxy carbonates have the potential to control the solubility of metals that are incapable of forming metal sulfides such as strontium (64). Also, carbonates and hydroxy carbonates may control the solubility of heavy metals in

absence of sulfide. Hydroxide acts as another precipitant for heavy metals in leachate. When hydroxide and sulfide are simultaneously present in leachate, the solubility is usually controlled by sulfide, except for trivalent chromium (59).

In complicated matrices such as leachate produced from a landfill, complexing agents both inorganic and organic in nature are present along with the potential precipitant species like sulfides, carbonates, and hydroxides. The occurrence of humic substances in landfill leachate has been reported by Chian and DeWalle (78), Raveh and Arnimelech (79) and Fuller and Artiole (80). Humic substances are complex polymers that possess both carboxyl and phenolic hydroxyl acidic functional groups. These functional groups are involved in metal complexation reactions (73).

The precipitation of heavy metals present in leachate by the potential-precipitant species, viz., sulfides, carbonates, and hydroxides, is counteracted by the formation of soluble metal-humate complexes; this leads to an increased solubilization of heavy metals in leachate. Pohland <u>et al</u>. (59, 64) reported the presence of aromatic hydroxyl as a mobilizing ligand for heavy metals in leachate from landfills receiving municipal refuse and varying quantities of hazardous wastes (metal sludges); the solubility of zinc and nickel was enhanced by aromatic hydroxyl even in the presence of sulfides, carbonates, and hydroxides. Heavy metals (copper, cadmium, nickel, zinc, manganese, lead, and iron) are known to form complexes with humic acid (67-77). Batch experiments on the extent to which heavy metal ions, namely, copper, lead, zinc, and cadmium are removed from aqueous solutions by humic acid revealed that

metal-humate complexes are usually formed within five days (67, 70); at equilibrium, humic acids enhanced the solubility of heavy metals even in presence of sulfide (67, 74).

#### 2.2.7 Metal Speciation by Chemical Modeling

If the medium under study is assumed to be in equilibrium, the concentration of each species of a given component (metal or ligand) can be calculated on the basis of thermodynamics, provided that the following are known: the total concentration of all the components; and the equilibrium constants of all the reactions that occur between the various species under the conditions (pressure, temperature, and ionic strength) of the medium being studied.

This approach has the considerable advantage of requiring only the determination of total concentrations of components. The model, however, suffers from limitations mentioned below:

> i) The equilibrium constants of all possible reactions should be known. Since little information is available on mixed complexes, their contribution to formation of complexes cannot be accurately assessed. Likewise, little is known about the adsorption of free ions, ligands and complexes on solids (65).

# Since, the approach is based on thermodynamic considerations, the medium is assumed to be in equilibrium. Kinetic aspects, however, are often important for reactions occurring with polymeric species, many of which are metastable compounds. Kinetics also play a major role in precipitation reactions and for processes involving mass

#### transfer at interfaces.

Even with these limitations, this approach can be used to describe the potential formation, at equilibrium, of metal-ligand precipitates and soluble complexes within the medium. Thus, the forms in which heavy metals may exist in the leachate and landfill environment during different phases of landfill stabilization, can be estimated.

A computer program for the calculation of chemical equilibrium composition of aqueous systems (MINEQL) developed by Westall <u>et al</u>. (66) was modified, then used to assess the potential formation of metal-ligand precipitates and soluble complexes for the set of metals and ligands that are summarized in Table 10.

The equilibrium composition of metal complexes in leachates (ionic strength = 0.03 M) at pH values of 5 and 7, are presented in Figures 4-7. As shown, at both pH values sulfide was the most powerful precipitant for heavy metals ( $Fe^{2+}$ ,  $Zn^{2+}$ ,  $Ni^{2+}$ , and  $Co^{2+}$ ) in the data set with the exception of trivalent chromium and manganese. The solubility of trivalent chromium was controlled by hydroxide, whereas the solubility of managanese was controlled by acetate at a pH value of 5.00 and by carbonates at a pH value of 7.00 (in absence of acetate). The solubility of cerium was controlled by acetate at a pH value of 5.00. In absence of acetate, however, at a pH value of 7.00 cerium was present as  $Ce^{3+}$ . Sodium and potassium were present as  $Na^+$  and  $K^+$ , respectively. Calcium and magnesium formed complexes with acetate at a pH value of 5.00, whereas at a pH value of 7.00 in the absence of acetate these metals formed complexes with carbonates.

Component	Concentration <sup>*</sup> M
Metal:	
$Ca^{2+}$ $Mg^{2+}$ $Sr^{2+}$ $K^{+}$ $Na^{+}$ $Fe^{2+}$ $Mn^{2+}$ $Zn^{2+}$ $Ni^{2+}$ $Co^{2+}$ $Cr^{3+}$ $Ce^{3+}$	1.0 x $10^{-2}$ 1.0 x $10^{-2}$ 1.0 x $10^{-5}$ 2.0 x $10^{-2}$ 2.0 x $10^{-2}$ 1.0 x $10^{-2}$ 5.0 x $10^{-4}$ 1.0 x $10^{-3}$ 5.0 x $10^{-4}$ 2.0 x $10^{-4}$ 1.0 x $10^{-4}$ 7.0 x $10^{-6}$
Ligand:	
co3 <sup>2-</sup> s <sup>23</sup>	2.0 x $10^{-2}$ , 1.0 x $10^{-2}$ 1.0 x $10^{-2}$
Acetate	5.0 x 10 <sup>-1</sup> , 1.0 x 10 <sup>-1</sup> , None

# Table 10. Data Set Used to Study Metal Speciation by Chemical Modelling (MINEQL)

\*Electroneutrality was ignored.

Note: The stability constants for formation of complexes and solids from metals and ligands are presented in Morel, Francois M. M., "Principles of Aquatic Chemistry," A Wiley-Interscience Publication New-York, 1983.

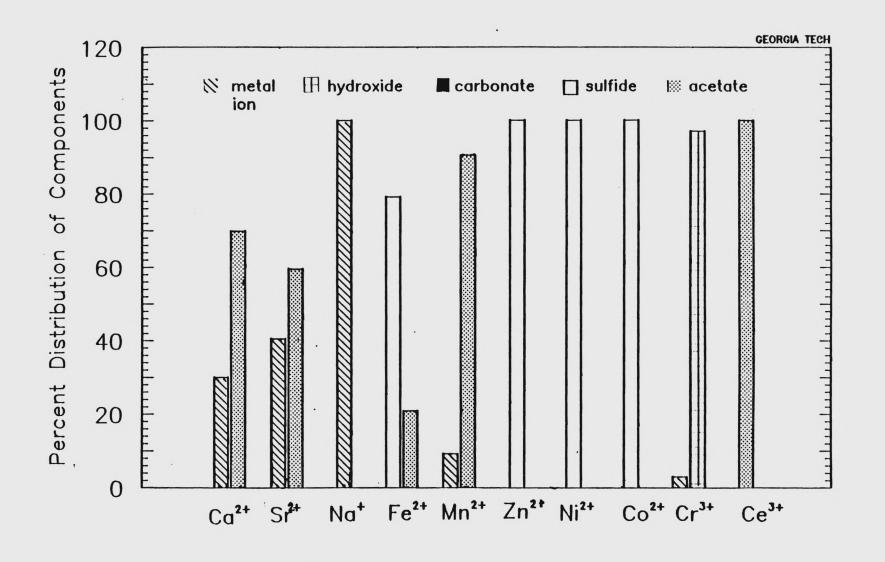


Figure 4. Speciation of Metals by Chemical Modeling (MINEQL) (Carbonate=.01M,Acetate=.5M,Sulfide=.01M, pH=5.00).

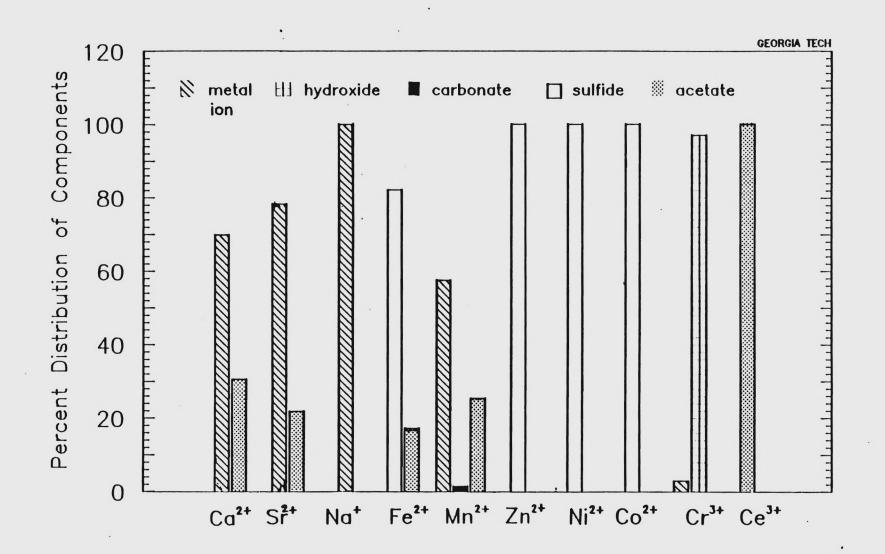


Figure 5. Speciation of Metals by Chemical Modeling (MINEQL) (Carbonate=.01M,Acetate=.1M,Sulfide=.01M, pH=5.00).

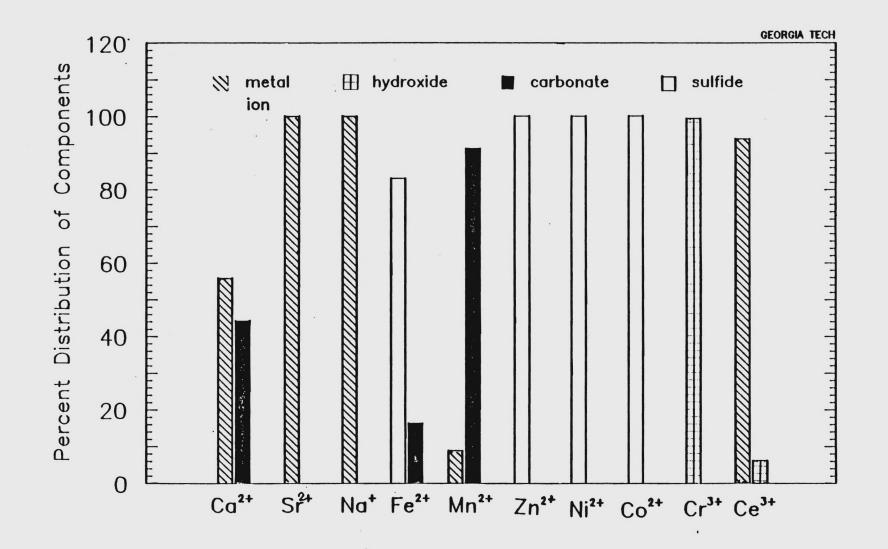


Figure 6. Speciation of Metals by Chemical Modeling (MINEQL) (Carbonate=.01M,Sulfide=.01M, pH=7.00).

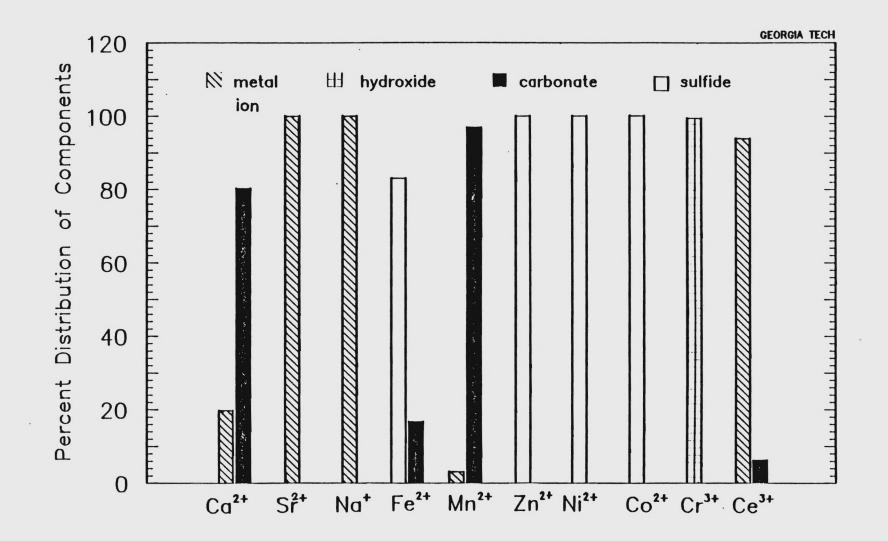


Figure 7. Speciation of Metals by Chemical Modeling (MINEQL) (Carbonate=.02M,Sulfide=.01M, pH=7.00).

#### 2.2.8 Complexation of Radionuclides with Humic Acid

The codisposal of LLRW and municipal refuse within laboratoryscale lysimeters, with and without leachate recycle, was investigated by Chang (21). One of the radionuclides (Co-58) utilized to simulate LLRW, exhibited a two-fold increase in leachate during the later phases of landfill stabilization. Chang (21) speculated that the increase in levels of Co-58 in leachate was probably due to the formation of soluble complexes of cobalt with humic substances present in leachate.

Swanson (81) reported enhanced mobility of Ni-63 and Pu-239 due to formation of complexes with organic ligands (EDTA, NTA, picolinate, citrate, and oxalate). Based on their studies on complexation of radionuclides with organic chelating agents present in nuclear waste, Toste <u>et al.</u>, (5) have identified a need for studies on complexation of radionuclides, that constitute LLRW, with naturally-occurring organics, such as humic acids.

#### CHAPTER III

## MATERIALS AND METHODS

## 3.1 Construction of Pilot-Scale Landfill Cells

The pilot-scale landfills were simulated in two cells 3.05-m (10 ft) square and 4.28-m (14-ft) deep. The cell walls were constructed with reinforced concrete blocks and installed on sloping reinforced concrete slabs. These cells were originally constructed and utilized for studies on "Controlled Landfill Stabilization by Leachate Recycle" (23). To prevent moisture infiltration from the adjoining soils, the inner faces of the cell walls were coated with successive coats of a foundation sealant and an epoxy based paint. Since containment of the radionuclides which were codisposed with shredded municipal refuse within the pilot-scale landfills was necessary, both cells were lined with 0.76-mm (30-mil) high density polyethylene liners (HDA Gundline Liner, Gundle Lining Systems, Inc., Houston, TX). The liner for each cell was assembled on-site, then placed above the underdrain network installed in a 15.2-cm (6-in) sand layer to provide continuous monitoring for liner integrity. Each underdrain network consisted of a 5.08-cm (2-in) PVC main with three 5.08-cm (2-in) laterals extending on either sides; these pipes were perforated with 0.95-cm (3/8-in) holes drilled 15.24 cm (6 in) on center. The mains extended through the front wall of each cell to a control sump. One of the cells (Cell A) was designed for single pass operation, while the other (Cell B) was

designed for leachate recycle.

Each cell was equipped with appurtenances for collecting leachate. The collection networks, similar in arrangement to the underdrain networks, were placed just above the liner at the bottom of each cell in a 15.24-cm (6-in) layer of sand. The collection network main extended through the liner and the front wall of each cell to two collection sumps. These collection sumps along with the collection sump for underdrain network were 208-L (55-gal) polyethylene containers; they were placed in a fiber glass coated plywood tank 2.43 m (8 ft) x 0.91 m (3 ft) x 1.52 m (5ft) (length x width x height). A submergible pump with epoxy coated housing and polyethylene impeller was utilized to recycle the leachate collected from Cell B. The leachate recycle network was constructed with a 5.08-cm (2-in) diameter Schedule 40 PVC pipe and compatible PVC fittings. A leachate distribution network, similar in arrangement to the underdrain network, was installed in a 0.3-m (1-ft) bed of gravel above the shredded municipal refuse placed in each cell. Figure 8 presents a sectional view of Cell Β.

The cells were tested for leaks by filling them with water and noting the change in water levels after 24 hours. An insignificant change in the water level in each cell was interpreted as an assurance of sealing of all the joints in the liner. After draining the water, a layer of Geocloth (Fabric, Gundle Lining Systems, Inc., Houston,TX) was placed above the liner in each cell, to separate the liner from any sharp edged material contained in the shredded municipal refuse. The cells were then ready for placement of shredded municipal refuse.

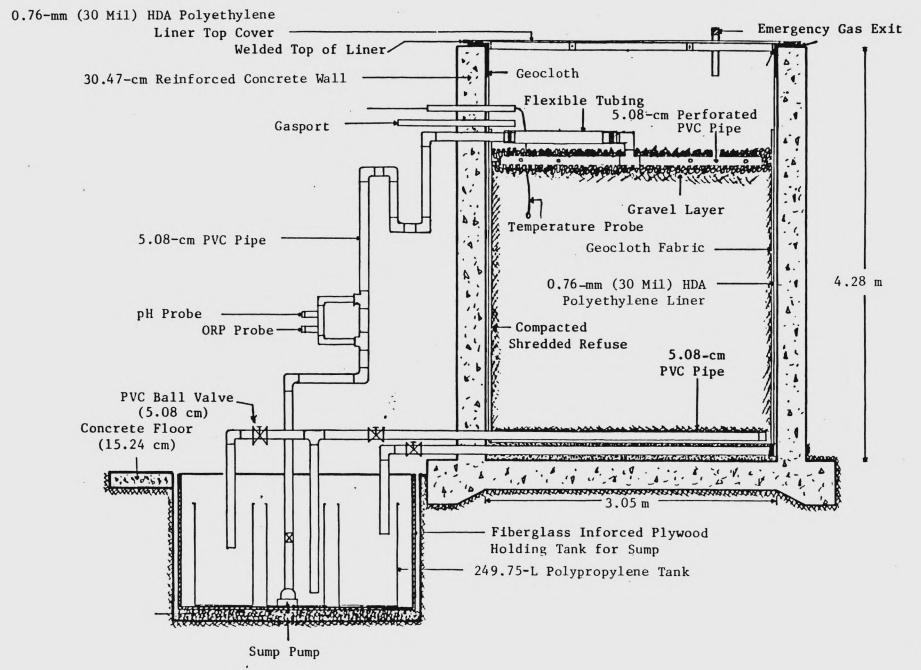


Figure 8. Schematic of simulated landfill with recycle.

#### 3.1.2 Placement of Shredded Municipal Refuse

Shredded municipal refuse was loaded in a transfer truck at the Shredding Facility in Dekalb County, Georgia on 1 July 1982 and delivered onsite the following morning. The refuse was placed within the lined cells with a front-end loader and a conveyor system. During placement, the refuse was compacted by two hand-held gasoline powered tampers. The density of the compacted refuse within the landfills was estimated by compacting representative samples of refuse in 208-L (55-gal) drums by hand-held tampers concomitantly with the loading operation, then determining the weight of the compacted refuse. Also, representative samples of refuse were collected in 3.78-L (1-gal) glass containers at the beginning and the end of each work day; these samples were analyzed for moisture content, carbon, hydrogen, nitrogen, and calorific values. Refuse placement was interrupted when height of compacted refuse reached 1.22 m (4 ft) in each cell and a 0.03-m (1.5-in) layer of SaniFoam (urea-formaldehyde based plastic foam) was sprayed as a cover on the surface of the refuse in both cells.

Placement of refuse was resumed on 3 July 1982. Prior to loading of refuse in each cell the cured foam layer was crushed manually. A solution containing Co-58 (halflife = 70.8 d), Sr-85(half life = 64.5 d), and Ce-141 (half life = 32.4 d) was diluted to 50 mL with 0.5 M HCl. A 0.05-mL (50 $\lambda$ ) aliquot was withdrawn from the 50-mL radionuclide solution, diluted to 400 mL with 0.5 M HCl and placed in a cottage cheese container for counting. Three such solutions were prepared for counting. The volume in the 50-mL volumetric flask was brought to 50 mL by adding 0.5 M HCl and 25 mL of this solution were spiked on the

refuse placed in each cell. The resulting amounts of Co-58, Sr-85, and Ce-141 in each cell were 0.91 mCi, 1.09 mCi, and 5.78 mCi, respectively. Compaction of refuse was resumed after a 0.52-m (2-ft) layer of refuse was placed above the crushed foam layer. Upon completion of refuse placement, the average heights of compacted refuse were 2.54 m (8 ft 4 in) and 2.72 m (8 ft 10 in) in Cell A and Cell B, respectively.

Final closure of the cells was initiated on 4 July 1982. In each cell a distribution network was embedded in a 0.3-m (1-ft) layer of gravel. On 6 July 1982,the top of each cell was sealed with the liner material and the creases along the joints were vacuum tested for leaks. The entire structure was covered by a plywood roof. An instrumentation shed was constructed on a reinforced concrete slab; it housed leachate collection sumps, pH/ORP meter, flow meter for gas measurement, temperature recorders, and 208-L (55-gal) drums for storing leachate from Cell A (single pass cell). The entire structure was fenced off and access to the shed was controlled. The project site was inspected and approved by the Radiological Safety Officer of Georgia Tech Nuclear Safeguards Committee, Georgia Tech, Atlanta, GA.

## 3.2 Moisture Addition

Leachate generation from each cell was initiated by raising the moisture content of the refuse to field capacity (Cell A, 101.8%; Cell B, 100.5%; moisture content was based on the dry weight of the refuse). Since rainfall occurred between the conclusion of refuse placement and the initiation of final closure, a rainfall equivalent of 492 L (130 gal) was included in the calculation for the volume of water needed to

raise the moisture content of the refuse to field capacity. The total volumes of water (tap water + rainfall equivalent) added to Cell A and Cell B were 1893 L (500 gal) and 1949 L (575 gal), respectively. Thereafter, rainfall was simulated by adding rainfall equivalent volumes to both cells through the distribution networks. Addition of rainfall equivalent was terminated on Day 56, since accumulation of leachate at the bottom of Cell B exceeded 0.3-m (1 ft) head. Rainfall data for Atlanta, GA were obtained from the Georgia Tech Research Institute (GTRI), Atlanta, GA.

#### 3.3 Analytical Methods

Leachate collection from each cell was initiated on 27 July 1982 (Day 0). Samples were collected at weekly intervals up to Day 150, at 2-week intervals from Day 150 to Day 570, and at monthly intervals from Day 570 to Day 970. From Day 290 to Day 350 collection of leachate samples was suspended and, the cells were tested for gas leaks during that period. After repeated testing, it was concluded that the cells were not leak proof for gas. Leachate collection was resumed on Day 350. Leachate recycle, however, was maintained at a frequency of once a week during that period (Day 290 to Day 350). Leachate samples were analyzed for pH, oxidation-reduction potential (ORP), five-day biochemical oxygen demand (BOD<sub>5</sub>), chemical oxygen demand (COD), total carbon (TC), total inorganic carbon (TIC), total organic carbon (TOC), conductivity, total alkalinity, individual and total volatile fatty acids (acetic, propionic, isobutyric, butyric, and valeric), sulfide, chloride, metals (Fe, Zn, Ni, Mn, and Co), and radionuclides (Co-58, Sr-85, and Ce-141). In addition to these parameters, nutrients such as nitrogen and phosphorus present in leachate are important indicator parameters which describe landfill stabilization. In this study, however, leachate samples were not analyzed for nutrients.

#### 3.3.1 Gross Parameters

Total alkalinity, BOD5, and COD were measured as per methods included in "<u>Standard Methods</u>" (1975). TC and TIC were measured with a Beckman Model 915 Total Carbon Analyzer (Fullerton, CA). A Fisher Model 144 pH/mV meter was used to measure pH and ORP. Conductivity was measured with a Yellow Springs Instrument Company Model 31 Conductivity Bridge (Yellow Springs, OH).

## 3.3.2 Volatile Fatty Acids

The volatile fatty acids present in leachate samples were measured using an Internal Standard Method on a Hewlett Packard Model 5710A gas chromatograph (GC) equipped with a flame ionization detector (FID; Avondale, PA). Leachate samples were filtered through a 0.45-µm filter (Gelman, Ann Arbor, MI), then acidified to a pH value less than 2 with 85% phosphoric acid (3.5 mL acid/100 mL sample). In addition, 1-butanol (20- L 1-butanol/100 mL acidified sample) was added as the internal standard. Table 11 lists the GC program conditions utilized to measure volatile fatty acids.

## 3.3.3 Heavy Metals

Leachate samples were analyzed for the following heavy metals: iron (Fe), zinc (Zn), nickel (Ni), manganese (Mn), and cobalt (Co). These heavy metals were chosen as they were expected to behave similar to the behavior exhibited by cobalt. In addition to these heavy metals

Gas Chromatograph	Hewlett Packard 5710A/ 3380A Integrator
Injection Temperature	250°C
Detector Temperature	250 <sup>0</sup> C
Oven Temperature Program Initial Temperature and Time Rate of Increase in Temperature Final Temperature and Time	100 <sup>0</sup> C for 4 min 4 <sup>0</sup> C/min 180 <sup>0</sup> C for 0 min
Column Details	
Material and Dimensions	Glass; 2m long by 2mm I.D.
Stationary Phase	<pre>1% Carbowax 20 M; 15%     phosphoric acid on     Carbopack B, #60H#80     mesh.</pre>
Carrier Gas	Nitrogen @ 40 mL/min

Table 11. Gas Chromatographic Conditions Used for Measuring Volatile Fatty Acids (83)

the leachate samples were also monitored for chromium (Cr) and copper (Cu). The analyses for these metals were discontinued after Day 50, since their concentrations in leachate were always less than 1 mg/L. The samples were preserved as per procedures included in "<u>Methods of Chemical Analysis of Water and Wastes</u>" (1979) (113). Two atomic absorption spectrophotometers, i.e., Perkin Elmer Model 303 and Perkin Elmer Model 703 (Norwalk, CT) were utilized to determine the concentration of each heavy metal present in digested leachate samples. Model 703 spectrophotometer was used to measure metal concentration in  $\mu$ g/L range, and Model 303 spectrophotometer was used to measure metal concentration in mg/L range.

Leachate samples, prior to analysis, were digested as per the following procedure (84): to 100 mL of sample contained in a 250-mL beaker, a 5-mL aliquot of 1:1 diluted nitric acid (HNO<sub>3</sub>; Ultrex) was added, covered with a watch glass, then evaporated (without boiling) to near dryness on a hot plate. After the sample was cooled, a 4-mL aliquot of concentrated HNO<sub>3</sub> (Ultrex) was added, and again evaporated to near dryness. After cooling, a 1-mL aliquot of 1:1 HNO<sub>3</sub> (Ultrex) and a 3-mL aliquot of 30% hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) were added to the sample which was then heated until the effervescence from the addition of H<sub>2</sub>O<sub>2</sub> subsided. After cooling, the addition of 30% H<sub>2</sub>O<sub>2</sub> in 1-mL aliquots and evaporation of the sample were repeated until the effervescence was minimal or the general sample appearance was unchanged. No more than 10 mL of 30% H<sub>2</sub>O<sub>2</sub> was added to the sample. After cooling, a 1-mL aliquot of 1:1 HNO<sub>3</sub> (Ultrex) was added and the sample was then heated for ten minutes. The sample was then cooled, diluted to 100 mL

with deionized water, and stored in a polyethylene container. Digestion blanks of deionized water were prepared along with digestion of leachates.

#### 3.3.4 Sulfide

Leachate samples were analyzed for sulfide with a silver/sulfide electrode (Orion Model 94-16; Orion Research, Inc., Cambridge, MA) and a pH/mV meter (Fisher Accumet Model 610; Fisher Scientific, Atlanta, GA). The method, included in the manufacturer's instructions, is presented in subsequent sub-sections.

3.3.4.1 <u>Preparation of Deoxygenated Deionized Water (DDW)</u>. Approximately 3 L of deionized water was deoxygenated by purging the water with nitrogen for approximately 30 minutes. This water, hereafter referred to as DDW, was used for preparing all working solutions.

3.3.4.2 <u>Sulfide Anti-Oxidant Buffer (SAOB 11)</u>. To 600 mL of DDW placed in a 1-L volumetric flask, 200 mL of 10 M NaOH, 35 g ascorbic acid, and 67 g disodium ethylenediaminetetraacetate were added. The mixture was swirled, then diluted to 1 L with DDW. Fresh SAOB 11 ranged in color from clear to pale yellow-brown and was stored in a tightly stoppered bottle. Upon oxidation, SAOB 11 turned dark brown and was discarded.

3.3.4.3 <u>Sulfide Standards</u>. A stock solution of saturated sodium sulfide was prepared by dissolving approximately 100 g of reagent grade  $Na_2S \cdot 9H_20$  in 100 mL of DDW. The mixture was well shaken, allowed to stand overnight, and stored in a tightly stoppered bottle.

A Weekly Sulfide Standard was prepared by pipetting 1 mL of the

stock solution into a 100-mL volumetric flask, then adding 50 mL of SAOB 11. The mixture was diluted to 100 mL with DDW and the exact concentration C was determined by titrating 40 mL of Weekly Sulfide Standard with 0.1 M lead perchlorate  $Pb(Cl0_4)_2$  and noting the change in potential (mV). At the end point of titration the change in potential ( $\Delta$ mV) was the largest.

$$C = 3200 (V_{+}/V_{S})$$
(1)

where;

C = concentration of sulfide, mg/L

 $V_t$  = volume of titrant at end point, mL

 $V_{S}$  = volume of standard; mL

To prepare a calibration curve, sulfide standards were prepared by making ten-fold serial dilutions of the Weekly Sulfide Standard with equal volumes of SAOB 11 and DDW.

3.3.4.4 <u>Sulfide Measurement</u>. Three sulfide standards were prepared by serial dilution of the Weekly Sulfide Standard (Section 3.3.4.3). The silver/sulfide electrode was placed in the beaker containing the least concentrated standard and the potential (mV) was noted while the standard was stirred gently. The electrodes were then taken out, rinsed, blot dried, and the procedure was repeated for the mid-range standard and the most concentrated standard. A calibration curve was prepared by plotting sulfide concentrations (log axis) versus millivolt readings (linear axis) on a semilogarithmic paper.

To measure sulfide present in a leachate sample, 50 mL of SAOB 11 was added to 50 mL of sample in a 150-mL beaker. The solution was

stirred gently and the potential (mV) was noted. The concentration was read directly from the calibration curve which was prepared within two hours.

## 3.3.5 Chloride

Leachate samples were analyzed for chloride using a potentiometric method similar to the method used for sulfide (Orion Research, Inc., Cambridge, MA). Pretreated leachate samples were titrated with 0.0142 N silver nitrate (AgNO<sub>3</sub>).

Leachate samples, prior to titration, were pretreated as per the following method: a 10-mL aliquot of sample was diluted to 100 mL with deionized water, acidified with a 2-mL aliquot of concentrated sulfuric acid ( $H_2SO_4$ ), and boiled for five minutes. After cooling, a 5-mL aliquot of 50% hydrogen peroxide ( $H_2O_2$ ) was added, and the sample was boiled again for 15 minutes. After cooling, a 10-mL aliquot of 10 M sodium hydroxide (NaOH) was added, and the sample was diluted to 100 mL with deionized water. The sample was boiled again for five minutes, then filtered while hot with a glass fiber filter (Gelman, Ann Arbor, MI). After cooling, a 4-mL aliquot of nitric acid ( $HNO_3$ ; Ultrex) was added and the sample was diluted to 100 mL with deionized water. This pretreated sample was then titrated 0.0142 N solution of silver nitrate.

## 3.3.6 Temperature

Temperature within the refuse in each cell was recorded on a temperature recorder (Series 9354; Cole-Parmer Instrument Company, Chicago, IL) with a temperature probe (Series 703, YSI; Cole-Parmer Instrument Company, Chicago, IL) which was embedded within the refuse

at 0.6 m from the refuse surface.

# 3.3.7 Radioactivity

Leachate samples in 400-mL aliquots were filtered through 0.45-  $\mu$ m filters (Gelman, Ann Arbor, MI), placed in cottage cheese containers, and analyzed for Co-58, Sr-85, and Ce-141 on a Ge(Li) detector. Table 12 lists the equipment used to detect Co-58, Sr-85, and Ce-141.

## 3.4 Complexation Studies

During landfill stabilization, the precipitation of heavy metals, that are present in the leachate, by potential-precipitant species, such as sulfides, carbonates, and hydroxides is counteracted by the potential formation of soluble metal-humate complexes. During the course of investigation, however, the radionuclides, Co-58, Sr-85, and Ce-141, present in leachate, decayed to below detectable levels. Thus, their potential to form soluble metal-humate complexes could not be evaluated. Since Chang (21), based on a study on codisposal of municipal refuse, Co-58, Sr-85, Cs-137, and H-3, reported an increase only in the Co-58 levels in leachate during the methane fermentation phase of landfill stabilization, a companion laboratory-scale investigation was embarked upon to evaluate the potential of humic substances in leachate to mobilize Co-58 from previously precipitated cobalt sulfide by forming soluble cobalt-humate complexes. Leachate samples from the pilot-scale landfill with leachate recycle (Cell B) were utilized to assess the potential of the humic substances present in leachate to mobilize Co-58 from cobalt sulfide.

The analytical methodology utilized for the investigation of

Table 12. 'Equipment Used to Measure Co-58, Sr-85, and Ce-141 Present in Leachate Samples Collected from the Pilot-Scale Landfills

Detector Company Shield Liquid Nitrogen	Ge(Li) Canberra 1.75 mm lead wall with 3.17 mm copper liner (ADT Co.) Type LR-31 Serial Number 80RF-76 (Union Carbide Co.)
Preamplifier Company Model	Canberra #2001
High Voltage Power Supply Company Model Operating Voltage	Canberra #3105 +2500 V
Spectroscopy Amplifier Company Model Coarse Gain Fine Gain	Canberra #141 10 4.34
Multichannel Analyzer Company Model Counting Time	Canberra #8100; One channel equivalent to 1 KeV 2000 sec
Printer Company Model	Hewlett Packard #9830A
Calculator/Plotter Company Model	Hewlett Packard #9862A

potential complexation of Co-58 with leachate was developed using model compounds, since reproducibility and reliability of the results produced from the methodology utilized were key to the success of this investigation. A swamp-based humic acid (A-2HA) was selected as the model humic acid, since it was well-characterized (85) and was of aquatic origin. Cobalt was selected as the model metal since the shredded municipal refuse was spiked with Co-58 to simulate codisposal of municipal refuse and LLRW within the pilot-scale landfills. Analytical grade cobalt sulfide (CERAC, Milwaukee, WI) was utilized to simulate precipitated cobalt within landfills. The precipitated cobalt sulfide within a landfill, however, will be associated with the precipitates of other heavy metals and inert solids. The use of analytical grade cobalt sulfide to examine the potential of humic substances present in leachate to mobilize cobalt from cobalt sulfide precipitated within a landfill will yield higher levels of cobalt in leachate, since the humic substances in leachate will only complex cobalt, as opposed to the case in an actual landfill, wherein cobalt along with other heavy metals will be complexed by the humic substances. Thus, the values generated, at equilibrium, for the mobilization of Co-58 from cobalt sulfide by humic substance in leachate can be regarded as "worst-case" results. Stable cobalt was replaced by a mixture of Co-58 and stable cobalt in the confirmatory study since Co-58 is measured more accurately than stable cobalt in dilute solutions of cobalt and the results of this study ascertained the complexation of Co-58 by humic substances present in leachate. Development of the analytical methodology was implemented in

five phases:

i) development of the molecular-weight fractionation scheme;

- ii) development of the analytical methods for equilibrium studies on complexation of cobalt with model humic acid (A-2HA);
- iii) development of the analytical methods for kinetic studies on complexation of cobalt with model humic acid (A-2HA);
- iv) equilibrium and kinetic studies on complexation of cobalt with leachate from the pilot-scale landfill with leachate recycle (Cell B); and,
- v) equilibrium studies on complexation of Co-58 with model
   humic acid (A-2HA) and leachate from Cell B.

### 3.4.1 Molecular-Weight Fractionation Scheme

A molecular-weight fractionation scheme, which included ultrafiltration (UF) and gel permeation chromatography (GPC) was developed for this study. Chian and DeWalle (78) reported that parallel ultrafiltration of leachate samples resulted in a more representative separation than serial ultrafiltraton; thus, parallel ultrafiltration was incorporated in the molecular-weight fractionation scheme. Aqueous solutions of the model humic acid (A-2HA) at pH values of 5.5, 7.0, and 8.5 were prepared using the procedure outlined in Figure 9. These solutions and leachate samples were fractionated into different molecular-weight fractions by UF and GPC. Figure 10 presents the molecular-weight fractionation scheme.

3.4.1.1 <u>Ultrafiltration (UF)</u>. Ultrafiltration (UF) of model humic acid (A-2HA) and leachate samples was performed in a stirred UF

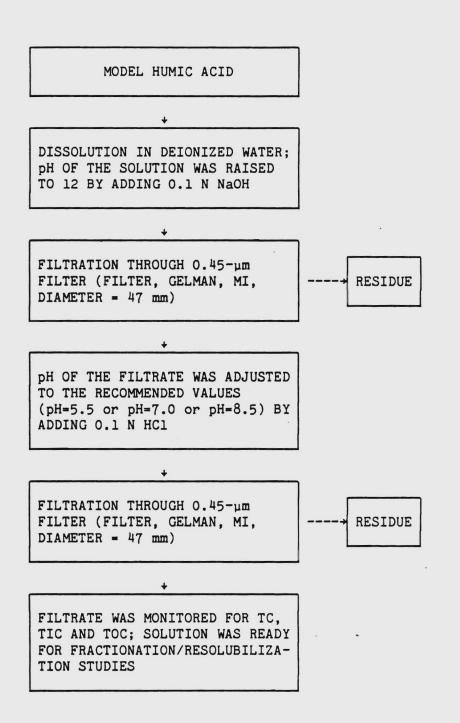


Figure 9. Preparative Procedure of Aqueous Solution of Model Humic Acid

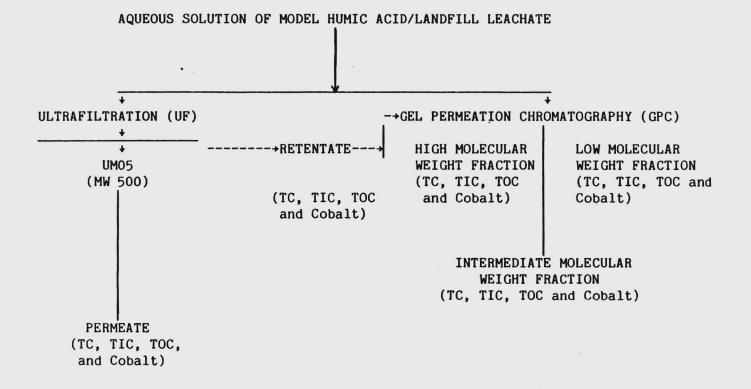


Figure 10. Molecular Weight Fractionation of Model Humic Acid and Landfill Leachates.

cell (Amicon, Bedford, MA) with UMO5 UF membrane (Diaflo Ultrafilters, Amicon, Bedford, MA). This membrane had a nominal weight cutoffs of 500 daltons. Prior to ultrafiltration, the UF membranes were checked for leaks by testing the retention of Blue Dextran (MW >  $2x10^6$  daltons). Nitrogen was used to pressurize the sample within the UF cell; the operating pressure was maintained at 2.41 x  $10^5$  Pa (35 psi). Table 13 presents the experimental details of ultrafiltration.

Ultrafiltration was used to determine the extent of cobalt bound to the high-molecular-weight fraction of both the model humic acid (A-2HA) and the humic substances present in the leachate. This technique is based on the premise that a UMO5 UF membrane (MW cutoff: 500 daltons) would retain the high-molecular-weight organic fraction and any cobalt associated with it. The original sample and its ensuing fractions were monitored for cobalt and TOC.

3.4.1.2 <u>Gel Permeation Chromatography (GPC)</u>. The determination of molecular-weight distribution profiles of humic substances found in soils, sediments, and natural waters by gel permeation chromatography (GPC) especially, with Sephadex gels (Pharmacia Fine Chemicals, NJ), has been reported by many researchers (87-108), Gel permeation chromatography is influenced by several factors that include column size, pH, volume, concentration, and composition of sample, and pH, type, and concentration of eluant. Furthermore, the validity of molecular-weight distribution profiles generated by GPC is enhanced by minimizing gel-solute interactions. There are two main kinds of gel-solute interactions: coulombic interaction caused by charged sites of gel and solute, and adsorption effects. The former, especially for

Ultrafiltration Cell			Ultrafiltration Membranes		
Cell Type	Operating Pressure Pa(psi)	Gas Used to Pressurize the Sample	Туре	Nominal Molecular Weight Cutoff (daltons)	Nominal <sup>*</sup> Pore Diameter (µm)
Stirred UF	2.41 x 10 <sup>5</sup> (35)	Nitrogen	UM05	500	0.0021
Cell; Max Sample					
Volume = 500 mL					
Max pressure =	3.44 x 10 <sup>5</sup> (50)				

# Table 13. Experimental Protocol Utilized for Ultrafiltration of Leachate

\*Perry, R. H., and Chilton, C. H., "Chemical Engineering Handbook", 5th Ed., McGraw Hill Book Company, pp 19-84. polyelectrolytes impose an additional charge-exclusion resulting in an apparent increase in the nominal molecular weight, while the latter results in an apparent decrease. Thus, molecular-weight distribution profiles generated under controlled conditions can be compared, while the practice of assigning nominal molecular weights to the excluded fractions of humic acids is questionable. In this study, GPC under controlled conditions was used to compare molecular-weight distribution profiles of the model humic acid (A-2HA) and the leachate samples with/without soluble cobalt-humate complexes present. Sephadex gels G-50 and G-75 (Pharmacia Fine Chemicals, Piscataway, NJ) were used as the stationary phase. Deionized water was used as the mobile phase and pumped through the entire system by a constant flowrate pump (Milton Roy Co., St. Petersburg, FL). The valve system used for sample application consisted of two SRV-4 four-way valves (Pharmacia Fine Chemicals, Piscataway, NJ) and a 1-mL sample loop. The valve system was operated in three configurations: column standby; filling the sample loop; and, sample application and normal running. A detailed block diagram of the apparatus and the operating sequence of the valve system are presented in Figure 11.

Blue Dextran (MW >  $2 \times 10^6$  daltons) and phenol (MW=94 daltons) were used as standards for the high-molecular-weight fraction and the low-molecular-weight fraction, respectively. The high-molecular-weight cut and the low-molecular-weight cut were established by measuring the absorbance of the ensuing fractions at  $\lambda = 254$  nm. Standards, model humic acid (A-2HA), and leachate samples were injected into the sample loop in 0.5-mL aliquots; the mobile phase, pumped through the sample

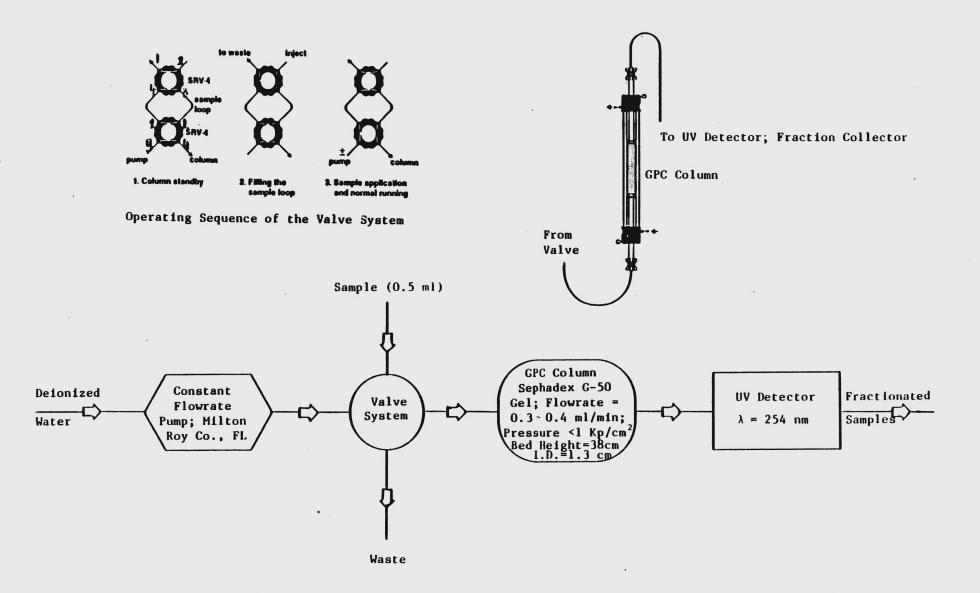


Figure 11. A Block-Diagram of the Apparatus Used for Fractionating A-2HA and Landfill Leachates by Gel Permeation Chromatography (GPC).

loop, introduced the sample into the column, and the ensuing fractions of the model humic acid (A-2HA) and the leachate samples were analyzed for TOC and cobalt. The experimental details of GPC are presented in Table 14.

# 3.4.2 Equilibrium Studies

A quality assurance and quality control (QA/QC) study was performed to determine the significance of the difference in the equilibrium levels of cobalt solubilized from cobalt sulfide by deionized water and the model humic acid (A-2HA), respectively. Measured amounts of cobalt sulfide were added to 20 replicates of deionized water (CoS = 0.065 g  $\pm$  0.011 g) and 17 replicates of the model humic acid (A-2HA; 0.5 g/L; pH = 7.00, CoS = 0.061 g  $\pm$  0.008 g), each contained in a 125-mL French Square bottle without any headspace. These bottles were loaded in a rotary shaker (1 rpm) and allowed to equilibrate for three weeks. The shaker was housed in an incubator; the temperature within the incubator was maintained at 20°C  $\pm$  1°C.

After the equilibration period, samples were withdrawn from the rotary shaker and centrifuged in a ultracentrifuge (Model J2-21, Rotor: JA-20.1, Beckman Instruments, Inc., Palo Alto, CA) at  $20.0^{\circ}C \pm 0.5^{\circ}C$ . The centrates from the centrifuged samples were digested as per the digestion procedure included in Section 3.3.3; the digested samples were analyzed for cobalt with an atomic absorption spectrophotometer (Model 703, Graphite Furnace, Perkin Elmer, Norwalk, CT).

After completion of the QA/QC study, equilibrium studies on complexation of cobalt (from cobalt sulfide), by model humic acid at pH values of 5.5, 7.0, and 8.5 were performed. These pH values were

Stationary Phase		Mobile Phase	Standards	Samples
Gel	Column Details			Fractionated
Sephadex: a modified dextran obtained by crosslinking linear macromolecules, con- sisting of a three- dimensional network of polysaccharide chains.	Bed Height = 38.5 cm Inner Diameter (I.D.) = 1.3 cm	Deionized Water (Flowrate: 0.4~0.5 mL/min)	Blue Dextran (MW>2x10 <sup>6</sup> ) Phenol (MW=94)	Standard, A-2HA and filtered landfill leach- ate (0.45-μm filter; Gelman, MI)
G-50 (Fractionation range: 500 MW-10,000 MW) G-75 (Fractionation range: 1,000 MW-50,000 MW)				

.

Table 14. Experimental Protocol Utilized for Gel Permeation Chromatography (GPC) of Leachate

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selected since the pH of landfill leachate may vary from 4.5 during acid formation phase to 9.0 as may be the case due to generation of ammonia by microbially-mediated reduction of nitrates or presence of caustic metal sludges in the landfill. Moreover, microenvironments with different pH values are known to coexist within a landfill. Thus, equilibrium studies on complexation of cobalt with model humic acid (A-2HA) were performed at different pH values.

The procedure utilized for these studies was similar to the procedure developed for the QA/QC study. In addition, the centrates from centrifuged samples were fractionated as per the molecular-weight fractionation scheme included in Section 3.4.1 (Figure 3). In the UF fractionation scheme, however, only a UMO5 UF membrane (MW cutoff: 500 daltons) was used, thereby fractionating the sample into the permeate (low-molecular-weight) and the retentate (high-molecular-weight) fractions. The sample, the permeate, and the retentate were digested as per the procedure included in Section 3.3.3, then analyzed for cobalt by atomic absorption spectroscopy (Perkin Elmer Model 703, Graphite Furnace Atomic Absorption Spectrophotometer, Norwalk, CT). The procedure utilized for equilibrium studies on complexation of cobalt (from cobalt sulfide) by filtered leachate samples (0.45  $\mu$ m filter, Gelman, Ann Arbor, MI) was similar to the aforementioned procedure, and is presented in Figure 12.

3.4.3 Kinetic Studies

To determine the time required by the model humic acid (A-2HA) for solubilizing cobalt to equilibrium levels from cobalt sulfide, a kinetic study was performed. Known amounts of cobalt sulfide were

MODEL HUMIC ACID (A-2HA)/LEACHATE SAMPLES

ADDITION OF COBALT SULFIDE TO SIMULATE PRECIPITATED COBALT SULFIDE WITHIN THE LANDFILL

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EQUILIBRATION TIME

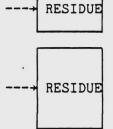
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+

FILTRATION: 0.45-µm FILTER

BECKMAN ULTRACENTRIFUGE; MODEL J2-21 ROTOR: JA- 20.1, JA-10 SPEED: 5,000 RPM TIME: 60 MIN

+



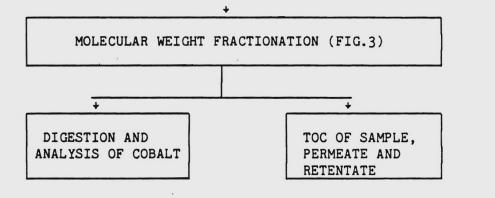


Figure 12. Methods Used for Equilibrium Studies on Complexation of Cobalt by Model Humic Acid (A-2HA) and Leachate Samples.

added to the model humic acid (A-2HA; 0.5 mg/L; pH=7.04; and TOC = 276.8 mg/L) in 125-mL French Square bottles; these bottles were loaded in a rotary shaker. Samples were withdrawn at predetermined intervals filtered, centrifuged, digested, and analyzed for cobalt by atomic absorption spectroscopy (Perkin Elmer Model 703, Graphite Furnace, Atomic Absorption Spectrophotometer, Norwalk, CT). The procedure utilized for leachate samples was similar to the procedure developed for the model humic acid (A-2HA) and is presented in Figure 13.

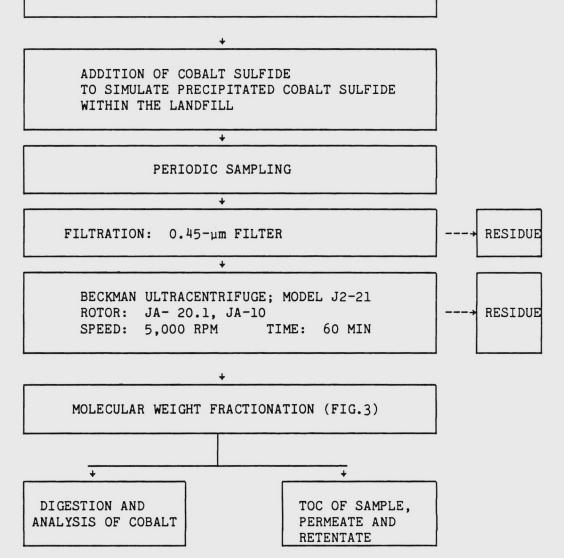
### 3.5 Equilibrium Studies with Co-58

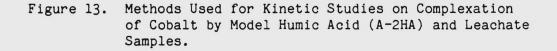
The solubilization of Co-58 from cobalt sulfide (containing Co-58) by model humic acid (A-2HA) and leachate samples was investigated. Upon reaching equilibrium, the samples containing resolubilized Co-58 were fractionated with gel permeation chromatography (GPC); thus, molecular-weight distribution profiles of radioactive cobalt were generated.

The radioactive cobalt (Co-58, 1 mCi) was purchased from Amersham (Arlington Heights, IL), and received by the Radiological Safety Officer, Office of Radiological Safety, Georgia Institute of Technology, Atlanta, GA. The procedure utilized in this study was approved by the Georgia Tech Nuclear Safeguards Committee and the experiments were performed in the Radiochemistry Laboratory of the Nuclear Research Center at Georgia Tech.

The detailed procedure of the equilibrium experiment is presented in four sections: preparation of the tracer solution, precipitation of cobalt sulfide, vacuum filtration step, and the equilibrium experiments.

MODEL HUMIC ACID (A-2HA)/LEACHATE SAMPLES





### 3.5.1 Preparation of Tracer Solution

The tracer solution (20  $\mu$ Ci/mL) was prepared with the addition of a 2-mL aliquot of Co-58 tracer containing 1 mCi to approximately 20 mL of 0.01 M hydrochloric acid (HCl) in a 50-mL volumetric flask; the final volume was adjusted to 50 mL with 0.01 M HCl. This tracer solution was used in the subsequent cobalt sulfide preparation step.

# 3.5.2 Precipitation of Cobalt Sulfide

To approximately 10 mL of deionized water in a 25-mL volumetric flask, 8 mL of cobalt solution (Atomic Absorption Standard, 1 mL = 1 mg; Fisher Chemicals, Atlanta, GA) and 0.5 mL of Co-58 tracer solution were added. The final volume was adjusted to 25 mL and a 0.2-mL aliquot was withdrawn for radioactivity counting. The standard cobalt solution, containing both stable and radioactive cobalt, was transferred into a 125-mL Erlenmeyer flask, then a 5-mL aliquot of sodium sulfide (NaS; 4.3 gm/L) was added. The formation of a black cobalt sulfide suspension was observed.

#### 3.5.3 Vacuum Filtration Step

The cobalt sulfide suspension generated in the previous step (3.5.2) was filtered through a  $0.45-\mu m$  filter (Micro Filtration Systems, Dublin, CA). The Erlenmeyer flask and the filter holder were rinsed with deionized water and the rinse water was filtered through the same paper. The volume of the filtrate was recorded and a 5-mL aliquot was withdrawn for radioactivity counting.

### 3.5.4 Equilibrium Experiment

The filter papers, together with the residues, were placed within correspondingly labelled vials (volume: 24 mL); the vials were

then filled (no headspace) with the model humic acid (A-2HA) at different pH values (5.5, 7.0, and 8.5) or leachate samples, and capped tightly with screw tops. These vials, along with packing material, were placed within wide-mouthed square polyethylene (PE) reagent bottles (180 mL Fisher Chemicals, Atlanta, GA). This minimized movement of the vials during the equilibration period. Furthermore, it reduced the possibility of contamination which could result from an accidental breakage of the vials. The reagent bottles were loaded in a rotary shaker (1 rpm) and the samples were allowed to equilibrate.

The solutions in the vials were monitored for radioactivity at intervals of 12, 28, and 90 days. These intervals were chosen, since equilibrium experiments on mobilization of stable cobalt by humic acid (A-2HA) and landfill leachate indicated that equilibrium was reached within 14 days. After 12 days, the vials were withdrawn from the PE bottles. One-mL aliquots were withdrawn for radioactivity counting and 1-mL aliquots of corresponding ligand solutions were added to the vials. The vials were placed within the PE bottles and allowed to equilibrate in the rotary shaker for 16 more days. After a total of 28 days, 1-mL aliquots were withdrawn and fractionated into different molecular-weight fractions with gel permeation chromatography (GPC). The samples were further equilibrated for a total of 90 days and 1-mL aliquots were withdrawn for radioactivity counting. The samples and the ensuing fractions were analyzed for radioactivity using a sodium iodide detector with a thallium activator [NaI(T1)]; the other equipments used for radioactivity counting are mentioned in Table 12.

#### CHAPTER IV

# RESULTS AND DISCUSSION

Results of analyses performed on the shredded municipal refuse samples which were collected concomitantly with the loading operation and the leachate samples from the two pilot-scale landfill cells along with the environmental data are presented in this chapter. In the time scale used for data presentation, Day 0 corresponds to the initiation of leachate collection on July 27, 1982. Also included in this chapter are the results of the studies performed on the solubilization of Co-58 from cobalt sulfide (containing both Co-58 and stable cobalt) by model humic acid (A-2HA), and leachate samples from the landfill cell equipped with leachate recycle (Cell B). Since the only radionuclide present in the samples was Co-58, the radioactivity in samples has been expressed in counts per minute normalized to Day 0 (March 7, 1985) of the solubilization study.

# 4.1 Characterization of Shredded Municipal Refuse

The characteristics of shredded municipal refuse placed in the pilot-scale landfills (Cell A and Cell B) with respect to as placed refuse density, moisture content, carbon, hydrogen, and nitrogen (CHN), and heat value are presented in Table 15. The as placed refuse density,  $379.9 \text{ kg/m}^3$ , was computed by simulating compaction of municipal refuse

379.9
43.8
5.8
6.2
40
15,210

Table 15. Original Characteristics of Shredded Municipal Refuse Placed within Pilot-Scale Landfills

\*Average of four samples \*\*The as placed density was computed by simulating compaction of municipal refuse with hand-held tampers within 208-L drums concomitantly with the loading operation.

with hand-held tampers within 208-L drums concomitantly with the loading operation; this value, albeit within the range of refuse density reported by Tchobanoglous <u>et al</u>. (53), is higher than value (318.6 kg/m<sup>3</sup>) reported by Pohland (23). The mass of refuse, on an as placed basis, in Cell A and Cell B was 8,976 kg (19,789 lb) and 9,584 kg (21,130 lb), respectively. The mass of refuse placed in each cell was calculated by multiplying the volume of compacted refuse in each cell with the as placed refuse density (379.9 kg/m<sup>3</sup>).

The carbon, hydrogen, and nitrogen contents of the shredded municipal refuse, on dry weight basis, were 43.8%, 5.8%, and 6.2%, respectively. These values are similar to values reported in the literature (23,53), albeit the nitrogen content of 6.2% was higher than values reported by Pohland (23) and Chang (21) for shredded municipal refuse from the same shredding facility. Since the chemical composition of shredded municipal refuse at the same shredding facility may vary seasonally, the higher nitrogen content may be attributed to higher content of food wastes, garden trimmings, textile wastes, and leather wastes in the shredded municipal refuse used in this study as compared to the municipal refuse samples used by Chang (21) and Pohland (23). The heat value of refuse, on a dry basis, was 15,210 kJ/kg; this value was within the range of values reported by Tchobanoglous <u>et al</u>. (53).

# 4.2 Moisture Balance

A periodical evaluation of the moisture present within a landfill is important since it is one of the factors which enhances landfill stabilization. The volume of moisture  $(m^3)$  retained within pilot-scale landfill cells at any time, t, can be expressed as:

$$R/t=t = MA/t=0 + M/t=0 + WR/t=t - L/t=t - LS/t=t - LF/t=t$$
 (1)

where:

- R = the volume of moisture retained within the landfill cell  $(m^3)$ .
- MA = the volume of water added to bring the landfill cell to field capacity (100% moisture content based on dry weight of refuse placed within the cell) (m<sup>3</sup>).
- M = the volume of water corresponding to the initial moisture content of the refuse placed within the landfill cell (m<sup>3</sup>).
- WR = the cumulative volume of water added to the landfill cell to simulate incidental rainfall  $(m^3)$ .
- L = the cumulative volume of leachate drained from the landfill cell  $(m^3)$ .
- LS = the cumulative volume of leachate collected from the landfill cell for analysis (m<sup>3</sup>).
- LF = the cumulative volume of leachate lost from the landfill cell due to liner failure (m<sup>3</sup>).

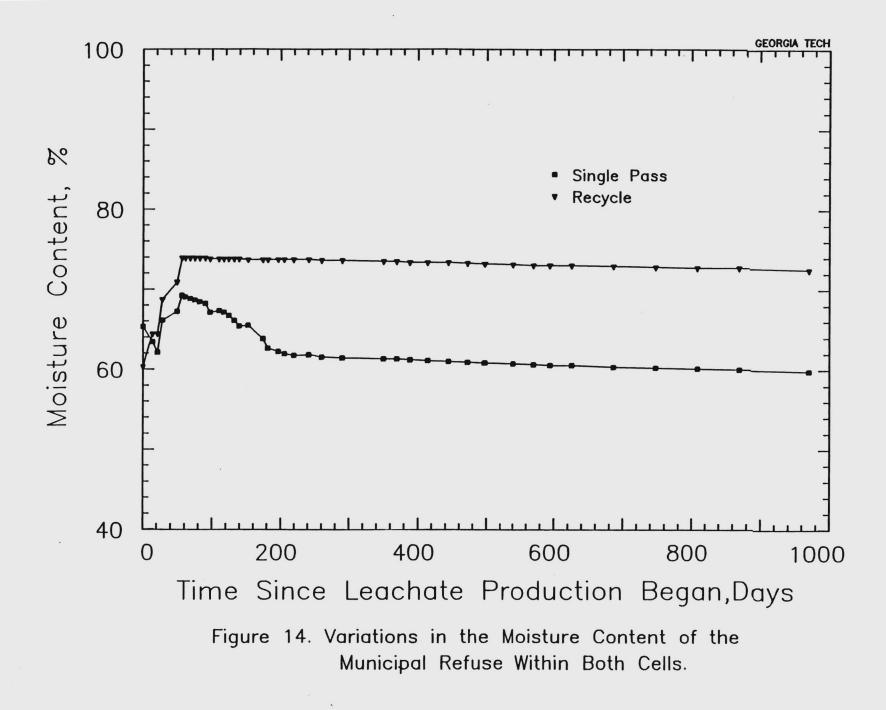
Since the cells were sealed, the entire structure was covered

with plywood roofing, and the leachate collection systems were housed in a shed, the volume of moisture lost due to evaporation could be considered negligible. Moreover, the term LF assumes significance only in an event of a liner failure. Since the absence of leachate in the underdrain system was noted throughout the period of this investigation, the term LF has been dropped from the expression for evaluating R; thus, the modified expression for the volume of moisture retained with the landfill cell at any time, t, is:

$$R_{t=t} = MA_{t=0} + M_{t=0} + WR_{t=t} - L_{t=t} - LS_{t=t}$$
(2)

The moisture balances for Cells A and B are presented in Figure 14. Water additions to the cells simulating incidental rainfall were terminated on Day 56 (September 22, 1982) for two reasons: to simulate the prevention of infiltration after sealing off the cells with covers (constructed from the liner material) and to limit the accumulation of leachate to 0.3 m (1 ft) over the liner at the bottom of Cell B. The moisture content (based on as placed weight of refuse) in Cell A varied between a low of 59.8% (Day 970) and a high of 69.2% (Day 56). The moisture content (based on dry weight of refuse) in Cell B varied between a low of 60.3% (Day 0) and a high of 73.9% (Day 56).

The maintenance of a minimal volume of leachate within landfills by limiting the volume to a 0.3-m head over the bottom of the landfill has two operational advantages: a reduction in size of the appurtenances required for recirculation of leachate and a reduction in volume



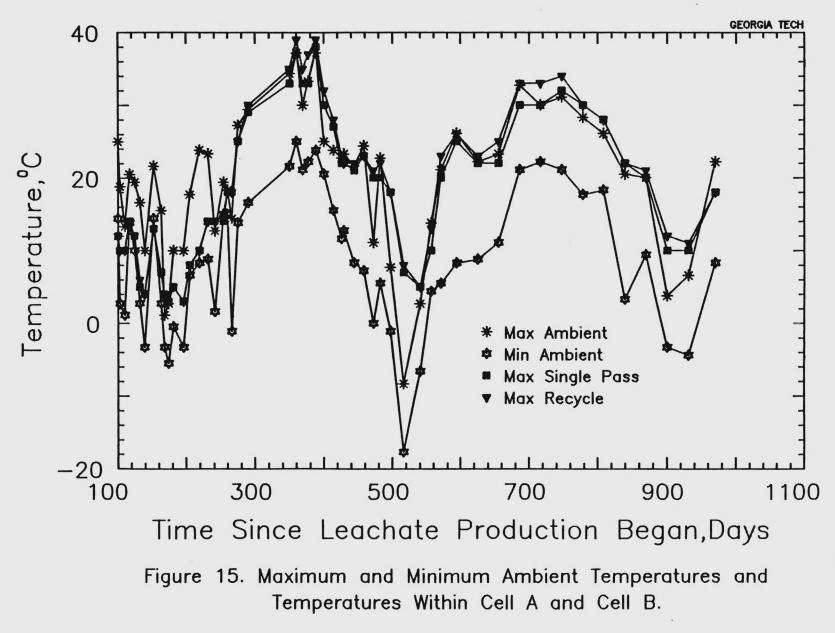
of leachate that may require external treatment. Also, the maintenance of a minimal volume of leachate within landfills will reduce the volume that can potentially migrate into the adjoining environment in the event of a liner failure. Since the presence of moisture content at levels greater than field capacity is essential for the maturation of stabilization processes, the volume of leachate in excess of field capacity can be minimized without disturbing the stabilization processes by recirculation of leachate as reported by Pohland (22).

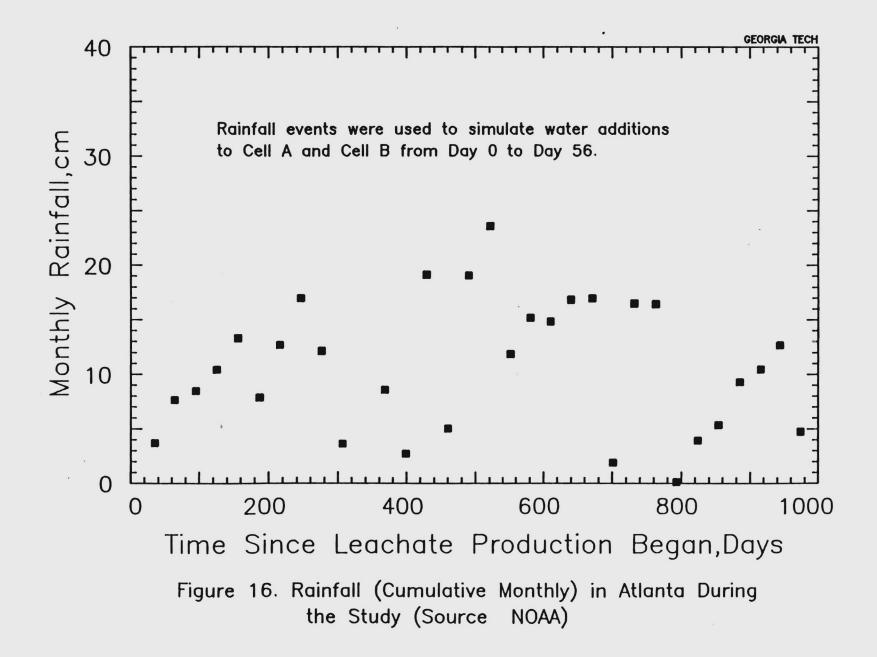
# 4.3 Environmental Conditions

The data on temperature, including maximum and minimum ambient temperatures and maximum temperatures within the cells, and incidental rainfall are presented in Figures 15 and 16, respectively. The maximum temperatures within the cells followed the variations in the ambient temperatures during the study.

### 4.4 Leachate Analysis

The causes of changes in the various constituents present in leachates from the landfill cells were different; in Cell A (single pass), the leachate generated was collected and stored, whereas in Cell B (leachate recycle), the leachate produced was recycled on a daily basis (Day 0 to Day 152 and Day 350 to Day 556), weekly basis (Day 152 to Day 350 and Day 556 to Day 970). The recirculation of leachate provided a continuous exposure of the complex constituents in the leachate and the refuse to the microbial population within the landfill cell. This prolonged exposure enabled the microbial population to



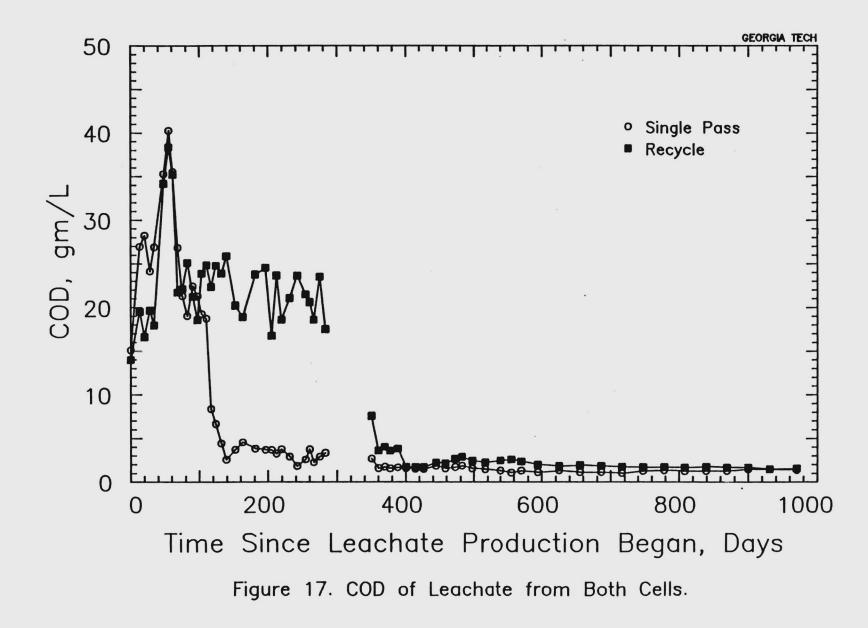


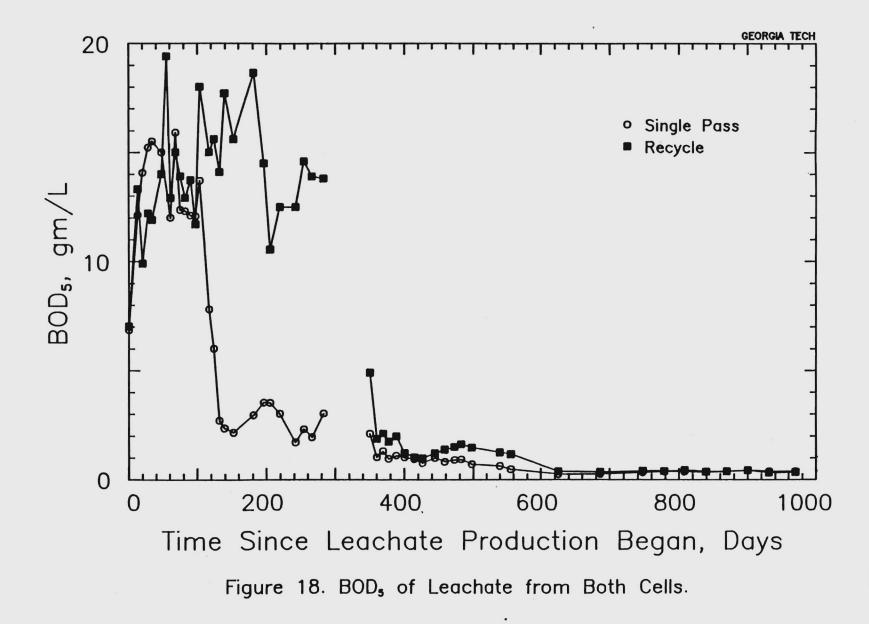
degrade the complex constituents in the leachate and the refuse, thus providing an on-site treatment of the leachate generated from the landfill cell.

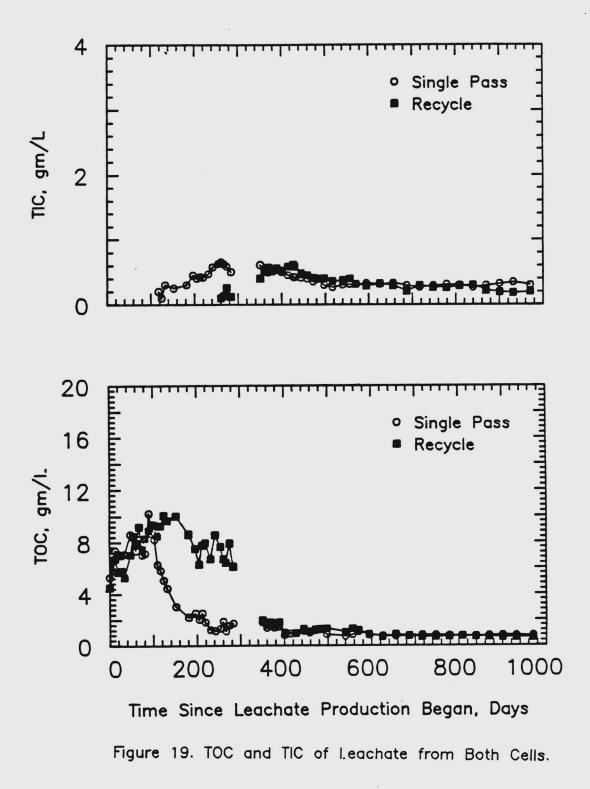
The causes for changes in constituents present in leachate from Cell A, in contrast to Cell B, were a combination of additions of rainfall-equivalent volumes of water, washout due to removal of leachate from the cell, and a result of microbially-mediated stabilization processes that occur in the landfill. Thus, the leachate collected from Cell A during the early stages of landfill stabilization, particularly, the Initial Adjustment Phase, the Transition Phase, and the Acid Formation Phase would require external treatment. In addition, since the addition of rainfall-equivalent volumes to both cells was terminated on Day 56, the changes in constituents present in leachate from both cells due to dilution were minimized beyond Day 56. 4.4.1 Organic Pollutant Parameters

Organic pollutant parameters (COD, BOD<sub>5</sub>, and TOC) were used to reflect the pollutional impact of the leachate produced from the landfill cells. The variations in COD, BOD<sub>5</sub>, and TOC along with the changes in ratios of BOD<sub>5</sub>/COD and COD/TOC are presented in Figures 17-21.

As indicated in Figures 17-19, COD, BOD<sub>5</sub>, and TOC exhibit a similar trend, wherein an increase is followed by a decrease in the concentrations. During the period COD, BOD<sub>5</sub>, and TOC concentrations decreased, the pH of leachate (Figure 25) increased, and the ORP (Figure 28) and the total volatile fatty acids (Figures 22 and 23) decreased. The decrease in COD, BOD<sub>5</sub>, and TOC in leachate from Cell B







occurred during the period (Day 290 to Day 350) when leachate collection was suspended. The cells, however, were tested for gas leaks during this period. Had leachate samples been collected and analyzed during this period (Day 290 to Day 350), the analyses would have reflected the decrease in COD, BOD<sub>5</sub>, and TOC in leachate and expected changes in other indicator parameters. The rate of decrease in these parameters in leachate from Cell B, however, can be envisioned as similar to the rate of decrease in corresponding parameters in the leachate from Cell A, since similar microbially-mediated stabilization processes occurred in both cells.

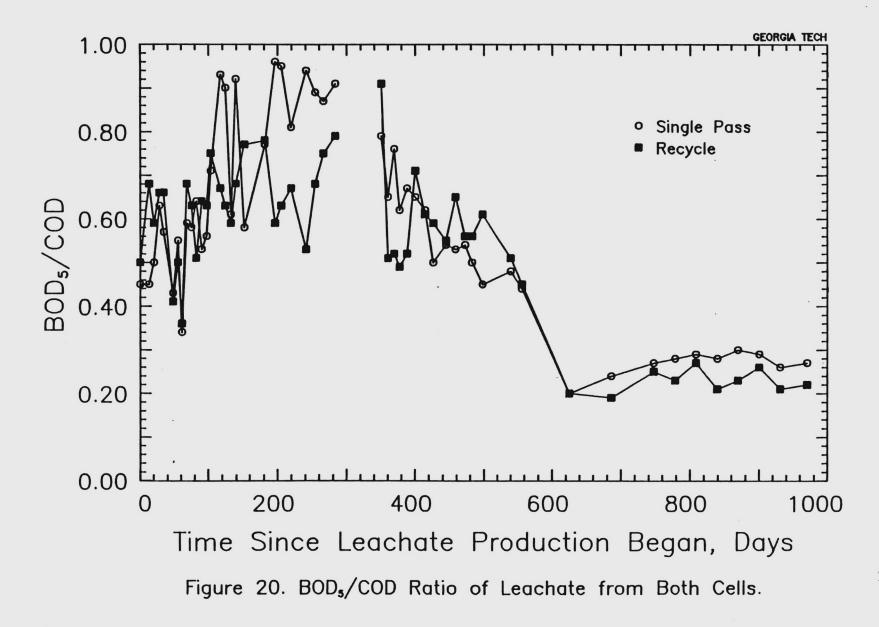
The initial concentrations of COD, BOD5, and TOC in leachate collected from Cell A were 15,100 mg/L, 6,849 mg/L, and 5,300 mg/L, respectively, whereas the final ones were 1,349 mg/L, 370 mg/L, and 767 mg/L. In comparison, the initial concentrations of COD, BOD5, and TOC in leachate collected from Cell B were 13,975 mg/L, 7,025 mg/L, and 4,500 mg/L, respectively, whereas the final ones were 1,522 mg/L, 342 mg/L, and 609 mg/L. The maxima of COD, BOD5, and TOC concentrations in leachate from Cell A were 40,256 mg/L (Day 55), 22,200 mg/L (Day 55) and 10,200 mg/L (Day 90), respectively. In contrast, the maxima of COD, BOD5, and TOC concentrations in leachate from Cell B were 38,327 mg/L (Day 55), 19,400 mg/L (Day 55), and 10,050 mg/L (Day 124), respectively. The changes in the maxima and the final concentrations of COD, BOD5, and TOC in leachate from Cell A represent reductions of 96%, 98%, and 92%, respectively. In comparison, the changes in the maxima and the final concentrations of COD, BOD5, and TOC in leachate from Cell B represent reductions of 96%, 98%, and 94%, respectively.

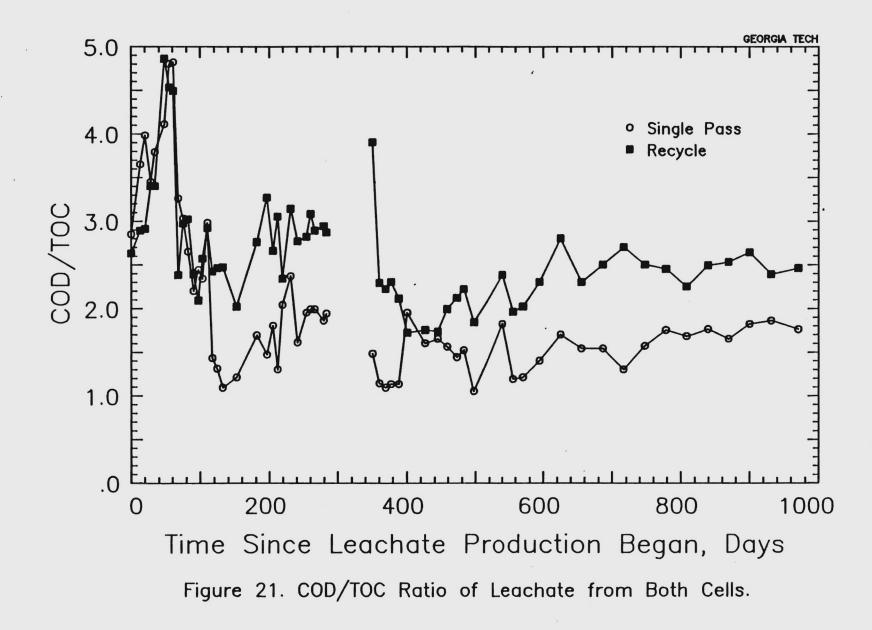
The delay in the occurrence of maxima of COD, BOD<sub>5</sub>, and TOC in leachate from Cell B as compared to Cell A can be attributed to the absence of washout of these parameters from Cell B due to recirculation of leachate.

The decline in COD, BOD5, and TOC concentrations in the leachate from Cell A between Days 55 and 139 was due to a combination of washout of the organic constituents and in situ treatment. During the same period, COD, BOD5, and TOC concentrations in the leachate from Cell B did not exhibit a similar decline since washout of the organic constituents was prevented by recycling leachate to the top of the refuse. In either case, concentrations were influenced by the relative dilution effects of water additions. Although gas production wasn't monitored because of the presence of gas leaks in the cells, the production of gas concomitantly with the decrease in the organic pollutant parameters is envisioned.

The TIC concentrations in the leachate samples from both cells are presented in Figure 19. As shown, the initiation of TIC concentrations in leachate in measurable levels occurred with an increase in the pH of leachate from acidic levels to neutral levels (Figure 25), a decrease in the volatile fatty acids in leachate (Figures 23 and 24) and a shift in the primary buffer system in the leachate from that of acetate to carbonate.

. Ratios of BOD<sub>5</sub>/COD and COD/TOC are presented in Figures 20 and 21, respectively. The BOD<sub>5</sub>/COD ratio is indicative of the biodegradability of the leachate; a decrease in this ratio indicates a decrease in biodegradability and consequently, a maturation of stabilization processes. The COD/TOC ratio is indicative of the oxidized state of the organic carbon that is available for biodegradability; a decrease in this



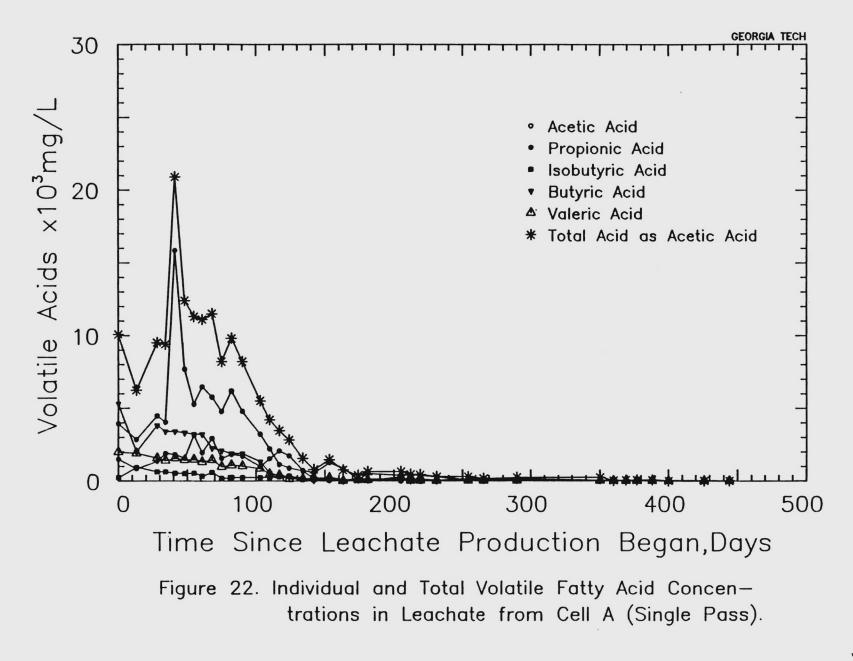


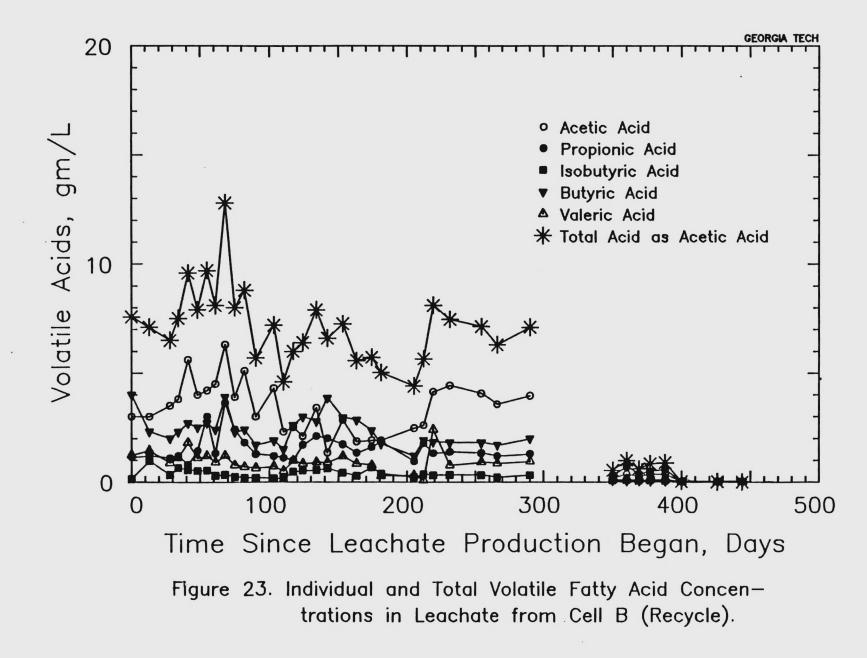
ratio represents a more oxidized state of organic carbon due to an increase in the presence of products of microbially-mediated processes which lead to the maturation of stabilization processes. As expected, these ratios decreased with the progress of landfill stabilization except from Day 205 to Day 290 when the BOD<sub>5</sub>/COD ratio increased for the leachate produced from Cell B; this increase was due to an increase in the total volatile acid concentration which resulted from increase in propionic acid concentration in the leac ate during the same period (Figure 23). The accumulation of propionic acid is attributable to the prevention of microbially-mediated degradation of propionic acid due to acidic conditions of the leachate.

# 4.4.2 Volatile Fatty Acids

In a landfill undergoing anaerobic stabilization, the concentration of volatile fatty acids in the leachate produced from the fill is one of the most important indicator parameters. Indeed, the stage and the progress of anaerobic conversion may be reflected by the concentration of these low molecular weight volatile fatty acids, which include acetic, propionic, isobutyric, butyric, and valeric acids.

The concentrations of individual and total volatile fatty acids in leachate from Cell A and Cell B are presented in Figures 22 and 23, respectively. As expected, the most abundant acid was acetic acid followed by butyric, propionic, valeric, and isobutyric acids. The maxima of total volatile fatty acid concentrations in leachate samples collected from Cell A and Cell B were 20,892 mg/L as acetic acid (Day 41) and 12,857 mg/L as acetic acid (Day 68), respectively. The total volatile fatty acid concentrations in leachate from Cell A and Cell B were undetectable beyond Day 426.





As mentioned earlier (Section 4.4), the causes of changes in the volatile fatty acid concentrations in leachate from Cell A and Cell B were different. After reaching a maximum of 20,892 mg/L as acetic acid on Day 41, the concentration of total volatile acid in leachate from Cell A steadily decreased due to washout and biodegradation; this decrease was accompanied with an increase in the pH (Figure 24). In comparison, during the same period (from Day 41 to Day 152), the leachate from Cell B did not exhibit a similar decrease in the pH and the total volatile acid concentration. Consequently, the frequency of leachate recycle was changed from once a day to once a week on Day 152 in an attempt to encourage the growth of methane formers by reducing its exposure to the high total volatile fatty acid concentrations and the low pH conditions prevailing in Cell B. This frequency of leachate recycle was maintained until Day 350. The frequency of leachate recycle was reduced to encourage the onset of methane formation phase since the acidic condition of the leachate was preventing the onset of the methane fermentation phase. During this period (Day 152 to Day 350), the pH of leachate increased to neutral pH levels and the total volatile acid concentration decreased concomitantly, thereby indicating a successful recovery of Cell B from acidic conditions. From Day 350 to Day 570 leachate was recycled once a day; thereafter, the frequency of leachate recycle was reduced to once a week, since most of the indicatory parameters (COD, BOD, TOC, pH, ORP, alkalinity, and conductivity) did not exhibit significant changes.

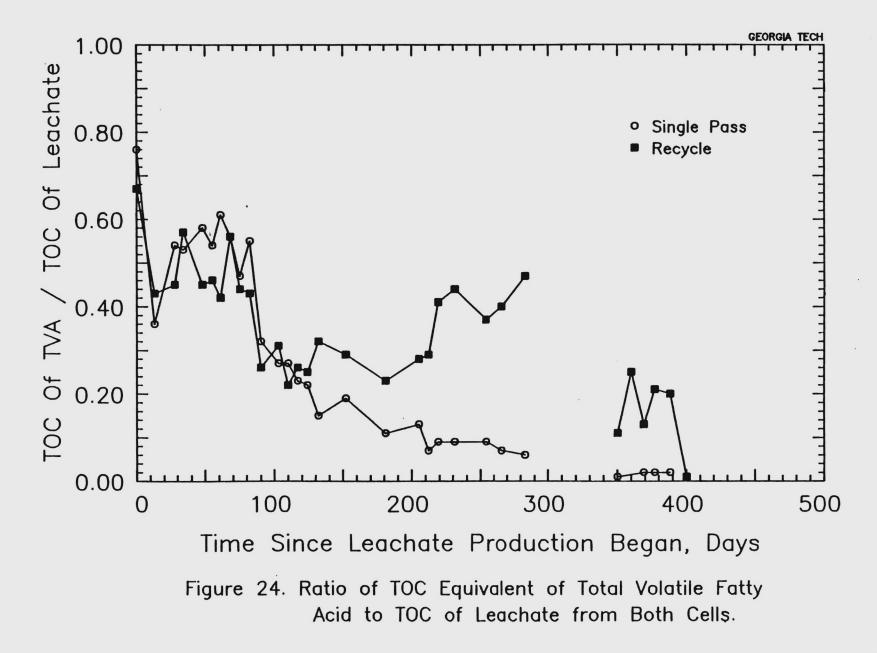
During the initial stages of landfill stabilization the organic carbon present in the total volatile acid constitutes a significant

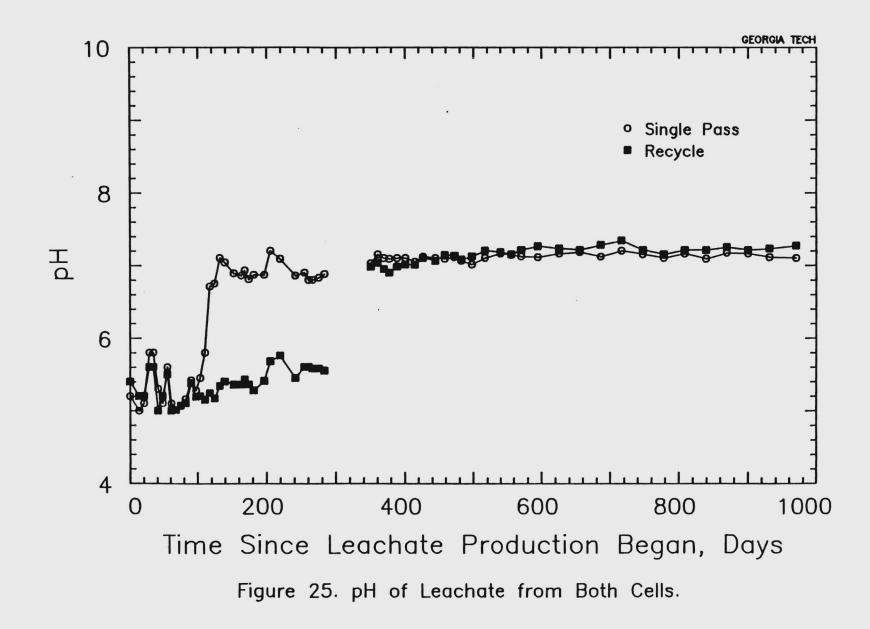
fraction of the TOC of leachate produced from the landfill. As stabilization progresses, the magnitude of this fraction decreases, due to the decrease in the total volatile fatty acid concentration. This decrease in the total volatile fatty acid concentration in the leachate is attributable to the landfill being in methane forming phase or final maturation phase, and a paucity of readily biodegradable organic compounds in the leachate. During the period when decreases in the volatile acid concentrations occurred, gas production couldn't be measured due to the presence of gas leaks in the liner. The production of gas, however, is expected with a decrease in the volatile acid concentrations.

The ratios of TOC equivalent of total fatty acid to the TOC of leachate from Cell A and Cell B, respectively, are presented in Figure 24. As shown, the decrease in this ratio corresponds with the decrease in the total volatile fatty acids (Figure 24) and an increase in the pH. 4.4.3 pH

The pH of the leachate produced from a landfill undergoing stabilization is a critical indicator parameter since it is an important determinant of the presence of volatile fatty acids and type and capacity of the predominant buffer system (viz., acetate or carbonate). Increases in the pH of leachate are usually accompanied by decreases in the volatile fatty acid concentrations.

The pH values of the leachate samples collected from Cell A and Cell B during this study are presented in Figure 25. The initial and the final pH values of the leachate from Cell A were 5.2 and 7.1, respectively. The pH values of the leachate from Cell A were within a





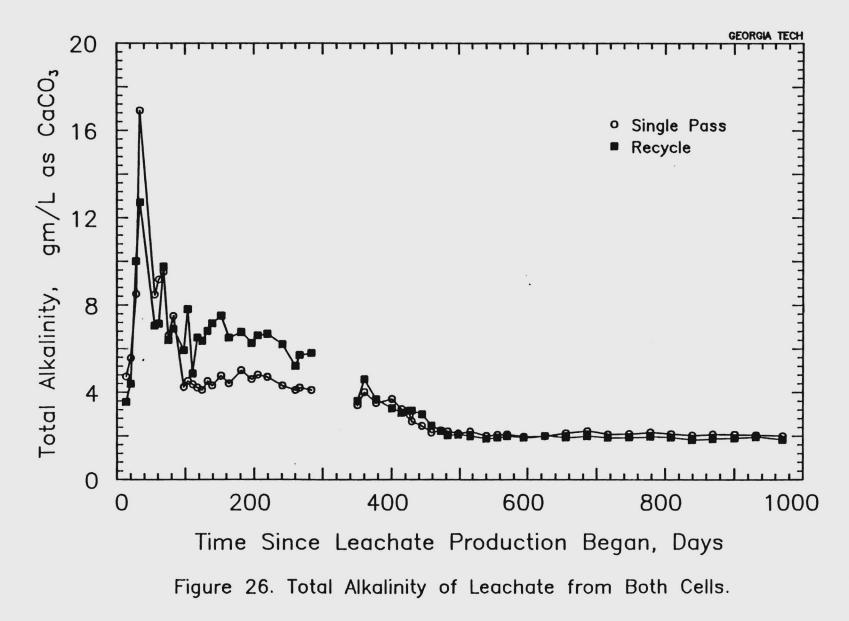
range 5.00 to 5.60 from Day 0 to Day 103, then increased to 7.14 in 36 days. This increase in pH was concomitant with a decrease in the total volatile fatty acid concentration and a shift in the buffer system from acetate to carbonate. Thereafter, the pH values of the leachate samples from Cell A were within a range 6.80 to 7.21.

The initial and the final pH values of the leachate from Cell B were 5.40 and 7.21, respectively. During the period of daily recycling of leachate (Day 0 to Day 152), the pH values were within a range 5.00 to 5.40. From Day 152 to Day 350, when weekly recycle of leachate was practised, the pH values were within a range 5.28 to 6.98; the increase in pH presumably due to the growth of methane formers within microenvironments of the landfill. This cause for an increase in the pH was further substantiated by a concomitant decrease in the total volatile acid concentration. From Day 350 to Day 970, the pH values were within a range 6.98 to 7.27. Similar to the behavior exhibited by the leachate from Cell A, the buffer system present in the leachate from Cell B shifted from acetate to carbonate as the pH increased from 5.00 to 7.27.

## 4.4.4 Alkalinity

The alkalinity of leachate is indicative of its buffer capacity. Also, alkalinity together with pH and total volatile fatty acid concentration indicate the predominant type of buffer system present in the leachate produced from a landfill.

The total alkalinity present in leachate samples collected from Cell A and Cell B during this investigation is presented in Figure 26. The total alkalinity present in leachate from Cell A increased from



4,717 mg/L as  $CaCO_3$  (Day 13) to 16,900 mg/L as  $CaCO_3$  (Day 34), then decreased to 1,990 mg/L as  $CaCO_3$  (Day 970). In comparison, the total alkalinity present in leachate from Cell B increased from 3,547 mg/L as  $CaCO_3$  (Day 13) to 12,700 mg/L as  $CaCO_3$  (Day 34), then decreased to 1,825 mg/L as  $CaCO_3$  (Day 970).

The increase in total alkalinity of leachate was due to increase in total volatile acid concentration since the volatile acids would add to the alkalinity pool as volatile acid salts. The decrease in volatile acid concentration would reduce the contribution of the volatile acids to the alkalinity pool. Thus, the trend exhibited by total alkalinity (Figure 25) of leachate closely follows the trend exhibited by volatile acids (Figures 22 and 23). An increase in pH (Figure 24) along with a decrease in volatile acid concentration and total alkalinity indicated a shift in the predominant buffer system from acetate to carbonate.

### 4.4.5 Conductivity

Conductivity of leachate is a measure of the presence of ionic species and along with total volatile fatty acid and alkalinity present in the leachate produced from a landfill undergoing stabilization, tends to have an inverse relationship with pH. Variations in conductivity result from microbially mediated processes, physicalchemical removal processes and dilution effects which occur within the leachate and the landfill environment.

The conductivity of a landfill leachate is indicative of its ionic strength. Since ionic strength of leachate inversely affects the activity coefficient of ions present in leachate, an increase in

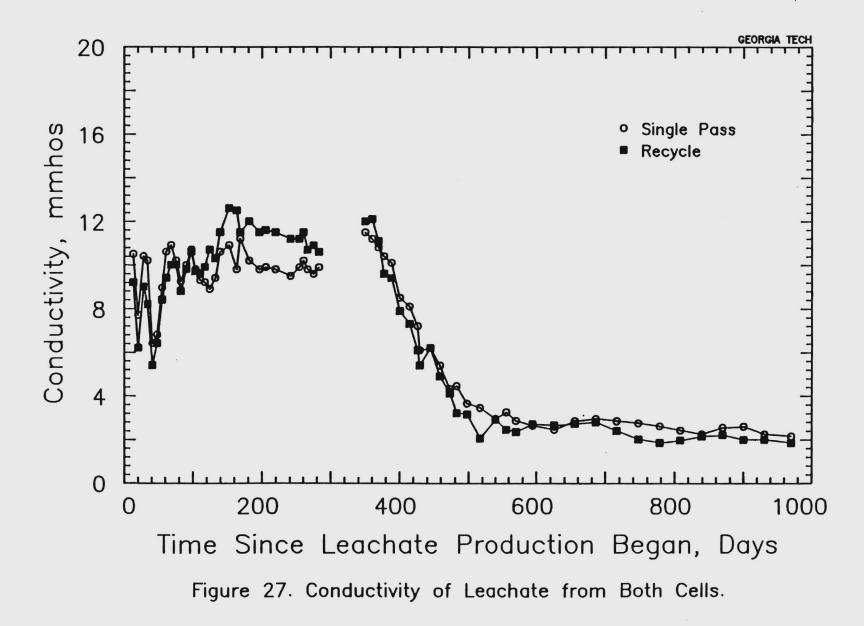
conductivity of leachate results in a decrease in the activity coefficient, thereby leading to an increase in the concentration of ions in leachate.

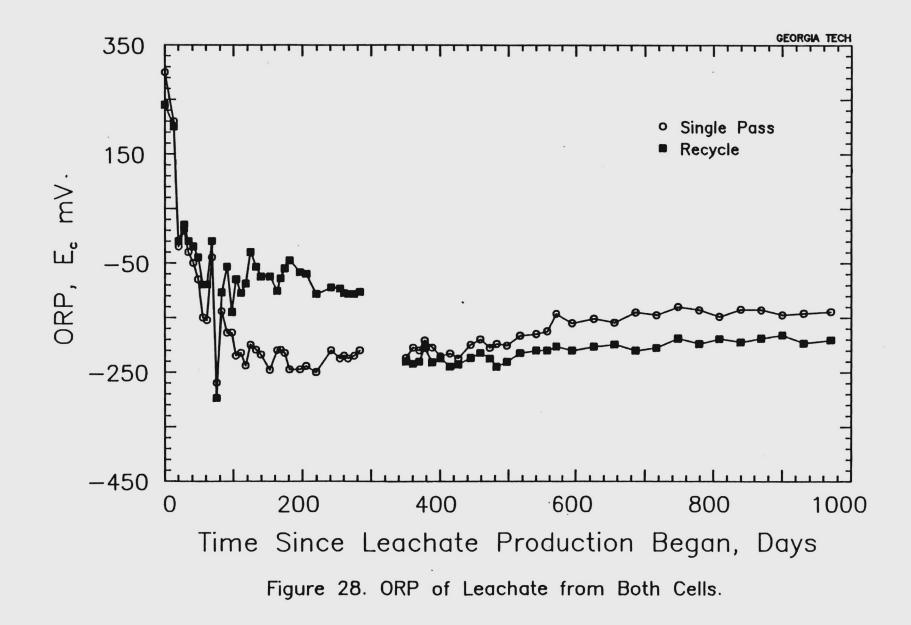
The conductivity of the leachate samples collected from Cell A and Cell B are shown in Figure 27. As shown, the conductivity of leachate from Cell A increased from 6,400 µmhos to 11,500 µmhos, then decreased to 2,150 µmhos, whereas the conductivity of leachate from Cell B increased from 5,400 mhos to 12,600 µmhos, then decreased to 1,855 µmhos.

The increases in the conductivity of leachate from Cell A and Cell B were due to leaching of organics and inorganics from the refuse and microbially mediated production of volatile acids. After reaching a maximum, the conductivity of leachate decreased due to a decrease in total volatile fatty acid concentrations, organic and inorganic salts, and metals, which were precipitated by sulfides. In addition, washout contributed to the decrease in conductivity of leachate from Cell A. 4.4.6 Oxidation-Reduction Potential (ORP)

The ORP of the leachate from a landfill is an indicator parameter which indicates the presence of oxidizing or reducing conditions within the leachate and the landfill environment. Except for the early phases of stabilization (Initial Adjustment Phase and Transition Phase), the ORP should reflect the presence of reducing conditions, particularly when methane fermentation is effective in converting the intermediate organic leachate constituents to more stabilized forms.

The ORP of leachate samples from Cell A and Cell B are presented in Figure 28. During the early phase of landfill stabilization (Day O





to Day 34) the ORP of leachate from both cells decreased from positive to negative values (Cell A: 300 mV  $E_c$  to -30 mV  $E_c$ ; Cell B: 240 mV  $E_c$ to -10 mV  $E_c$ ) which indicated the onset of reducing conditions within the leachates and the landfill environments. Thereafter, the ORP of the leachate from both cells remained negative till Day 970. ORP values of -400 mV to -500 mV that are optimum for methane fermentation were never recorded during the period of this investigation; this is probably due to inherent problems with the instrumental measurement of this parameter since the exposures of leachate, collected from a highly reducing environment, to atmosphere during measurement will record ORP values that are more positive than the actual ORP values.

Without new sources of oxidizing potential and entry of oxygen, the reducing environment established by microbially-mediated processes within a landfill will likely persist for an indefinite period of time. From Day 34 to Day 970 the ORP values reflected the existence of reducing conditions within both cells; these conditions along with increases in pH values and decreases in volatile acids induced sulfide formation and precipitation of heavy metals as subsequently discussed. 4.4.7 Sulfide

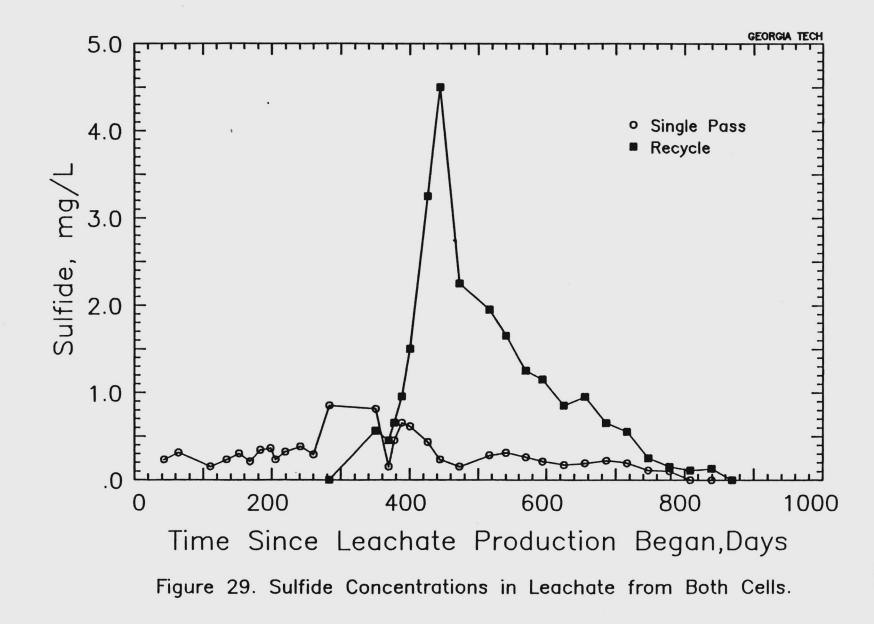
Sulfide is produced within a landfill environment during the Acid Formation and the Methane Fermentation Phases of stabilization as a result of microbial reduction of sulfate under reducing conditions by microorganisms which are strict anaerobes and use sulfate as an electron acceptor. The sulfide so produced precipitates most heavy metals present in the leachate as metal sulfides.

The total sulfide concentration in the leachate samples from

Cell A and Cell B are presented in Figure 29. The total sulfide concentration in the leachate from Cell A increased from 0.23 mg/L (Day 43) to 0.85 mg/L (Day 283), then decreased to 0.1 mg/L (Day 778), whereas the total sulfide concentration in the leachate from Cell B increased from 0.45 mg/L (Day 350) to 4.50 mg/L (day 444), then decreased to 0.13 mg/L (Day 839). The maximum sulfide concentrations in the leachate samples from Cell A and Cell B corresponded to ORP values of -210 mV  $E_c$  and -224 mV  $E_c$ , respectively.

In a sulfate-sulfide system, the reduction of sulfate to sulfide usually occurs around -500 mV  $E_c$ . Since ORP of leachate is a very sensitive parameter, it usually increases sharply as soon as the leachate is exposed to atmosphere (for measurement); thus, the "asmeasured" value will probably be higher than the actual value. The trends reported for leachate samples from both cells, however, would remain unchanged. Presence of sulfide in leachate samples was accompanied by precipitation of heavy metals (shown later in Figures 36-40), The maximum sulfide concentration (4.5 mg/L) in leachate from Cell B was higher than the maximum sulfide concentration (0.85 mg/L) in leachate from Cell A; this was attributed to the contrasting modes of management of leachate from Cell A (single pass) and Cell B (leachate recycle).

Sulfide exists in solution in three forms:  $H_2S$ ,  $HS^-$ , and  $S^{2-}$ . The presence of these forms is pH dependent with  $H_2S$  and  $HS^-$  being the dominant forms at acidic pH values (pH=5.0). However, with an increase in the pH of leachate from 5 to 7 as is evidenced in landfills, sulfide is also present in  $S^{2-}$  form along with  $H_2S$  and  $HS^-$ ; this is important,

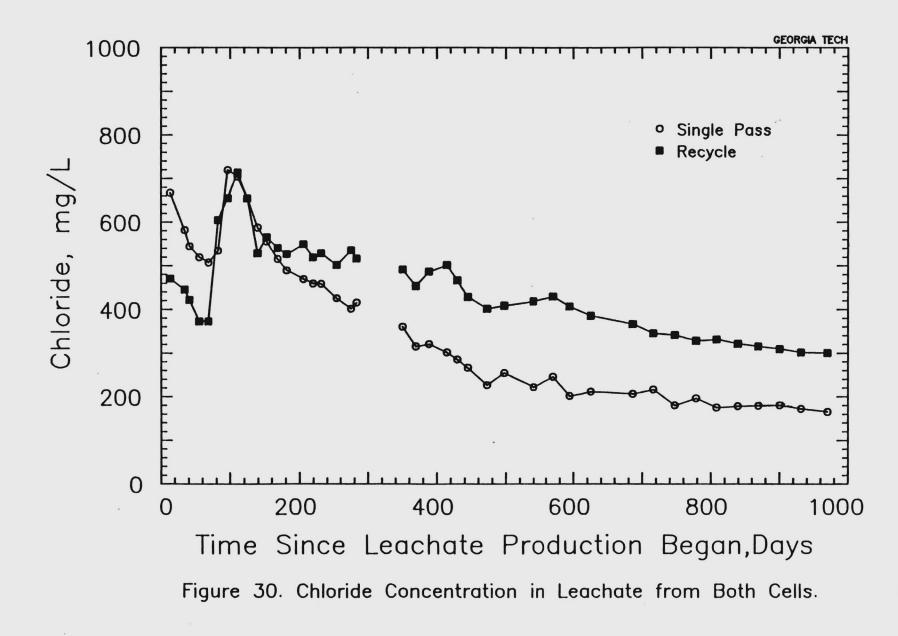


since  $S^{2-}$  is responsible for precipitation of heavy metals. 4.4.8 Chloride

In an anaerobic system such as a landfill undergoing stabilization, a conservative constituent is often utilized as a tracer to assess dilution, mixing, and washout effects. Chloride (Cl<sup>-</sup>) has been selected as a tracer, since it is essentially a non-reactive component with a high solubility in water. The chloride concentrations in the leachate samples from Cell A and Cell B are shown in Figure 30.

Chloride concentration in leachate from Cell A decreased from an initial value of 667 mg/L (Day 13) to 507 mg/L (Day 68), then increased to a maximum of 719 mg/L (Day 96); thereafter, the concentration decreased steadily to a final value of 165 mg/L (Day 970). In comparison, chloride concentration in leachate from Cell B decreased from an initial value of 470 mg/L (Day 13) to 372 mg/L (Days 55 and 68), then increased to a maximum of 713 mg/L (Day 110); thereafter, the concentration decreased steadily to a final value of 300 mg/L (Day 970).

The initial decline in the chloride concentration in leachate from both cells was due to dilution caused by continual water additions in rainfall-equivalent volumes to the cells through the distribution networks placed over the refuse in each cell to simulate incidental rainfall. The increase in concentrations can be ascribed to the leaching of chloride from the refuse. After reaching a maximum, the chloride concentration in leachate from both cells declined steadily, and as expected, chloride concentration in leachate from Cell A declined more than the chloride concentration in leachate from Cell B.



This behavior can be ascribed to washout resulting from single pass operation.

A correction factor, based on chloride concentrations, was utilized to assess the changes in constituent levels in leachate samples from both cells. The correction factor (C.F.) is defined as follows:

$$C.F. = \frac{[C1]_{t}}{[C1]_{max}}$$
(3)

where;

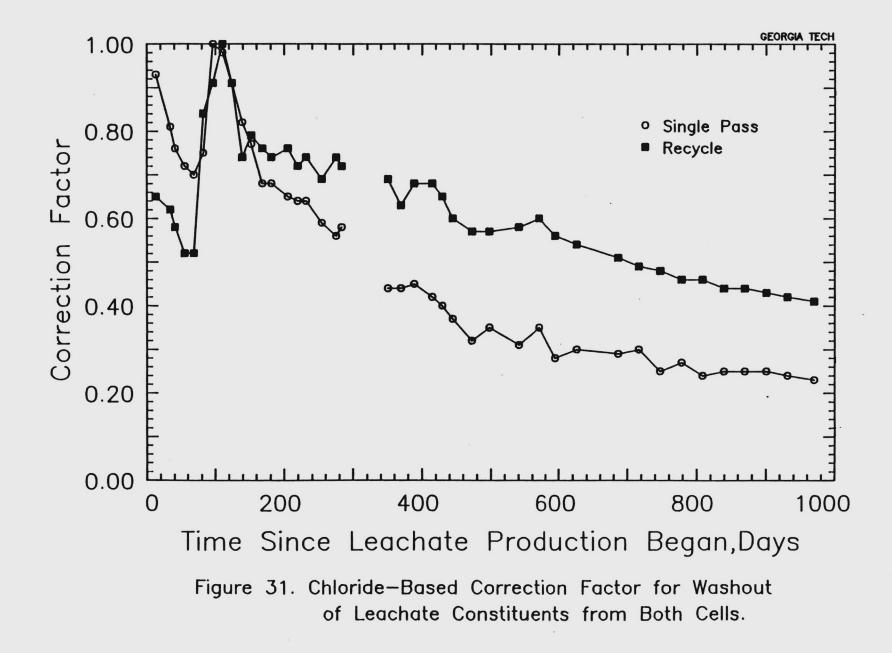
C.F. = concentration factor

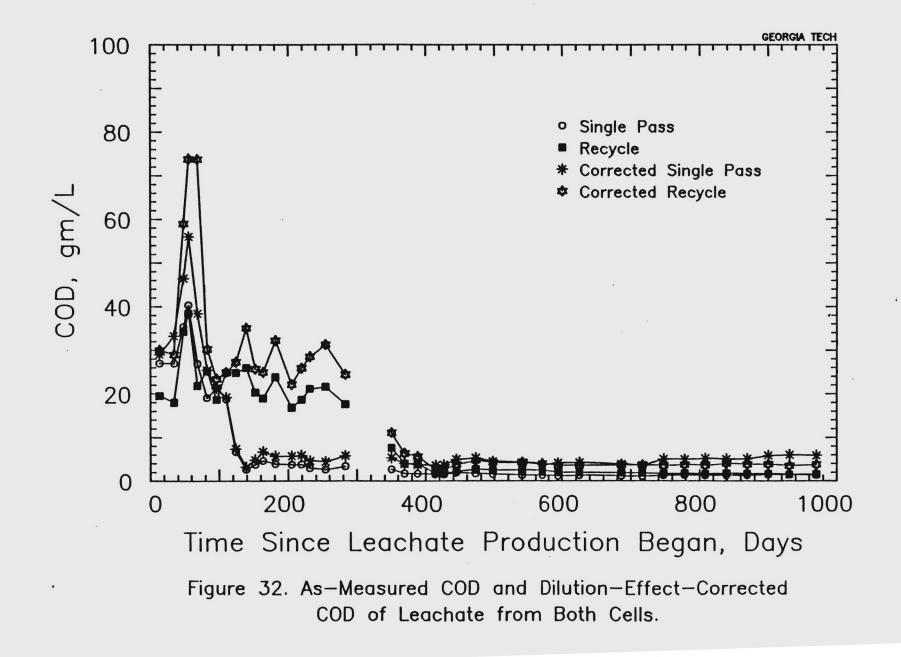
 $[Cl^-]_t$  = chloride concentration at any time t, mg/L

[C1<sup>-</sup>]<sub>max</sub> = maximum chloride concentration, mg/L

The correction factors for leachate samples collected from both cells are shown in Figure 31.

The correction factor for leachate samples from Cell A decreased from 1.00 (Day 96) to 0.23 (Day 970), whereas that from Cell B decreased from 1.00 (Day 110) to 0.41 (Day 970). To further illustrate the washout effect, the "As-Measured" COD and the "Corrected" COD values of leachate samples from both cells are presented in Figure 32. Based on the maximum "Corrected" COD values of leachate samples from Cell A and Cell B, the reductions in COD by Day 970 were 89% and 95%, respectively. In comparison, the reduction based on maximum "As-Measured" COD values of leachate from both cells was 96%. Thus, the difference in reductions in COD values (89% vs. 96%) of leachate samples from Cell A can be attributed to washout.





#### 4.4.9 Ionic Strength and Activity

Ionic strength (I) indicates the intensity of the electric field in the leachate from a landfill. The ionic strength variations are caused by microbially-mediated processes, physical-chemical removal processes, and dilution effects within the leachate and the landfill environment.

The ionic strength (I) of leachate from Cell A and Cell B was computed using the following equation:

$$I = 1.6 \times 10^{-5} \times C$$
 (4)

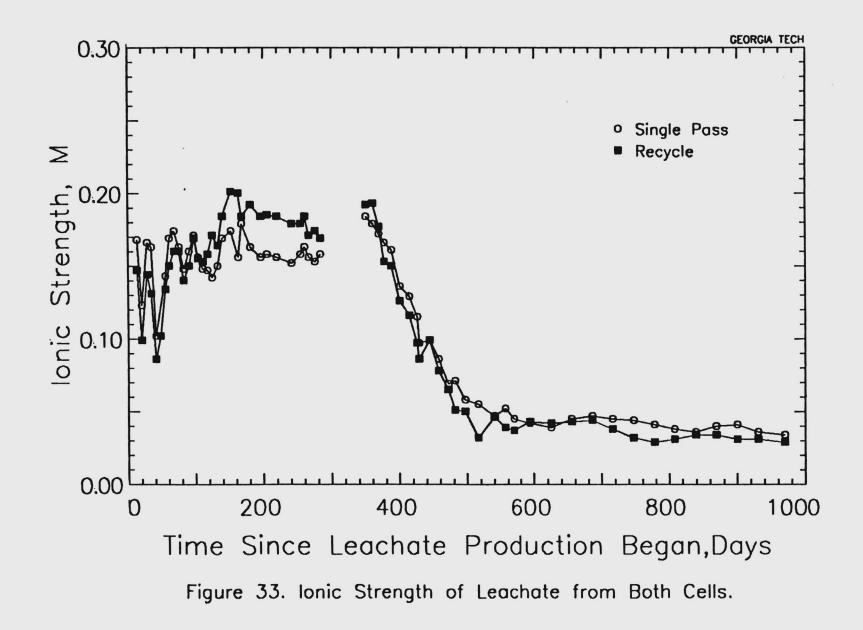
where:

I = Ionic Strength, M

 $C = Conductivity, \mu mhos$ 

Equation 4 was reported by Lind (110) and Russell (111). The ionic strength of leachate from Cell A decreased from 0.168 M (Day 13) to 0.102 M (Day 41), then increased to 0.174 M (Day 68); between Days 68 and 350, the ionic strength fluctuated between 0.142 M and 0.184 M. After Day 350, the ionic strength declined steadily to 0.034 M (Day 970). In comparison, the ionic strength of leachate from Cell B decreased from 0.147 M (Day 13) to 0.086 M (Day 41), then increased to 0.201 M (Day 152). After Day 360, the ionic strength declined steadily to 0.029 M (Day 970; Figure 33).

The initial decline in the ionic strength of leachate from both cells can be ascribed to dilution. Reducing conditions coupled with enhanced biological conditions within the leachate and the landfill environment caused the production of ionic species such as sulfides and



ionized volatile fatty acids as the pH of leachate increased to neutral levels from acidic levels. The reductions in ionic strength and conductivity were concomitant with removal of ionic species such as sulfides and heavy metals primarily by precipitation with heavy metals (Figures 29, 36-40).

The activity coefficient can be computed using any of the four following approximations: Debye-Huckel Approximation, Extended Debye-Huckel Approximation, Guntelberg Approximation, and Davies Approximation (112).

Debye-Huckel Approximation (Ionic Strength < 0.5 x  $10^{-3}$  M)

$$\log_{1} = -0.51 Z_{1} I^{1/2}$$
(5)

where:

i = activity coefficient of species i

 $Z_i$  = charge on species i

I = ionic strength

Extended Debye-Huckel Approximation (Ionic Strength <0.1M)

$$\log Y_{1} = -\frac{A Z_{1}^{2} I^{\frac{1}{2}}}{1 + Ba_{1} I^{\frac{1}{2}}}$$
(6)

where;

i = activity coefficient of species i  $Z_i$  = charge on species i I = ionic strength  $A = 0.509 \text{ mol}^{-1/2} \text{ kg}^{1/2}$ , at 25°C for water  $B = 3.291 \text{ x } 10^9 \text{ m}^{-1} \text{ mol}^{-1/2} \text{ kg}^{1/2}$ , at 25°C for water  $a_i$  = effective diameter of the ion, m Guntelberg Approximation (Ionic Strength < 0.1M)

$$\log \tilde{\gamma}_{i} = -\frac{0.5 \, z_{i} \, I^{\frac{1}{2}}}{1 + I^{\frac{1}{2}}} \tag{7}$$

where:

i = activity coefficient of species i

 $Z_i$  = charge on species i

I = ionic strength

Davies Approximation (Ionic Strength < 0.5m)

$$\log \gamma_{i} = -0.51 Z_{i}^{2} \left( \frac{I^{\frac{1}{2}}}{1+I^{\frac{1}{2}}} - 0.3I \right)$$
(8)

where;

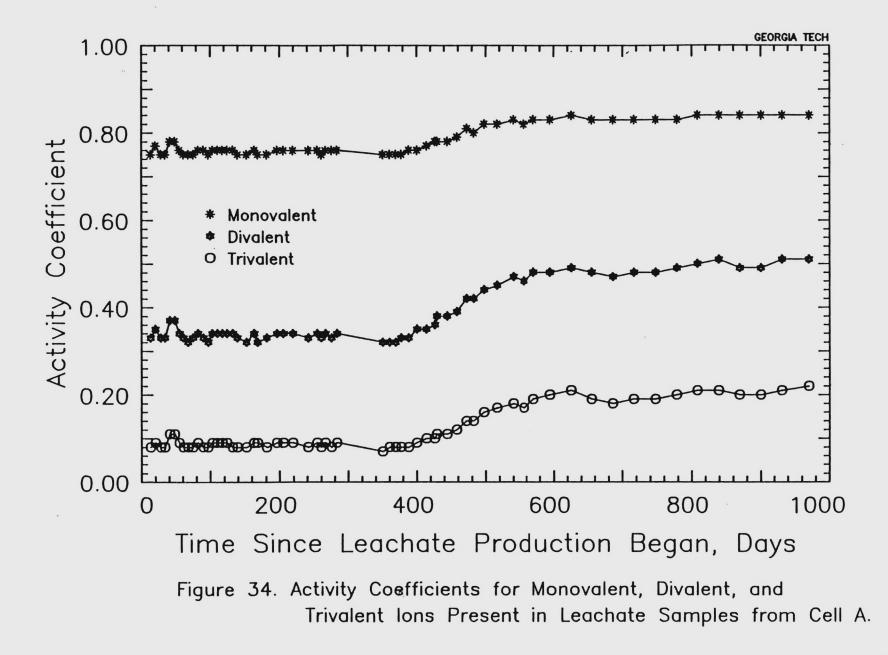
i = activity coefficient for species i

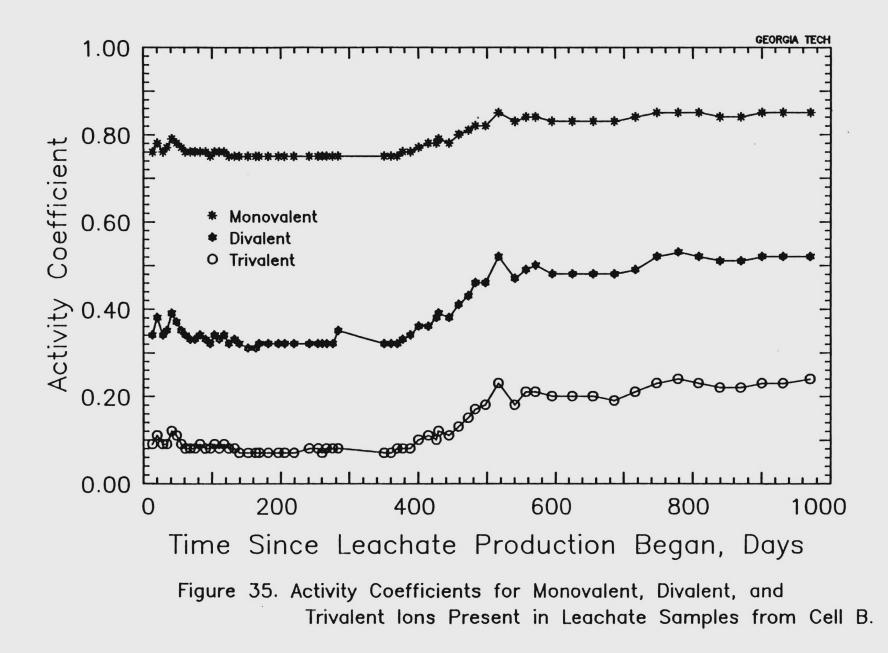
 $Z_i$  = charge on species i

I = ionic strength

Davies Approximation was used to compute the activity coefficient of leachate samples collected from Cell A and Cell B, since the ionic strength values were greater than 0.1 M from Day 13 to Day 414, except on Days 20 and 41, when the ionic strength of leachate samples collected from Cell B was less than 0.1 M. The activity coefficients for monovalent, divalent, and trivalent ions present in leachate samples from Cell A and Cell B are shown in Figures 34 and 35, respectively.

An increase in the activity coefficients of monovalent, divalent, and trivalent ions was accompained by a decrease in ionic strength, conductivity, and alkalinity. The activity coefficients of





monovalent, divalent, and trivalent ions were between 0.75 and 0.85, 0.33 and 0.53, and 0.08 and 0.24, respectively. These values are in agreement with the values reported by Chang (21) and Pohland (23).

The activity coefficient of an ion affects the presence of that ion in the leachate produced from a landfill. In general, lower activity coefficients result in higher levels of ions in leachate. For example, the solubility of cobalt sulfide can be described as;

$$\{C_0^{2^+}\} \{S^{2^-}\} = K_{so}$$
(9)

assuming an activity coefficient of 0.5 for divalent ions Equation 9 will be modified to;

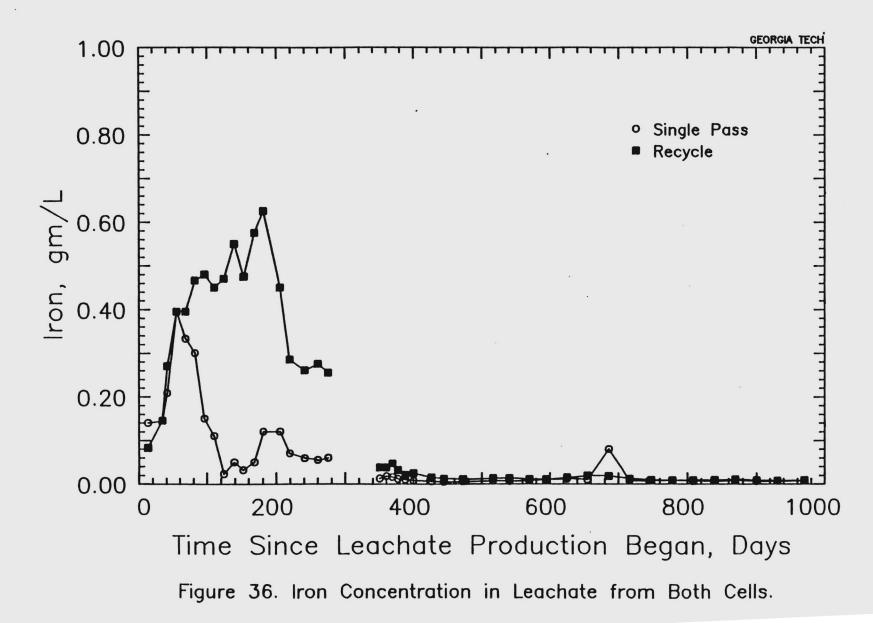
$$0.5 [Co^{2+}]0.5[S^{2-}]=K_{so}$$
 (10)

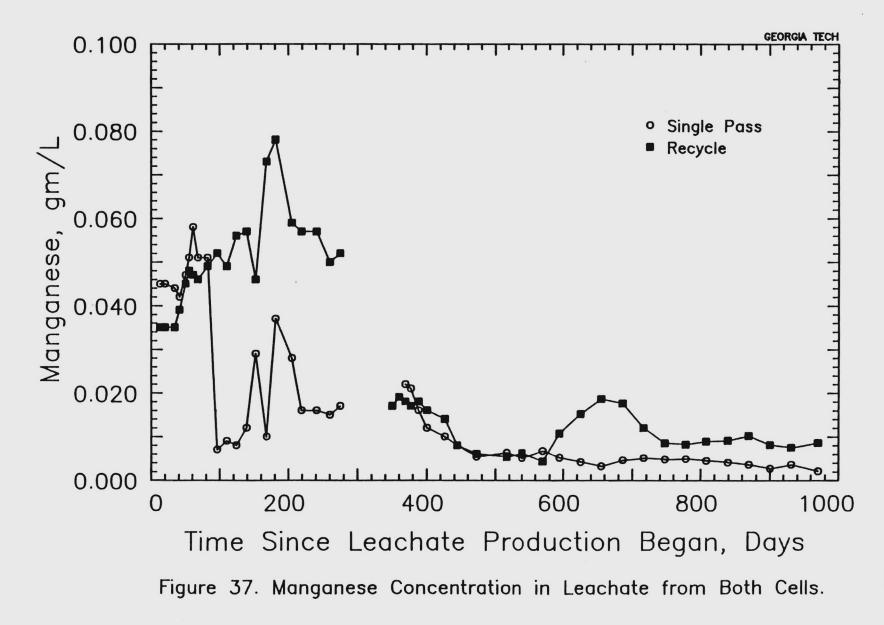
$$[Co^{2^{+}}][S^{2^{-}}]=4K_{so}$$
(11)

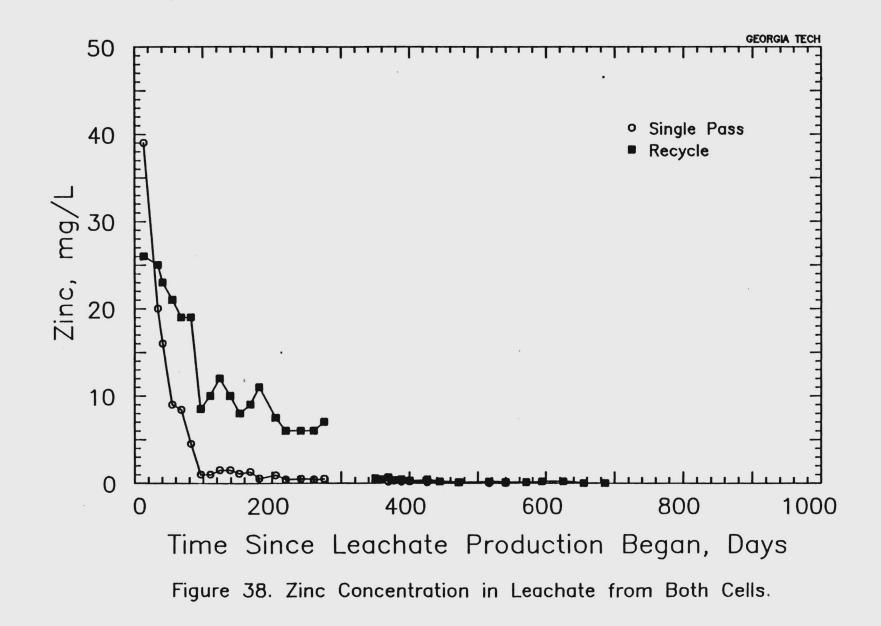
Since  $K_{SO}$  does not change, the product of cobalt and sulfide concentrations in leachate increases by a factor of four; this results in higher cobalt and sulfide concentrations in leachate at equilibrium ' than the concentrations predicted for dilute solutions.

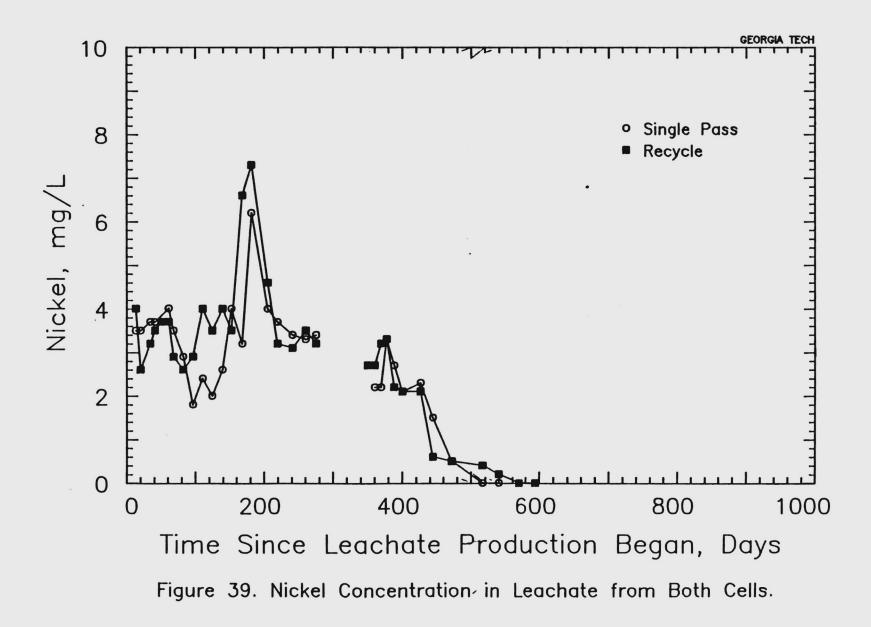
## 4.4.10 Iron, Manganese, Zinc, Nickel, Cobalt, and Strontium

Iron, manganese, zinc, nickel, cobalt and strontium concentrations in leachate samples collected from Cell A and Cell B are presented in Figures 36 through 40. Iron concentration in leachate from Cell A increased from 140 mg/L (Day 13) to 395 mg/L (Day 55), then decreased to an ultimate value of 8.1 mg/L (Day 970), whereas iron concentration in leachate from Cell B increased from 83 mg/L (Day 13) to 625 mg/L









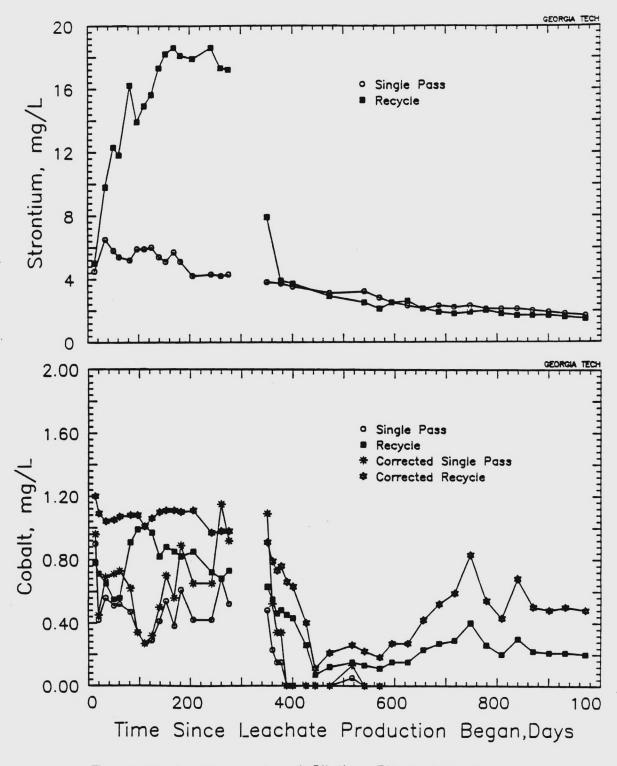


Figure 40. As-Measured and Dilution-Effect-Corrected Cobalt Concentrations and Strontium Concentrations in Leachate from Both Cells.

(Day 152), then decreased to an ultimate value of 8.6 mg/L (Day 970). Manganese concentration in leachate from Cell A increased from 45 mg/L (Day 13) to 51 mg/L (Days 55, 68, and 82), then decreased to an ultimate value of 2.1 mg/L (Day 970), whereas manganese concentration in leachate from Cell B increased from 35 mg/L (Day 13) to 79 mg/L (Day 181), then decreased to an ultimate value of 2 mg/L (Day 970). Zinc concentrations in leachate samples from both cells decreased steadily from 39 mg/L (Day 13; Cell A) to 0.1 mg/L (Day 472; Cell A) and 26 mg/L (Day 13, Cell B) to 0.2 mg/L (Day 625; Cell B). Nickel concentrations in leachate from Cell A increased from 3.5mg/L (Day 13) to 6.2mg/L (Day 181), then decreased to an ultimate value of 0.5 mg/L (Day 472), whereas nickel concentration in leachate from Cell B increased from 4.0 mg/L (Day 13) to 7.3 mg/L (Day 181), then decreased to an ultimate value of 0.2 mg/L (Day 540). Cobalt concentration in leachate from Cell A fluctuated between 0.27 mg/L (Day 110) and 0.90 mg/L (Day 13) till Day 350, then decreased sharply to 0.15 mg/L by Day 377. In contrast, cobalt concentration in leachate from Cell B increased from an initial value of 0.78 mg/L (Day 13) to 1.01 mg/L (Day 110), then decreased to 0.07 mg/L (Day 444), and finally increased to an ultimate value of 0.20 mg/L (Day 970). The strontium concentration in leachate from Cell A varied from 4.5 mg/L (Day 13) to 1.7 mg/L (Day 970), with a high of 6.0 mg/L (Day 124), whereas in leachate from Cell B, the strontium concentration varied from 5.0 mg/L (Day 13) to 1.5 mg/L (day 970), with a high of 18.6 mg/L (Days 168 and 241).

The increase in concentrations of iron and manganese in leachate from both cells during the transition and the acid formation phases of landfill stabilization was primarily due to acidic pH of the leachate; the concentrations decreased with an increase in pH from acidic to neutral levels together with the appearance of sulfide (Figure 29) and a shift of buffer system from acetate to carbonate in leachate from both cells, thereby indicating precipitation of these metals as sulfides and carbonates. In addition to precipitation, washout also contributed to the decrease in concentrations of iron and manganese in leachate samples from Cell A.

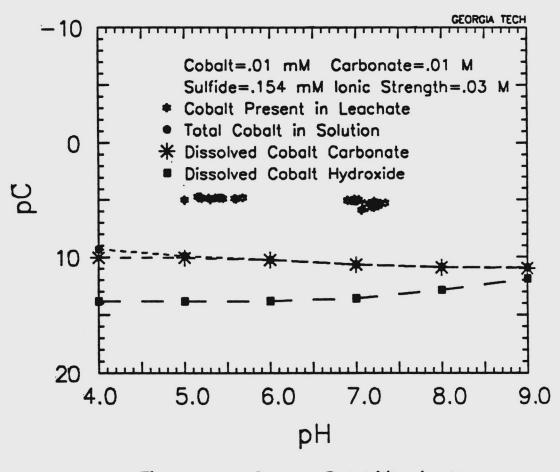
Zinc and nickel behaved similarly to iron. Thus, the primary removal mechanism for heavy metals was precipitation; this is in agreement with the evidence reported by other researchers (21,25,59 and 109).

The "as-measured" and the "dilution-effect-corrected" concentrations of stable cobalt in leachate from both cells are presented in Figure 40. As expected, cobalt concentrations in leachate increased during the transition and the acid formation phases of landfill stabilization due to acidic pH of the leachate; the concentrations then decreased with an increase in pH from acidic to neutral levels together with the appearance of sulfide (Figure 29) and a shift in the primary buffer system from acetate to carbonate in leachate from both cells. While precipitation of cobalt by the potential-precipitant species such as, carbonates and sulfides was the major removal mechanism for removal of cobalt from leachate, the "asmeasured" concentrations of cobalt in leachate were higher than those predicted by solubility.

As shown in Figure 40, the strontium concentrations in leachate from Cell A and Cell B increased during the transition and the acid

formation phases of landfill stabilization due to acidic pH of the leachate. The trend, however, was more pronounced in the strontium concentration in leachate from Cell B compared to the strontium concentration in leachate from Cell A. This behavior can be attributed to the prolonged acidic pH of leachate from Cell B as compared to leachate from Cell A, and prevention of washout of constituents present in leachate from Cell B due to recirculation of leachate. With the shift of the primary buffer system from acetate to carbonate along with an increase in pH of leachate from acidic levels to neutral levels, and appearance of TIC concentrations in leachate, strontium concentrations in leachate from both cells decreased. Based on the solubility product of strontium carbonate (log K = 9.0), strontium concentration in a leachate (ionic strength = 0.03 M, Figure 33, activity coefficients for divalent ions = 0.5, Figure 35) will be 5.5 mg/L. This value is comparable to the residual strontium concentrations (2 mg/L) in leachate. Thus, the decrease in strontium concentration, albeit more pronounced in leachate from Cell B, is attributed to the probable precipitation of strontium as strontium carbonate.

The solubility of cobalt as a function of hydroxide, carbonate, and sulfide is presented in Figure 41; sulfide is the strongest precipitant for cobalt, followed by carbonate and hydroxide. The "as-measured" cobalt concentrations in leachate from Cell B are also shown in Figure 41, these concentrations are divided into two pH ranges: one group with pCo varying from 4.7 to 5.0 at pH values ranging from 5.00 to 5.60 and the other group with pCo varying from 5.0 to 5.9 at pH values ranging from 6.90 to 7.34.



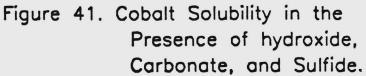


Figure 41 indicates that while sulfide and carbonate (shift of the primary buffer system from acetate to carbonate due to increase in pH values of leachate from acidic levels to neutral levels and reduction in the concentrations of volatile acids to below detection limits) precipitated cobalt, cobalt concentrations were present in excess of the concentrations predicted by solubility. This can be attributed to the probable complexation of cobalt by humic substances present in leachate.

#### 4.4.11 Radionuclides

The results of the radionuclide analyses performed on the leachate samples collected from Cell A and Cell B are summarized in Table 16. The causes for the decrease in the radionuclide levels in leachate from Cell A and Cell B were different; while washout contributed to the decrease in the radionuclide levels in Cell A, the radionuclides in leachate from Cell B were contained within the landfill system (lined landfill with cover, leachate collection, recirculation, distribution networks, and leachate collection sumps) and allowed to decay.

The maximum levels of Ce-141 present in leachate from both Cell A and Cell B, during the period of investigation, were less than 0.22 nCi/L. Based on Day O concentrations of Ce-141 in Cell A and Cell B, less than 0.03 percent of the Ce-141 spiked on the refuse were present in the leachate from both cells. Thus, greater than 99.97 percent of the Ce-141 spiked on the refuse were retained within the cells. The results of chemical modeling on speciation of metals (MINEQL) indicate

Time Since Leachate Production	Radionuclide Concentration Cell A Cell B					
	Ce-141	Sr-85	Co-58	Ce-141	Cell B	0
Began,	C6-141	31-05	C0-50	CE-141	Sr-85	Co-50
Days	nCi/L	nC1/L	nC1/L	-01/1	nC1/L	-01/1
vajo	101/1	101/6	IIC1/L	nC1/L	nc1/L	nC1/L
0	<0.22	4.35	4.00	<0.22	3.60	3.50
13	<0.21	4.37	3.68	<0.24	6.37	5.84
20	<0.22	6.90	5.79	<0.21	6.42	5.78
27	<0.21	4.94	3.83	<0.33	8.36	6.70
34	<0.34	6.10	4.56	(0.39	9.72	7.68
41	<0.27	5.41	3.77	(0.43	9.46	6.99
48	<0.31	5.02	2.75	(0.44	10.8	8.23
55	<0.32	5.36	2.70	<0.47	11.7	8.83
62	<0.37	5.22	1.98	<0.64	11.3	7.96
69	<0.60	5.48	2.10	<0.77	12.7	9.02
76	<0.45	5.20	1.70	<0.74	15.6	10.40
83	<0.53	5.64	1.67	<1.1	16.2	10.80
90	<0.62	5.77	1.73	<1.2	15.2	8.97
97	<1.1	6.58	2.14	(1.2	14.0	8.45
103	<1.3	6.54	1.66	(1.6	17.6	9.83
110	<1.6	6.43	1.29	<2.0	16.3	9.69
117	<1.9	6.23	0.40	(2.4	17.3	9.48
124	<2.2	5.90	0.22	(2.5	17.4	10.3
132	<1.3	5.52	0.32	(2.1	16.8	9.07
139	(1.5	5.99	0.30	(2.5	18.9	10.8
153	<4.1	1.73	0.30	<3.8	20.6	9.66
160	<4.4	5.07	0.25	(3.5	19.5	9.89
168	<5.1	5.35	0.15	(4.2	20.3	10.2
174	<6.3	4.71	0.15	(5.2	20.8	10.5
181	<7.1	5.31	0.16	<5.4	21.0	10.4
190	<7.0	4.72	0.50	<8.4	20.4	11.2
196	<8.6	2.88	0.16	<9.6	18.0	9.88
205	<4.9	4.34	0.30	<7.6	20.4	9.58
220	<8.1	5.29	0.25	<12.0	22.1	11.0
231	<26.0	2.00	0.20	<29.0	21.6	8.36
241	<26.0	4.70	0.20	<17.0	22.9	10.4
283	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
350	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

# Table 16. Radionuclide Concentrations, nCi/L, (on July 2, 1982) in the Leachate Samples from Cell A (Single Pass) and Cell B (Recycle)

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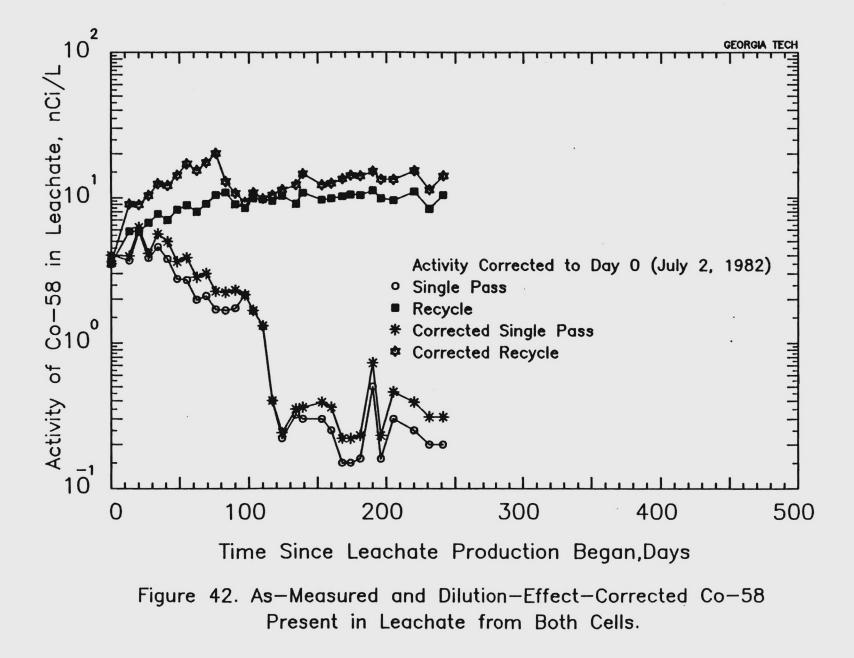
\*None Detectable

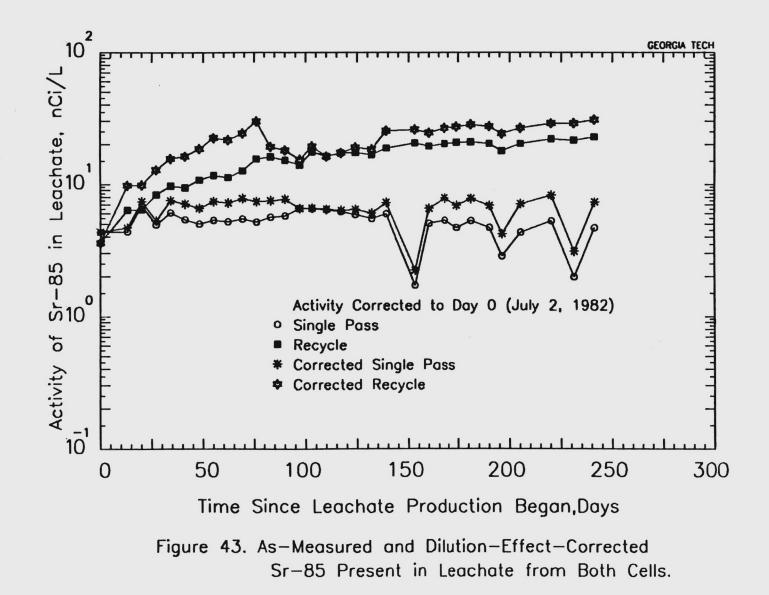
Notes:

Volume of Water in Cell A = 5,475 L Volume of Water in Cell B = 5,785 L Radionuclide Concentration in Cells on 2 July 1982 - Cell A: Co-58 = 165 nCi/L, Sr-85 = 200 nCi/L, and Ce-141 = 1,055 nCi/L - Cell B: Co-58 = 157 nCi/L, Sr-85 = 190 nCi/L, and Ce-141 = 1,000 nCi/L

that Ce-141 is expected to form soluble complexes with acetic acid in the presence of acetic acid, carbonate, sulfide, and hydroxide, at pH =5.00, and exist as Ce<sup>+3</sup> in the presence of carbonate, sulfide, and hydroxide, at pH = 7.00. This model (MINEQL), however, does not account for physical-chemical separation, viz., adsorption and ionexchange. Since less than 0.03 percent of the Ce-141 spiked on the refuse was present in the leachate from both cells, the remaining ( 99.97%) Ce-141 was probably adsorbed on the refuse placed in the cells.

The as-measured and the dilution-effect-corrected concentrations of Co-58 and Sr-85 in leachate from both cells are presented in Figures 42 and 43, respectively. In comparison with Chang's findings on codisposal of low-level radioactive waste within landfills (21), the concentrations of Co-58 and Sr-85 in leachate from the cell with leachate recycle (Cell B), behaved similarly. The different behavior of Co-58 and Sr-85 in leachate from single-pass cell (Cell A), when compared to the results reported by Chang (21), is attributed to the variation in simulations of infiltration of moisture due to incidental rainfall; Change (21) added one liter of water per week to the lysimeters, to simulate infiltration of moisture due to incidental rainfall (one liter per week was an average rainfall-equivalent volume over a three year period), throughout the period of investigation, whereas, in this study, actual rainfall-equivalent volumes, based on incidental rainfall in Atlanta, GA during the period of investigation, were added to the cells, Moreover, the additions of rainfallequivalent volumes were terminated on Day 56 since a 0.3-m (1 ft) head





of leachate had accumulated on the bottom of the cell; this 0.3-m value was computed by assuming the porosity of refuse as 0.4. In Chang's study, the constituents present in the refuse placed in the single-pass cell leached continually due to continual addition of water to the cell, whereas, in this study, the leaching of constituents from the refuse, due to addition of rainfall equivalent volumes, ceased after Day 56.

Based on the initial spiked levels of Co-58 and Sr-85, the maximum levels of Co-58 and Sr-85 in leachate from Cell B were 12.7 percent and 15.7 percent, respectively. The radionuclides in leachate from both Cell A and Cell B decayed to below detectable limits by Day 350.

The specific activities of Co-58 and Sr-85 in leachate from both cells are presented in Table 17. The decrease in specific activity of Co-58 in leachate from Cell A is attributable to the inconsistent leaching patterns of Co-58 and stable cobalt due to termination of addition of rainfall-equivalent volumes to the cell. In leachate from Cell B, the specific activities of Co-58 and Sr-85 were 11.21 nCi/mg  $\pm$ 2.14 nCi/mg and 1.07 nCi/mg  $\pm$  0.009 nCi/mg, respectively. The coefficient of variation for the specific activities of Co-58 and Sr-85 in leachate from Cell B were 19.1% and 8.4%, respectively. The coefficient of variations for Co-58 and Sr-85 indicate that trends exhibited by cobalt and strontium in leachate from Cell B can be used to simulate the behavior of Co-58 and Sr-85 within Cell B.

One of the objectives of this study was to investigate the potential solubilization of precipitated LLRW by humic substances

Time Since		Specifi	c Activity	
Leachate	Co	-58		-85
Production	Cell A	Cell B	Cell A	Cell B
Began,				
Days	nCi/mg	nCi/mg	nCi/mg	nCi/mg
13	4.0	7.4	1.0	1.3
20	13.7	8.1	-	-
34	8.1	11.8	0.9	1.0
50	-	-	0.9	-
61	3.8	14.2	1.0	0.8
82	3.6	11.8	1.1	1.0
96	6.3	8.5	1.1	1.1
110	4.8	9.7	1.0	1.1
124	0.8	10.6	0.9	1.1
139	0.8	13.2	1.2	1.1
152	0.6	11.0	0.3	1.2
168	0.4	12.0	1.0	1.0
181	0.2	12.6	1.1	1.1
205	0.7	11.3	1.0	1.2
241	0.5	14.4	1.0	1.3

Table 17. Specific Activities of Co-58 and Sr-85 in Leachate from Cell A (Single Pass) and Cell B (Leachate Recycle)

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present in the landfill leachate during the later phases of landfill stabilization (methane formation phase and final maturation phase). Since the radionuclides chosen to simulate LLRW had decayed to levels below the detectable limits by Day 350, a solubilization study was simulated, wherein Co-58 was chosen as the model radionuclide and its solubilization from cobalt sulfide (previously precipitated to simulate precipitation of LLRW by sulfide) by leachate from Cell B (during the later phases of stabilization) was investigated. Co-58 was chosen, since Chang (21) had reported an increase in Co-58 in the leachate from a lysimeter (equipped with leachate recycle) during the later phases of landfill stabilization. The results of the solubilization study are presented in the following section.

### 4.5 Solubilization Study

The results of the study on the solubilization of Co-58, from cobalt sulfide (containing both Co-58 and stable cobalt) by model humic acid (A-2HA) and leachate samples from Cell B are presented in the following sections. Also presented in the following sections are the results of the Quality Assurance/Quality Control (QA/QC) study, wherein cobalt sulfide and a swamp based humic acid (A-2HA) were utilized as model compounds.

#### 4.5.1 Fractionation of Model Humic Acid and Leachate

Ultrafiltration (UF) and gel permeation chromatography (GPC) were utilized to fractionate model humic acid (A-2HA) and leachate samples into different molecular-weight fractions. The results of UF and GPC fractionations are summarized in Tables 18 and 19, respectively.

As shown in Table 18, model humic acid (A-2HA) at three different pH values (5.5, 7.0, and 8.5) and three leachate samples were fractionated into permeate (low-molecular-weight) and retentate (highmolecular-weight) fractions. The percent local membrane rejection, R, reported for each sample was computed using the following expression;

$$R = 1 - \frac{\ln (1 - \frac{V_p C_{pc}}{V_f C_f})}{\ln (1 - \frac{V_p}{V_f})}$$
(9)

where;

- R = local membrane rejection, \$
- Vp = permeate volume, mL
- $V_{f}$  = feed volume, mL

 $C_{DC}$  = permeate concentration (composite), mg/L

 $C_{f}$  = feed concentration, mg/L

The local membrane rejections indicate the percentage of TOC associated with the high-molecular-weight fraction which will be retained on an UF membrane at any time during ultrafiltration.

The local membrane rejection of the UM05 ultrafiltraton membrane (nominal cutoff = 500 daltons; Amicon, Bedford, MA) for the model humic acid (A-2HA) samples at pH values of 5.5, 7.0, and 8.5 were 97.5%  $\pm$ 0.2%, 95% and 96%, respectively. In comparison, the local membrane rejections for the leachate samples were 46.9% (Day 749, pH=7.15), 57.2%  $\pm$  2.1% (Day 810, pH=7.21), and 63.1%  $\pm$  1.2% (Day 841, pH=7.21). The significant difference in the local membrane rejections for the

Sample	рН	TOC	Ultrafiltration UM05 (500 MW)			
			Permeate	Retentate	Local Membrane Rejection, R	
		mg/L	mg/L	mg/L	%	
A-2HA (1.0 g/L)	7.0	525 (200 ml) <sup>*</sup>	22.8 <u>+</u> 1.3 (148.5 ml <u>+</u> 1.3 ml)	1981.6 <u>+</u> 34.2 (51.5 ml <u>+</u> 1.3 ml)	97.5 <u>+</u> 0.2	
A-2HA <sup>**</sup> (0.5 g/L)	5.5	282 (100 ml)	20.2 (49 ml)	592 (51 ml)	95	
A-2HA <sup>**</sup> (0.5 g/l)	8.5	284 (100 ml)	16.5 (51 ml)	591 (49 ml)	96	
Leachate, <sup>**</sup> Day 749	7.15	665 (200 ml)	445.9 (146 ml)	1312.6 (54 ml)	46.9	
Leachate, Day 810	7.21	720 <u>+</u> 25.4 (100 ml)	371 <u>+</u> 9.6 (49 m1 <u>+</u> 1.8 m1)	1040 <u>+</u> 37.6 (51 ml <u>+</u> 1.8 ml)	57.2 <u>+</u> 2.1	
Leachate, Day 841	7.21	698 <u>+</u> 32.4 (100 ml)	309 <u>+</u> 13.3 (47.3 ml <u>+</u> 3.7 ml)	1024 <u>+</u> 59.1 (52.7 ml <u>+</u> 3.7 ml)	63.1 <u>+</u> 1.2	

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Table 18. Total Organic Carbon (TOC) Distribution in Different Molecular Weight Fractions of Model Humic Acid (A-2HA) and Leachate Samples

\*Volume of sample. \*\*Results are average of two samples.

model humic acid (A-2HA) samples and the leachate samples can be attributed to absence of the fulvic acid (low-molecular-weight) fraction in the model humic acid (A-2HA), whereas the leachate samples contained both fractions.

Model humic acid at three pH values (5.5, 7.0, and 8.5) and two leachate samples were fractionated into high-molecular-weight, intermediate-molecular-weight, and low-molecular-weight fractions using gel permeation chromatography. The gel used (Sephadex G-50; Pharmacia Fine Chemicals, Piscataway, NJ) had exclusion limits of 10,000 MW and 500 MW. As shown in Table 19, the TOC distributions obtained by gel permeation chromatography were similar to the local membrane rejections obtained by ultrafiltration (Table 18). In comparison with the model humic acid (A-2HA), the leachate samples exhibited a higher TOC content in the low-molecular-weight fraction. This can be attributed to the absence of the fulvic acid (low-molecular-weight) fraction in the model humic acid (A-2HA), since the fulvic acid fraction was separated from the model humic acid (A-2HA) during its isolation by Reuter (85). The percent recovery based on TOC ranged from 82.8% to 91.2% for model humic acid (A-2HA) and leachate samples, respectively.

#### 4.5.2 Quality Assurance/Quality Control (QA/QC) Study

In order to test the significance of the difference in the solubilization levels of cobalt sulfide in deionized water and model humic acid (A-2HA) a quality assurance/quality control (QA/QC) study was conducted. The results obtained from this study are summarized in Table 20. The cobalt concentrations in model humic acid and deionized water were 8.3 mg/L  $\pm$  1.2 mg/L and 3.7 mg/L  $\pm$  0.7 mg/L, respectively.

Sample	рН	TOC	Volume of Sample		T	DC	
		mg/L	Injected Into The GPC Column mL	High Molecular Weight mg/L	Intermediate Molecular Weight mg/L	Low Molecular Weight mg/L	Recovery x <u>+</u> <sup>g</sup> x
A-2HA (4)*	7.0	525	0.5	6.5 <u>+</u> 0.5	5.5 <u>+</u> 0.2	1.6 <u>+</u> 0.3	88.1 <u>+</u> 6.1
A-2HA (2)	5.5	520	0.5	5.4	5.9	1.8	86.9
A-2HA (2)	8.5	560	0.5	6.7	6.6	1.5	91.2
Leachate, Day 810 (4)	7.21	720	0.5	2.6 <u>+</u> 0.4	6.6 <u>+</u> 0.4	9.1 <u>+</u> 0.7	85.4 <u>+</u> 6.0
Leachate, Day 841 (4)	7.21	698	0.5	2.9 <u>+</u> 0.3	6.3 <u>+</u> 0.4	8.0 <u>+</u> 0.8	82.9 <u>+</u> 6.3

## Table 19. Total Organic Carbon (TOC) Distribution in Different Molecular Weight Fractions (Generated by GPC) of Model Humic Acid (A-2HA) and Leachate

\*Number of replicates.

.

Sample	Cobalt Sulfide Added	Cobalt Detected	t-Test
	$(\overline{x}+\sigma), g/L$	$(\bar{x}+\sigma)$ , mg/L	
A-2HA (17) <sup>*</sup> (0.5 g/L; pH=7.0)	0.49 <u>+</u> 0.07	8.3 <u>+</u> 1.2	t <sub>exp</sub> = 15.3 t <sub>v</sub> =35, α=0,05
Deionized Water (20) <sup>*</sup>	0.52 <u>+</u> 0.09	3.7 <u>+</u> 0.7	t <sub>ν</sub> =30, α=0.05 (Significant)

Table 20. Cobalt Solubilized from Cobalt Sulfide by Model Humic Acid (A-2HA) and Deionized Water in the Quality Assurance/Quality Control (QA/QC) Experiment

\*Number of replicates.

The Student t-Test on the means revealed that the difference between the amounts of cobalt solubilized by model humic acid (A-2HA) and deionized water is statistically significant; this indicated probable formation of cobalt-humic complexes. The calculations of the Student t-Test are presented in Appendix A.

### 4.5.3 Equilibrium Study

The results of the equilibrium study on solubilization of cobalt, from cobalt sulfide, by model humic acid (A-2HA), leachate samples, and deionized water (blank) are presented in Table 21. Deionized water solubilized 6.1 mg/L of cobalt, whereas model humic acid (A-2HA) at pH values of 5.5, 7.0, and 8.5 solubilized 10.8 mg/L, 8.3 mg/L, and 26.1 mg/L of cobalt, respectively. In comparison, the leachate samples, which were collected on Days 749, 810, and 941, solubilized 40.5 mg/L, 45.6 mg/L, and 42.3 mg/L of cobalt, respectively. The local membrane rejection for blank was 15%, whereas for model humic acid (A-2HA) at pH values of 5.5, 7.0, and 8.5 the local membrane rejections were 74.8%, 91.5%, and 94.9%, respectively. In comparison, the local membrane rejections of the leachate samples collected on Days 749, 810, and 841 were 64.1%, 43.3%, and 42.5%, respectively.

The local membrane rejection of cobalt indicates the percentage of solubilized cobalt retained along with the high-molecular-weight fraction on an ultrafiltration membrane. Thus, higher rejections of cobalt (i.e., 64.1%, 43.3%, and 42.5%) by the UM05 ultrafiltration membrane (nominal cutoff = 500 daltons; Amicon, Bedford, MA) for the leachate samples than for the blank (15%) indicate that significant amounts of solubilized cobalt are associated with the molecular-weight

Sample	pН			Cobalt Detected		
	•	Background	Solubilized	Permeate (UM05)	Retentate (UM05)	Local Membrane Rejection
		mg/L	mg/L	mg/L	mg/L	%
Blank (CoS +						
Deionized Water)	6.8	N.D.*	6.1	5.4 (45 mL)**	6.7 (55 mL)**	15.0
A-2HA (0.5 g/L)	5.5	N.D.*	10.8	3.5 (52 mL)	18.0 (48 mL)	74.8
A-2HA (0.5 g/L)	7.0	N.D.*	8.3	1.2 (148 mL)	27.9 (52 mL)	91.5
A-2HA (0.5 g/L)	8.5	N.D.*	26.1	1.8 (50 mL)	47.8 (50 mL)	94.9
Leachate, Day 749	7.15	0.4	40.5	17.1 (42 mL)	59.1 (58 mL)	64.1
Leachate, Day 810	7.21	0.2	45.6	29.3 (47 mL)	61.2 (53 mL)	43.3
Leachate, Day 841	7.21	0.3	42.3	27.5 (47 mL)	54.3 (53 mL)	42.5

# Table 21. Cobalt Solubilized from Cobalt Sulfide by Model Humic Acid (A-2HA), Leachate Samples, and Deionized Water

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\*None Detectable \*\*( ) Volume of fractionated sample.

fractions greater than 500 daltons. This can be attributed to the complexation of cobalt by humic substances present in the leachate samples. The increase in local membrane rejection of cobalt with pH for model humic acid (A-2HA) can be attributed to the increase in the solubility of model humic acid (A-2HA) with pH.

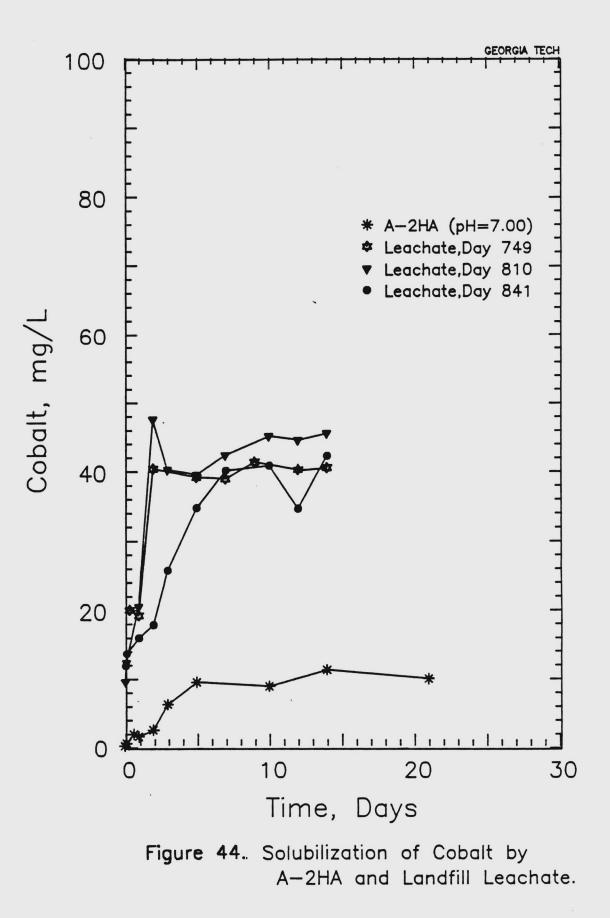
## 4.5.4 Kinetic Study

Results of the kinetic study (Figure 44) indicate that 80% of the final levels of solubilized cobalt were attained within three days. Thus, solubilization of cobalt from cobalt sulfide by leachate samples can be considered as a fairly rapid reaction since a period of three days can be envisioned as a short period when compared to the time period (measured in years) a landfill usually resides in the methane formation phase and the final maturation phase.

### 4.5.5 Confirmatory Study Using Cobalt-58

The solubilization of Co-58 from cobalt sulfide (containing both Co-58 and stable cobalt) by model humic acid (A-2HA; at pH values of 5.5, 7.0, and 8.5), leachate and deionized water (blank) was investigated and the results are presented in this section. The characteristics of the leachate used in this study are summarized in Table 22. Prior to utilization, the leachate was filtered through a 0.45 m filter. Since Co-58 was the only radionuclide present in the samples, radioactivity in each sample has been reported in counts per minute normalized to Day 0 (March 7, 1985) of the confirmatory study.

As shown in Table 23, the radioactivity in the residue (CoS) was the difference between the average radioactivity in the initial cobalt solutions and the radioactivity in the filtrate. The average radio-



Co-58 and Stable Cobalt)	÷
Parameter	Value
pH	7.27
ORP	-191 mV E <sub>C</sub>
Total alkalinity as CaCo <sub>3</sub>	1,825 mg/L
Conductivity	1.85 mmhos
Ionic strength	0.04 M
COD	1,522 mg/L
BOD <sub>5</sub>	342 mg/L
тос	619 mg/L
Total volatile acid as acetic acid	N.D.*
Cobalt	N.D.
Iron	8.6 mg/L
Manganese	2.0 mg/L
Sulfide	N.D.
· ·	

Table 22. Characteristics of Leachate (Day 970) That Was Utilized to Investigate the Solubilization of Co-58 from Cobalt Sulfide (Containing both Co-58 and Stable Cobalt)

\*None Detectable

Sample	РH	Cobalt Solution	Counts Per Filtrate	Cobalt in Cobalt Sulfide Residue
I	II	(x10 <sup>-6</sup> ) III	(x10 <sup>-6</sup> ) IV	on Filter Paper (x10 <sup>-6</sup> ) V=III <sub>avg</sub> *-IV
A-2HA, #1 A-2HA, #2 A-2HA, #3 A-2HA, #4 A-2HA, #5 A-2HA, #5 A-2HA, #6 A-2HA, #7 A-2HA, #8	7.00 7.00 7.00 5.50 5.50 8.50 8.50	20.79 19.50 19.67 19.78 19.23 17.95 14.83 16.13	4.73 4.87 4.99 4.69 5.01 5.71 4.24 5.22	12.95 12.81 12.69 12.99 12.67 11.97 13.44 12.46
Leachate,	#1 7.27 #2 7.27 #3 7.27 #4 7.27	16.49 17.17 16.84 17.26	5.10 5.23 5.71 5.11	12.58 12.45 11.97 12.57
Blank <sup>***</sup> , #1 Blank, #2 Blank, #3	6.80	16.07 16.15 17.47	5.27 5.20 4.88	12.41 12.48 12.80

Table 23. Radioactivity of the Cobalt Solution and the Filtrate

\*III<sub>avg</sub> = 17.66x10<sup>6</sup> counts per minute
\*\*Leachate Sample from Cell B; Day 970
\*\*\*Deionized Water

activity in the initial cobalt solutions was  $17.68 \times 10^6$  counts per minute and the average radioactivity retained on the filter papers (0.45 µm, MSI, Fisher Scientific, Atlanta, GA) was  $11.94 \times 10^6$  counts per minute. Thus, the radioactivity retained on a filter paper was  $71.3\% \pm 0.07\%$ .

The levels of Co-58 solubilized from cobalt sulfide on Days 12 and 28 are presented in Table 24. As shown, the levels of Co-58 solubilized increased significantly from Day 12 to Day 28. Also, the percent cobalt solubilized increased for each ligand from Day 12 to Day 28. These increases indicate that the solubilized levels of Co-58 were not at equilibrium on Day 12. No significant increases in Co-58 levels were observed on Day 90. The data presented in Table 24 correspond to equilibrium solubilized cobalt levels (Day 28) of 6.2 mg/L, 15.6 mg/L, 17.7 mg/L, 20.9 mg/L and 24.9 mg/L, respectively, for blank, leachate, A-2HA (pH=8.5), A-2HA (pH=5.5), and A-2HA (pH=7.0). As expected, these values are similar to the solubilized cobalt levels reported for stable cobalt (Table 21).

The different fractions obtained by fractionating Day 28 samples with gel permeation chromatography were analyzed for radioactivity. The relative radioactivity in each fraction expressed as a percent of the total radioactivity in the sample, is shown in Figures 45 through 48. Blue dextran (MW >  $2x10^6$  daltons) and phenol (MW = 94 daltons) were used as standards and for a given set of conditions (column details, flow rate of mobile phase, and type of mobile phase) the elution pattern indicated the volumes within which blue dextran (highmolecular-weight fraction) and phenol (low-molecular-weight fraction)

Sample	рН	Counts Per Day 12	Day 28		58 Solubilized a bay 12		t Sulfide ay 28
		(x10 <sup>-5</sup> )	(x10 <sup>-5</sup> )	% ×1	$\frac{\pi}{x_1} \pm \sigma_{x_1}$	<b>%</b> x <sub>2</sub>	$\frac{\frac{\pi}{2}}{x_2} \pm \sigma_{x_2}$
A-2HA, #1 A-2HA, #2	7.00 7.00	6.39 6.46	10.05 9.75	4.9 5.0	50+02	7.7 7.6	75+01
A-2HA, #3 A-2HA, #4	7.00 7.00	6.47 6.98	9.57 9.66	5.0 5.3	5.0 <u>+</u> 0.2	7.5 7.4	7.5 <u>+</u> 0.1
A-2HA, #5	5.5	6.54	9.07	5.1	4.6*	7.1	6.3*
A-2HA, #6 A-2HA, #7	5.5 8.5	4.95 5.05	6.64 7.33	4.1 3.7		5.5 5.4	
A-2HA, #8	8.5	3.68	6.65	2.9	3.3*	5.3	5.35*
Leachate, #1	7.27	4.89	-	3.8			
Leachate, #2	7.27	4.13	4.35	3.3	3.5 <u>+</u> 0.2	3.4	4.7 <u>+</u> 1.1
Leachate, #3 Leachate, #4	7.27 7.27	4.26 4.29	6.50 6.89	3.5 3.4		5.4 5.5	
Blank, #1	6.80	2.42	2.89	1.9		2.3	
Blank, #2 Blank, #3	6.80 6.80	2.07 1.50	2.38 1.92	1.6 1.2	1.6 + 0.3	1.9 1.5	1.9 <u>+</u> 0.4

Table 24. Co-58 Solubilized from Cobalt Sulfide by Model Humic Acid (A-2HA), Leachate Sample from Cell B (Day 970), and Deionized Water (Blank)

\*Average of two samples.

were eluted. These volumes were 15 mL  $\pm$  0.16 mL and 35 mL  $\pm$  0.52 mL for blue dextran and phenol, respectively.

As shown in Figures 45-48, the Co-58 solubilized by blank (deionized water) coeluted with phenol. However, the elution patterns for the solubilized Co-58 in model humic acid (A-2HA) and leachate sample exhibited a shift towards left relative to the profile for the blank; this indicates that a fraction of the solubilized Co-58 is associated with high-molecular-weight and intermediate-molecular-weight organics. As expected, the largest shift to the left occurred for model humic acid (A-2HA) at pH=8.5 and the smallest shift occurred for model humic acid (A-2HA) at pH=5.5. This can be attributed to a decrease in the presence of the low-molecular-weight fraction of the model humic acid (A-2HA) with an increase in pH from 5.5 to 8.5 since the solubility of the model humic acid (A-2HA) increased with the increase in pH. Thus, the molecular-weight profiles generated by GPC confirm the complexation of Co-58 by humic substances present in landfill leachate.

# 4.6 <u>Migratory Activity Levels of Co-58, Sr-85, and Ce-141</u> from the Landfills in the Event of a Liner Failure

The loading levels, based on dry weight or refuse, of Co-58, Sr-85, and Ce-141 utilized in this study were 1.6 x  $10^{-7}$  Ci/kg, 2.0 x  $10^{-7}$  Ci/kg, and 1.06 x  $10^{-6}$  Ci/kg, respectively. During the period of investigation, the maximum levels, expressed as a percent of initial loading levels, of Co-58, Sr-85, and Ce-141 in leachate from the pilot-scale landfill with provisions for leachate recycle (Cell B) were 12.7%, 15.7%, and 0.03%, respectively. The maximum levels of Co-58,

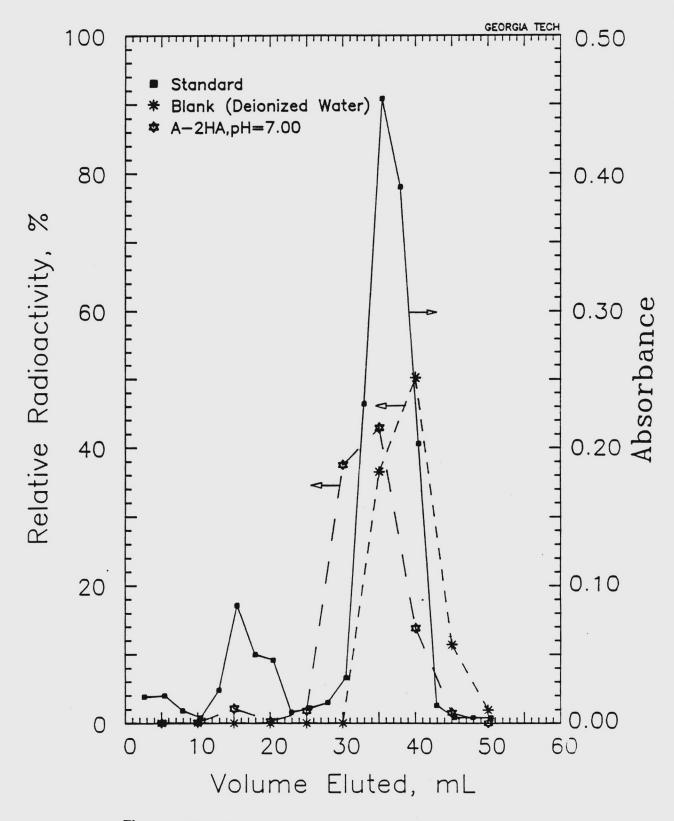


Figure 45. Radioactivity Present in Different GPC Fractions of Blank and A-2HA (pH=7.00).

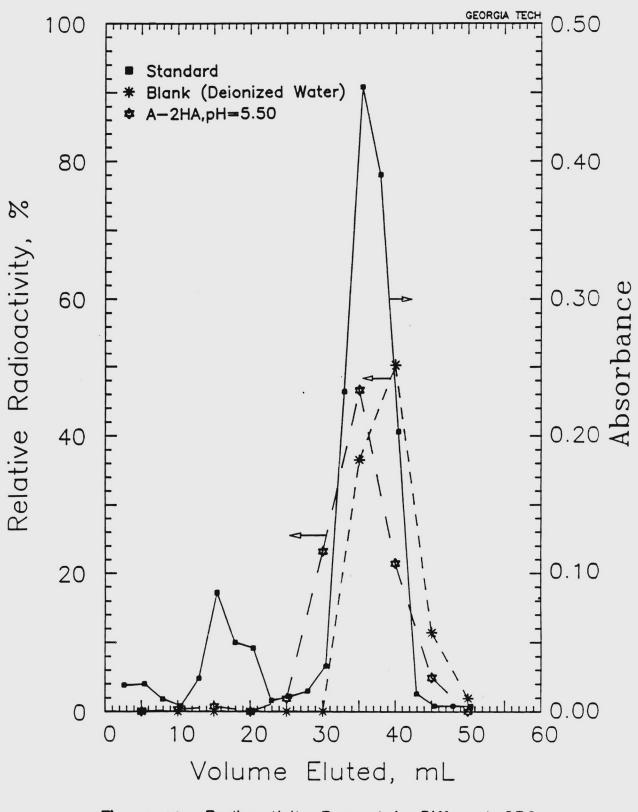


Figure 46. Radioactivity Present in Different GPC Fractions of Blank and A-2HA (pH=5.50).

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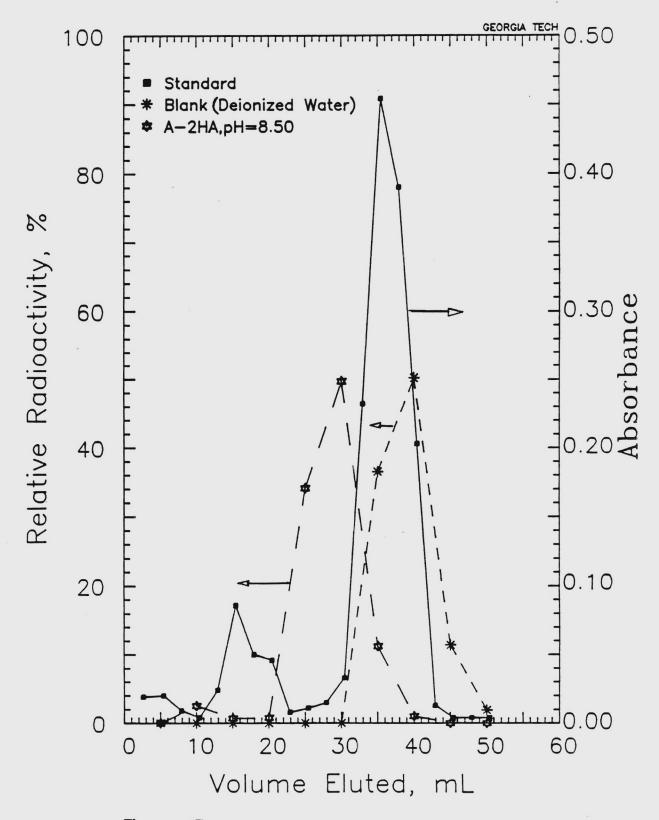


Figure 47. Radioactivity Present in Different GPC Fractions of Blank and A-2HA (pH=8.50).

GEORGIA TECH 100 Standard 2 Blank (Deionized Water) Leachate Ż 0.40 80 Relative Radioactivity, % o o Absorbance 60 \* 40 0.10 20 00.0L 0 30 20 10 40 50 60 0 Volume Eluted, mL

Figure 48. Radioactivity Present in Different GPC Fractions of Blank and Leachate.

Sr-85, and Ce-141 in leachate from the pilot-scale landfill without provisions for leachate recycle (Cell A) were lower than the levels present in leachate from Cell B. Based on these maximum levels present in leachate (Co-58 = 12.7%, Sr-85 = 15.7%, and Ce-141 = 0.03%), and neglecting any dilution, the migratory levels of Co-58, Sr-85, and Ce-141, in the event of a liner failure were 0.09 mCi/m<sup>3</sup>, 0.13 mCi/m<sup>3</sup>, and 0.0015 mCi/m<sup>3</sup>, respectively. Detailed computations are presented in Appendix C. These levels are below the maximum permissible concentrations of these radionuclides in water (Co-58 = 1 mCi/m<sup>3</sup>, Sr-85 = 3 mCi/m<sup>3</sup>, and Ce-141 = 3 mCi/m<sup>3</sup>; 10CFR, Part 20, Appendix B). Thus, the codisposal of the selected radionuclides, Co-58, Sr-85, and Ce-141, with municipal refuse is feasible at the loading levels (Co-58 = 1.6 x  $10^{-7}$  Ci/kg, Sr-85 = 2.0 x  $10^{-7}$  Ci/kg, and Ce-141 = 1.06 x  $10^{-6}$  Ci/kg) utilized in this study.

### CHAPTER V

#### CONCLUSIONS

Based on the results obtained from this study, the following conclusions can be drawn:

- 1. Co-58, Sr-85, and Ce-141 can be safely codisposed with municipal refuse within landfills at the loading levels utilized in this study, since the migratory activity levels of these radionuclides, in the event of a liner failure, will be less than the maximum permissible concentrations of these radionuclides in water above natural background. The loading levels, based on dry weight of refuse, for Co-58, Sr-85, and Ce-141 were 1.6 x 10<sup>-7</sup> Ci/kg, 2.0 x 10<sup>-7</sup> Ci/kg, and 1.06 x 10<sup>-6</sup> Ci/kg, respectively.
- 2. The containment mechanism for Co-58 within the landfills was the formation of insoluble cobalt sulfide. The actual cobalt concentrations in leachate, however, were greater than equilibrium concentrations of cobalt in the presence of sulfide, carbonate, and hydroxide. This increase in the concentrations of cobalt in leachate is attributed to the formation of soluble complexes with other ligands (viz., humic substances) present in leachate during landfill stabilization. A separate confirmation of complexation of cobalt with humic substances present in leachate was provided

by companion laboratory-scale studies, wherein, leachate samples from the landfill operated as a landfill with leachate recycle (Cell B) solubilized Co-58 from cobalt sulfide precipitate.

- 3. The containment of Sr-85 within the landfill may be attributed to the formation of insoluble strontium carbonate complexes. This behavior was particularly reflected by strontium in leachate from the landfill operated as a landfill with leachate recycle (Cell B), as the strontium concentrations in leachate decreased with the change in the predominant buffer system from acetate to carbonate and increases in pH and TIC of leachate.
- 4. During landfill stabilization, the maximum levels of Co-58 and Sr-85 in leachate from Cell B, expressed as a percent of the initial loading levels, were 12.7% and 15.7%, respectively. The activity of Co-58 and Sr-85 in leachate corresponding to these levels, however, were lower than the maximum permissible concentration of these radionuclides in water above natural background.
- 5. Greater than 99.97% of the initial loading level of Ce-141 was retained within the landfills. The containment of Ce-141 within the landfills may be attributed to its adsorption onto the refuse.
- 6. During landfill stabilization, the behavior exhibited by Co-58 and Sr-85 in leachate from the landfill operated as a landfill with leachate recycle (Cell B) was similar to the

behavior exhibited by their respective stable forms. Thus, there was no preferential partitioning of the radionuclides as compared to their stable forms.

- 7. Since the total volatile fatty acid concentration in leachate from the landfill operated as landfill with leachate recycle (Cell B) persisted at a high level (7-10 g/L) up to Day 152, an attempt was made to enable the stabilization processes to proceed from the acid formation phase to the subsequent phases, namely, the methane fermentation phase and the final maturation phase, by reducing the frequency of leachate recycle from once a day to once a week between Days 252-350. This led to the successful recovery of Cell B with respect to progress of stabilization processes from the acid formation phases.
- 8. In a landfill receiving both municipal refuse and Co-58, the mobilization of Co-58 by humic substances present in leachate can be minimized by terminating leachate recycle after the degradation of readily degradable organics has occurred (final maturation phase), since the termination of leachate recycle will reduce both the formation of humic substances, and the contact opportunities provided to the humic substances in leachate to complex Co-58.

#### CHAPTER VI

#### RECOMMENDATIONS

Based on the results of this study, the codisposal of Co-58, Sr-85, and Ce-141 with municipal refuse, at the loading levels of 1.6 x  $10^{-7}$  Ci/kg, 2.0 x  $10^{-7}$  Ci/kg, and 1.06 x  $10^{-6}$  Ci/kg, respectively, within landfills is recommended. In addition, further studies at higher loading levels of these radionuclides are recommended. Furthermore the technical feasibility of codisposal of radionuclides, namely, P-32, Cr-51, Se-75, Mo-99, I-131, Pu-238, Pu-239, and Pu-240, with municipal refuse within landfills should be evaluated. These radionuclides constitute LLRW and are generated from two sources: nuclear fuel cycle and nuclear medicine. The codisposal of LLRW, that are short-lived (half-life less than 300 days), with municipal refuse within landfills is an attractive alternative to the disposal of these radionuclides by shallow land burial, since it may alleviate some of the storage problems for such radionuclides. Dominant scenarios and potential pathways with respect to LLRW codisposed with municipal refuse within landfills should be evaluated. The results obtained from such studies would aid in assessment of the behavior of LLRW, within landfills, during landfill stabilization.

APPENDIX A

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Sample Calculation of Testing Means by Student t-test.

A-2HAD.I. Water
$$n_1 = 17$$
 $n_2 = 20$  $v = (n_2-1) = 16$  $v_2 = (n_2-1) = 19$  $\overline{x}_1 = 8.3 \text{ mg/L}$  $\overline{x}_2 = 3.71 \text{ mg/L}$  $\sigma_1 = 1.2 \text{ mg/L}$  $\sigma_2 = 0.7 \text{ mg/L}$ SS1 = 22.9SS2 = 8.8

$$s^{2} = \frac{22.9 + 8.8}{16 + 19}$$

$$s_{D} = s_{X} - s_{X} = \sqrt{s^{2}(\frac{1}{n_{1}} + \frac{1}{n_{2}})} = 0.3$$

$$t = \frac{8.3 - 3.7}{0.3} = 15.33$$

From Tables:

$$t_v = 40, \alpha = 0.05 = 2.021$$
  
 $t_v = 30, \alpha = 0.05 = 2.042$   
 $\therefore \bar{x}_1 \neq \bar{x}_2$ 

APPENDIX B

С С С MINEQL WAS MODIFIED FOR CDC APPLICATION BY S.B.GHOSH С С PROGRAM MAIN (INPUT, INPUT2, INPUT3, OUTPUT, TAPE5=INPUT, 1TAPE10=INPUT2, TAPE11=INPUT3, TAPE6=OUTPUT) COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN (6), NNN 00000010 COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),00000020 1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400) 0000030 С 00000040 NYDIM=400 00000050 NXDIM=30 00000060 ITMAX=100 00000070 EPS=1.E-4 00000080 ITER=0 0000090 · THRSH=.01 00000100 XMU=0.01 00000110 С 00000120 CALL INPUT 00000130 CALL INION 00000150 CALL IONCOR (XMU) 00000160 CALL OINCMP 00000180 CALL OINSPC 00000190 С RUN A SERIES OF COMPUTATIONS WITH PH FIXED FROM PH=(1 - 14)DO 2222 J=1,14 ITER=0 GR(IADY(50)) = J10 CONTINUE 00000200 CALL SOLID 00000210 CALL SOLVE 00000220 CALL SOLIDX(K) 00000230 IF (K.NE.0) GO TO 10 00000240 CALL OUTCMP 00000250 CALL OUTSPC 00000260 CALL OUTPC (THRSH) 00000270 2222 CONTINUE STOP 00000280 END 00000290 SUBROUTINE ERROR (I) 00000300 CHARACTER\*4 MSG(40) DATA MSG/ DATA MSG/ 1'COMP', 'PONE', 'NTS ', '> NX', 'DIM ', 2'SPEC', 'IES ', '> NY', 'DIM ', ', 3'ID ', 'NOT ', 'FOUN', 'D: I', 'NPUT', 4'ID ', 'NOT ', 'FOUN', 'D: I', 'ADY ', 5'ID ', 'NOT ', 'FOUN', 'D: I', 'ADX ', 6'PHAS', 'E RU', 'LE V', 'IOLA', 'TION', 7'ITER', 'ATIO', 'NS >', 'ITMA', 'X ', 8'SING' 'ULAP' 'Z M' 'ATPL' 'X '/ 00000320 00000330 00000340 00000350 00000360 00000370 00000380 8'SING', 'ULAR', ' Z M', 'ATRI', 'X 00000390 С 00000400 I1=(I-1)\*5+100000410 12=1\*5 00000420 WRITE(6,600) I, (MSG(II), II=I1, I2) 00000430 FORMAT('1', '\*\*\*\* EXECUTION TERMINATED \*\* ERROR', I3, ' \*\* ', 5A4) 600 00000440 С 00000450 CALL OUTCMP 00000460 CALL OUTSPC 00000470 STOP 00000480 00000490 END

		101
	SUBROUTINE EXCOL(J0,JJ)	00000500
	COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN (6), NNN	00000510
	COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),	
		00000520
14	1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400)	00000530
С		00000540
	IV=IDX(JJ)	00000550
	IDX(JJ) = IDX(JO)	00000560
	IDX(JO) = IV	00000570
	V=X (00)	00000580
	X(JO) = X(JJ)	00000590
	X (JJ) = V	00000600
	V=GX (J0)	00000610
	GX(JO) = GX(JJ)	00000620
	GX(JJ) = V	00000630
	V=T(J0)	00000640
	T(JO) = T(JJ)	00000650
	T(JJ) = V	00000660
	DO 603 I=1,NYDIM	00000670
	V=A(I,JO)	
		00000680
	A(I, J0) = A(I, JJ)	00000690
	A(I,JJ)=V	00000700
603	CONTINUE	00000710
	RETURN	00000720
С		00000730
	ENTRY EXROW(IO, II)	00000740
	IV-IDY(II)	00000750
	IDY (II) - IDY (IO)	00000760
	IDY (10) = IV	00000770
	DO 46 J=1,NXDIM	00000780
	V=A(I0, J)	00000790
	A(IO, J) = A(II, J)	00000800
	A(II,J)=V	00000810
46	CONTINUE	00000820
	V=GK (10)	00000830
	GK(I0) = GK(II)	00000840
	GK(II)=V	00000850
	RETURN	00000860
	END	00000870
	FUNCTION IADY (IDYT)	00000880
	COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN (6), NNN	00000890
	COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),	00000900
	1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400)	00000910
С		00000920
	II=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)+NN(6)	00000930
	DO 10 I=1,II	00000940
	IF(IDY(I).EQ.IDYT) GO TO 20	00000950
10	CONTINUE	00000960
10	CALL ERROR (4)	00000970
20		
20	IADY=I	00000980
	RETURN	00000990
C.		00001000
	ENTRY IADX (IDXT)	00001010
	J J-NNN	00001020
	DO 110 J=1, JJ	00001030
	IF(IDX(J).EQ.IDXT) GO TO 120	00001040
110	CONTINUE	00001050
	CALL ERROR (5)	00001060
120		
120	IADX=J	00001070
	RETURN	00001080
	END	00001090

SUBROUTINE INION 00001100 COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN(6), NNN 00001110 COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),00001120 1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400) 00001130 DIMENSION IONZ (200) 00001140 С 00001150 GF0=0.0 00001160 READ(11,500) (IONZ(J), J=1,200) 00001170 500 FORMAT (4012) 00001180 C 00001190 RETURN 00001200 ENTRY CONCOR (XMU) 00001210 NC=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)+NN(6)00001220 00001230 NX=NNN С 00001240 ET=-0.5 00001250 00001260 SI=SORT(XMU) GF=ET\*(SI/(1.0+SI)-0.2\*XMU) 00001270 С 00001280 DGF=GF-GF0 00001290 GF0=GF 00001300 С 00001310 WRITE(6,60) XMU,GFO 00001320 FORMAT('0',' IONIC STRENGTH = ',1PE9.2,5X,' LOG F(Z=1) =',0PF6.2)00001330 60 00001340 C DO 20 I=1,NC 00001350 VJ=0.0 00001360 VI=0.0 00001370 DO 30 J=1,NX 00001380 VJ=VJ+A(I,J)\*IONZ(IDX(J))\*IONZ(IDX(J))00001390 VI=VI+A(I, J)\*IONZ(IDX(J)) 00001400 00001410 30 CONTINUE  $GK(I) = GK(I) + DGF^{*}(VJ - VI^{*}VI)$ 00001420 20 CONTINUE 00001430 RETURN 00001440 END 00001450 SUBROUTINE INPUT 00001460 00001470 COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN (6), NNN COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),00001480 1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400)00001490 С 00001500 DIMENSION IADXT(200), IAT(4), IDT(4)00001510 С 00001520 INITIALIZE ADDRESS 00001530 С 00001540 DO 20 J=1,200 20 IADXT(J)=000001550 00001560 C С INPUT PROBLEM DATA 00001570 00001580 J=0 READ(5,500) IDXT,GXT,TT 00001590 10 IF(IDXT.EQ.0) GO TO 90 00001600 J=J+100001610 IDX(J)=IDXT 00001620 GX(J) = GXT00001630 T(J) = TT00001640 X(J) = 10 \* GXT00001650 IADXT(IDXT) = J00001660 GO TO 10 00001670 90 CONTINUE 00001680 IF (J.GT.NXDIM) CALL ERROR(1) 00001690

	NNN-J	00001700
с		00001710
С	INITIALIZE NN	00001720
	DO 100 L=1,6	00001730
100	NN(L) = 0	00001740
С	INITIALIZE A	00001750
-	DO 110 I=1,NYDIM	00001760
	DO 110 J=1,NXDIM	00001770
110	A(I, J) = 0.0	00001780
c	INPUT BASIS IN A MATRIX	00001790
	JJ-NNN	00001800
	DO 200 I=1,JJ	00001810
	IDY(I) = IDX(I)	00001820
	A(I, I) = 1.0	00001820
200	GK(I)=0.0	00001840
200	NN(1)-JJ	00001850
с	MA(1) 55	00001860
č	INPUT THERMODYNAMIC DATA	00001870
Ŭ	I=NN(1)	00001880
	DO 400 L=2,6	00001890
	IO-I	00001900
	READ(10,510) IN	00001910
	IF(IN.EQ.0) GO TO 400	00001910
	DO 300 II-1, IN	00001920
	READ(10,510) IDYT, GKT, (IDT(J), IAT(J), J=1,4)	00001930
	DO 310 J=1.4	00001940
	JTEST-IDT (J)	00001950
	IF(JTEST.EQ.0) GO TO 310	00001970
	IF (IADXT (JTEST).EQ.0) GO TO 300	00001980
310	CONTINUE	00001990
510	I=I+1	00002000
	IDY(I)-IDYT	00002010
	GK(I) = GKT	00002020
	DO 320 J=1,4	00002030
	JTEST-IDT (J)	00002040
	IF(JTEST.EQ.0) GO TO 320	00002050
	A(I, IADXT(JTEST)) = IAT(J)	00002060
320	CONTINUE	00002070
300	CONTINUE	00002080
300	NN(L) = I - IO	00002090
400	CONTINUE	00002100
	CONTINUE	00002110
C C	READ SPECIES MODIFICATION & TYPE SPECIFICATIONS	00002120
C	ENTRY INTYPE	00002120
2000	READ(5,510) LTYPE	00002130
2000	IF (LTYPE.EQ.0) GO TO 1000	00002150
3000	READ(5,510) IDYT, GKT, (IDT(J), IAT(J), $J=1,4$ )	00002160
2000	IF(IDYT.EQ.0) GO TO 2000	00002170
с	SEARCH	00002180
C	II=0	00002190
	DO 710 L=1,6	00002200
	IF (NN (L).EQ.0) GO TO 710	00002210
	IO=II+1	
	II = II + NN (L)	00002220
		00002230
	DO 720 I=I0, II	00002240 00002250
	IF (IDY (I).NE.IDYT) GO TO 720	
	IF (GKT.NE.0.0) GK (I) = GKT	00002260
720	GO TO 900	00002270
720	CONTINUE	00002280
710	CONTINUE	00002290

c	SEARCH UNSUCCESSFUL: ENTER NEW SPECIES	00000000
С		00002300
	IALLO=0	00002310
	DO 810 J=1,4	00002320
	JTEST=IDT(J)	00002330
	IF (JTEST.EQ.0) GO TO 810	00002340
	IF (IADX(JTEST).EQ.0) CALL ERROR(3)	00002350
	IALLO=1	00002360
810	CONTINUE	00002370
010		
	IF (IALLO.EQ.0) CALL ERROR (3)	00002380
	I=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)+NN(6)+1	00002390
	NN(6) = NN(6) + 1	00002400
	L=6	00002410
	IDY(I)-IDYT	00002420
	GK(I)=GKT	00002430
	DO 820 J=1,4	00002440
	JTEST-IDT(J)	00002450
	IF(JTEST.EQ.0) GO TO 820	00002460
	A(I, IADX(JTEST)) = IAT(J)	00002400
000		
820	CONTINUE	00002480
С	MOVE SPECIES I FROM TYPE L TO LTYPE	00002490
900	CONTINUE	00002500
	IF(L.EQ.LTYPE) GO TO 910	00002510
	K-1	00002520
	11-0	00002530
	DO 940 LL=1,L	00002540
940	II=II+NN(LL)	00002550
	IF (LTYPE.GT.L) GO TO 920	00002560
	K=-1	00002570
	II=II-NN(L)+1	00002580
000		
920	CONTINUE	00002590
	NN(L) = NN(L) - 1	00002600
	NN (LTYPE) = NN (LTYPE) +1	00002610
930	CALL EXROW(I,II)	00002620
	L=L+K	00002630
	I=II	00002640
	II=II+NN(L)*K	00002650
	IF(L.NE.LTYPE) GO TO 930	00002660
910	CONTINUE	00002670
10	GO TO 3000	00002680
~	60 10 5000	
С		00002690
1000	CONTINUE	00002700
	RETURN	00002710
500	FORMAT(15,2X,F7.2,E7.2)	00002720
510	FORMAT(15,2X,F7.2,4(14,13))	00002730
	END	00002740
	SUBROUTINE OUTPUT	00002750
	CHARACTER*4 NAME(200), TYPE(42)	
	COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN (6), NNN	00002760
	COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),	
		00002770
	1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400)	00002780
	DIMENSION IAT(4), IDT(4)	
	DATA NAME/	
	1'CA ','MG ','SR ','K ','NA ','FE3 ','FE2 ','MN2 ','CU2 ',	00002800
	2'BA ','CD ','ZN ','NI ','HG ','PB ','CO2 ','CO3 ','AG ',	00002810
	3'CR ','AL ','CS ','LI ','BE ','SC ','TIO ','SN2 ','SN4 ',	00002820
	4'LA ','CE3 ','AU1 ','TH4 ','UO2 ','CU1 ','X34 ','X35 ','X36 ',	00002830
	513*' ','H ',48*' ','E- ',' ',	00002840
	6'CO3-','SO4 ','CL ','F ','BR ','I ','NH3 ','S ','PO4 ',	00002850
	7'P207', 'P010', 'SI03', 'S203', 'CN ', 'AC ', 'ACAC', 'CIT ', 'OX ',	00002860
	8'SAL ', 'TART', 'EN ', 'DIP ', 'SUSA', 'GLY ', 'GLU ', 'PIC ', 'NTA ',	00002870

9'EDTA', 'DCTA', 'CYST', 'NOR ', 'PHTH', 'ARG ', 'ORN ', 'LYS ', 'HIS ', &'ASP ', 'SER ', 'ALA ', 'TYR ', 'MET ', 'VAL ', 'THR ', 'PHE ', 'ISO ', 1'LEU ', 'PRO ', 'BOH4', 'SO3 ', 'SCN ', 'NHOH', 'MOO4', 'WO4 ', 'ASO4', 2'VO4 ', 'SEO4', 'NO3 ', 'TRIS', 'X159', 'X160', 'X161', 'X162', 'X163', 00002880 00002890 00002900 00002910 337\*' '/ 00002920 DATA TYPE/ 1'I - ', 'COMP', 'ONEN', 'TS ', 3\*' 00002940 1 1 - ', 'COM', 'PLEX', 'ES ',3\*' ', 2'II -', 'COM', 'PLEX', 'ES ',3\*' ', 3'III ','- FI', 'XED ','SOLI', 'DS ',2\*' ', 4'IV -', 'PRE', 'CIPI', 'TATE', 'D SO', 'LIDS',' ', 5'V - ', 'DISS', 'OLVE', 'D SO', 'LIDS',2\*' ', 6'VI -', 'SPE', 'CIES', 'NOT', 'CON', 'SIDE', 'RED'/ 00002950 00002960 00002970 00002980 00002990 C 00003010 C INPUT DATA COMPONENTS 00003020 ENTRY OINCMP 00003030 JJ=NNN 00003040 WRITE(6,700) 00003050 WRITE(6,610) 00003060 DO 10 J=1, JJ 00003070 10 WRITE (6, 620) IDX (J), X (J), GX (J), T (J), NAME (IDX (J))00003080 RETURN 00003090 00003100 С C INPUT DATA SPECIES 00003110 ENTRY OINSPC 00003120 II=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)+NN(6)00003130 JJ=NNN 00003140 L=0 00003150 M-1 00003160 DO 50 I=1.II 00003170 IF(M.NE.I) GO TO 30 00003180 20 L=L+1 00003190 IF (NN(L).EQ.0) GO TO 20 00003200 M=M+NN(L)00003210 L2=L\*7 00003220 L1=L2-6 00003230 WRITE (6,600) 00003240 WRITE (6,630) (TYPE(N), N=L1, L2) 00003250 30 K=0 00003260 DO 40 J=1.JJ 00003270 IF (A(I,J).EQ.0) GO TO 40 00003280 K=K+1 00003290 IDT(K) = IDX(J)00003300 IAT(K) = A(I, J)00003310 40 CONTINUE 00003320 50 WRITE(6,640) IDY(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 00003330 RETURN 00003340 C 00003350 C COMPONENT OUTPUT 00003360 ENTRY OUTCMP 00003370 II=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)+NN(6)00003380 JJ=NNN 00003390 WRITE(6,650) ITER 00003400 WRITE(6, 660)00003410 DO 60 J=1, JJ 00003420 60 WRITE (6, 670) IDX (J), X (J), GX (J), T (J), Y (J), NAME (IDX (J))00003430 RETURN 00003440 С 00003450 C SPECIES OUTPUT 00003460 ENTRY OUTSPC 00003470 II=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)+NN(6)00003480

L=0 M=1 D0 100 I=1,II IF (M.NE.I) GO TO 80 70 L=L+1 IF (NN(L).EQ.0) GO TO 70 M=M+NN(L) L2=L*7 000 WRITE (6,600 WRITE (6,600) WRITE (6,680) (TYPE(N),N=L1,L2) 80 K=0 D0 90 J=1,JJ IF (A(I,J).EQ.0) GO TO 90 K=K+1 10T(K)=IDX(J) IDT(K)=IDX(J) 100 WRITE (6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 000 C C OUTPUTS PERCENTAGES 000 C OUTPUTS PERCENTAGES 000 C OUTPUTS PERCENTAGES 000 WRITE (6,730) 000 WRITE (6,730) 000 WRITE (6,710) NAME(IDX(J)) WRITE (6,710) NAME(IDX(J)) 000 WRITE (6,710) NAME(IDX(J)) 000 001 001 001 001 001 001 0	0003490 0003500 0003510 0003520 0003530 0003540 0003550 0003560 0003560 0003570 0003580 0003600 0003610 0003610 0003620 0003620 0003630 0003640 0003650 0003650 0003650 0003650 0003650 0003650 0003650 0003710 0003710 0003720 0003730 0003740 0003750 0003750 0003750 0003750 0003750 0003750
L=0 M=1 D0 100 I=1,II IF (M.NE.I) GO TO 80 70 L=L+1 IF (NN(L).EQ.0) GO TO 70 M=M+NN(L) L2=L*7 000 WRITE (6,600 WRITE (6,600) WRITE (6,680) (TYPE(N),N=L1,L2) 80 K=0 D0 90 J=1,JJ IF (A(I,J).EQ.0) GO TO 90 K=K+1 10T(K)=IDX(J) IDT(K)=IDX(J) 100 WRITE (6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 000 C C OUTPUTS PERCENTAGES 000 C OUTPUTS PERCENTAGES 000 C OUTPUTS PERCENTAGES 000 WRITE (6,730) 000 WRITE (6,730) 000 WRITE (6,710) NAME(IDX(J)) WRITE (6,710) NAME(IDX(J)) 000 WRITE (6,710) NAME(IDX(J)) 000 001 001 001 001 001 001 0	0003500 0003510 0003520 0003520 0003520 0003520 0003550 0003560 0003560 0003570 0003580 0003600 0003610 0003620 0003620 0003630 0003640 0003650 0003650 0003650 0003650 0003650 0003670 0003710 0003710 0003720 0003720 0003720 0003720 0003740 0003750 0003760 0003760 0003770
M=1         000           D0 100 I=1, II         000           JF (M.NE.I) GO TO 80         000           70         L=L+1         000           IF (NN(L).EQ.0) GO TO 70         000           M=M+NN(L)         000           L2=L*7         000           L1=L2-6         000           WRITE (6,680) (TYPE(N), N=L1, L2)         000           80         K=0         000           D0 90 J=1, JJ         000           IF (A(I,J).EQ.0) GO TO 90         000           K=K+1         000           IDT(K)=IDX(J)         000           IDT (K)=A(I,J)         000           RETURN         000           C         000           C         000           WRITE (6,690) IDY(I), C(I), GC(I), GK(I), (NAME(IDT(J)), IAT(J), J=1, K)         000           MRITE (6,730)         000           JJ=NNN         000           WRITE (6,710) NAME(IDX(J))         000           WRITE (6,710) NAME(IDX(J))         000           WRITE (6,710) NAME(IDX(J))         000           WRITE (6,700) GO TO 140         000           IF(NN(3).EQ.0) GO TO 140         000           IF(NN(3).EQ.0) GO TO 140         000     <	0003510 0003520 0003530 0003540 0003550 0003550 0003570 0003580 0003590 0003610 0003610 0003620 0003620 0003630 0003650 0003650 0003650 0003660 0003670 0003710 0003710 0003720 0003730 0003740 0003750 0003760
D0 100 I=1, II 000 IF (H.NE.I) G0 T0 80 000 0 L=1-1 000 0 M=M+N(L).EQ.0) G0 T0 70 000 0 M=M+N(L).EQ.0) G0 T0 70 000 1 L=L2-6 000 0 MRITE (6,600) (YPE(N),N=L1,L2) 000 0 WRITE (6,680) (TYPE(N),N=L1,L2) 000 0 D0 90 J=1,JJ 000 0 CONTINUE 000 1 D1 (K)=DX(J) 000 0 CONTINUE 000 10 WRITE (6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 000 0 RETURN 000 0 CONTINUE 000 10 WRITE (6,730) 000 0 JJ=NNN 000 WRITE (6,710) NAME(IDX(J)) 000 WRITE (6,710) NAME(IDX(J)) 000 0 II 0 J=1,JJ 000 0 WRITE (6,710) NAME(IDX(J)) 000 0 WRITE (6,710) NAME(IDX(J)) 000 0 II 0 J=1,JJ 000 0 II	0003520 0003530 0003540 0003550 0003550 0003570 0003580 0003590 0003610 0003610 0003620 0003620 0003630 0003650 0003650 0003650 0003650 0003660 0003670 0003710 0003710 0003720 0003730 0003740 0003750 0003760
$ \begin{array}{ccccc} \mathrm{IF} (\mathrm{H}, \mathrm{NE}, \mathrm{I}) & \mathrm{GO} \ \mathrm{TO} \ 80 & 000 \\ \mathrm{70} & \mathrm{L}=\mathrm{L}+\mathrm{1} & 000 \\ \mathrm{IF} (\mathrm{NN}(\mathrm{L}), \mathrm{EQ}, 0) \ \mathrm{GO} \ \mathrm{TO} \ 70 & 000 \\ \mathrm{M}=\mathrm{M}+\mathrm{NN}(\mathrm{L}) & 000 \\ \mathrm{L}=\mathrm{L}=\mathrm{L}^{2}-\mathrm{K} & 000 \\ \mathrm{L}=\mathrm{L}=\mathrm{L}=\mathrm{L}^{2}-\mathrm{K} & 000 \\ \mathrm{MR} \ \mathrm{ITE} \ (6,600) & 000 \\ \mathrm{MR} \ \mathrm{ITE} \ (6,680) \ (\mathrm{TYPE}(\mathrm{N}), \mathrm{N}=\mathrm{L1}, \mathrm{L2}) & 000 \\ \mathrm{MR} \ \mathrm{RTE} \ (6,680) \ (\mathrm{TYPE}(\mathrm{N}), \mathrm{N}=\mathrm{L1}, \mathrm{L2}) & 000 \\ \mathrm{MR} \ \mathrm{RTE} \ (6,680) \ (\mathrm{TYPE}(\mathrm{N}), \mathrm{N}=\mathrm{L1}, \mathrm{L2}) & 000 \\ \mathrm{IF} \ (\mathrm{A}(\mathrm{I}, \mathrm{J}), \mathrm{EQ}, 0) \ \mathrm{GO} \ \mathrm{TO} \ 90 & 000 \\ \mathrm{K}=\mathrm{K}+\mathrm{1} & 000 \\ \mathrm{ID} \ \mathrm{MR} \ \mathrm{ITE} \ (6,690) \ \mathrm{ID} \ \mathrm{GO} \ \mathrm{TO} \ 90 & 000 \\ \mathrm{ID} \ \mathrm{ID} \ \mathrm{K}=\mathrm{LI} \ \mathrm{ID} \$	0003530 0003540 0003550 0003550 0003550 0003580 0003590 0003610 0003610 0003620 0003620 0003630 0003650 0003650 0003660 0003650 0003670 0003710 0003710 0003720 0003730 0003740 0003750 0003760
70 L=L+1 000 IF (NN(L).EQ.0) GO TO 70 000 M=H+NN(L) 000 L2=L*7 000 L1=L2=6 000 WRITE (6,600) (TYPE(N),N=L1,L2) 000 80 K=0 000 DO 90 J=1,JJ 000 IF (A(I,J).EQ.0) GO TO 90 000 K=K+1 000 IDT (K)=IDX(J) 000 IAT (K)=A(I,J) 000 90 CONTINUE 000 100 WRITE (6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 000 C 000 C 0UTPUTS PERCENTAGES 000 ENTRY OUTPC(TOL) 000 WRITE (6,730) 000 WRITE (6,710) NAME(IDX(J)) 000 WRITE (0,710) 000 W	0003540 0003550 0003560 0003570 0003580 0003590 0003600 0003610 0003620 0003620 0003630 0003640 0003650 0003650 0003660 0003670 0003700 0003710 0003710 0003720 0003740 0003750 0003760
IF (NN(L).EQ.0) GO TO 70 M=M+NN(L) 000 L2=L*7 000 U1=L2-6 000 WRITE (6,600) WRITE (6,680) (TYPE(N),N=L1,L2) 000 00 90 J=1,JJ 000 00 00 J=1,JJ 000 00 00 J=1,JJ 000 00 K=K+1 000 100 WRITE (6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 000 00 CONTINUE 000 00 CONTINUE 00 C	0003550 0003560 0003570 0003580 0003590 0003600 0003610 0003620 0003630 0003630 0003640 0003650 0003650 0003650 0003670 0003700 0003710 0003720 0003720 0003740 0003750 0003760
N=H+NN(L)       000         L2=L*7       000         U1=L2-6       000         WRITE (6,600)       000         WRITE (6,680) (TYPE(N),N=L1,L2)       000         80       K=0       000         D0 90 J=1,JJ       000         IF (A(I,J).EQ.0) GO TO 90       000         K=K+1       000         IDT (K) =IDX (J)       000         IDT (K) =A(I,J)       000         00       WRITE (6,690) IDY (I), C(I), GC (I), GK (I), (NAME (IDT (J)), IAT (J), J=1, K)         000       RETURN       000         001       WRITE (6,690) IDY (I), C(I), GC (I), GK (I), (NAME (IDT (J)), IAT (J), J=1, K)       000         001       WRITE (6,730)       000         001       JJ=NNN       000         WRITE (6,710)       000       000         WRITE (6,710)       000 <td< td=""><td>0003560 0003570 0003590 0003600 0003610 0003620 0003620 0003620 0003620 0003650 0003650 0003660 0003670 0003700 0003710 0003710 0003720 0003750 0003760 0003770</td></td<>	0003560 0003570 0003590 0003600 0003610 0003620 0003620 0003620 0003620 0003650 0003650 0003660 0003670 0003700 0003710 0003710 0003720 0003750 0003760 0003770
$\begin{array}{c} L2=L^{*,7} & 0000\\ L1=L2=6 & 0000\\ WRITE (6,600) & 0000\\ WRITE (6,680) (TYPE(N),N=L1,L2) & 0000\\ D0 90 J=1,JJ & 0000\\ D0 90 J=1,JJ & 0000\\ F=K+1 & 0000\\ IDT (K)=IDX (J) & 0000\\ IAT (K)=A(I,J) & 0000\\ 00000\\ IOW WRITE (6,690) IDY (I),C(I),GC(I),GK (I), (NAME (IDT (J)),IAT (J),J=1,K) & 000\\ 000000\\ C & 00TPUTS PERCENTAGES & 0000\\ ENTRY OUTPC (TOL) & 0000\\ JJ=NNN & 0000\\ WRITE (6,730) & 0000\\ WRITE (6,730) & 0000\\ WRITE (6,730) & 0000\\ WRITE (6,700) NAME (IDX (J)) & 0000\\ WRITE (6,710) NAME (IDX (J)) & 0000\\ WRITE (0,710) & 0000\\ WR$	0003570 0003580 0003590 0003600 0003610 0003620 0003630 0003630 0003650 0003650 0003660 0003670 0003700 0003710 0003710 0003720 0003730 0003740 0003750 0003760
$ \begin{array}{c} L2=L*7 \\ L1=L2-6 \\ WRITE (6,600) \\ WRITE (6,680) (TYPE(N),N=L1,L2) \\ 80  K=0 \\ D0 \; 90 \; J=1, JJ \\ IF (A(I,J).EQ.0) \; GO \; TO \; 90 \\ C=K+1 \\ IDT (K) = IDX (J) \\ IAT (K) = A(I,J) \\ 90  CONTINUE \\ 100  WRITE (6,690) \; IDY (I), C(I), GC (I), GK (I), (NAME (IDT (J)), IAT (J), J=1,K) \\ 000 \\ C \\$	0003570 0003580 0003590 0003600 0003610 0003620 0003630 0003630 0003650 0003650 0003660 0003670 0003700 0003710 0003710 0003720 0003730 0003740 0003750 0003760
L1=L2-6 WRITE (6,600) WRITE (6,680) (TYPE(N),N=L1,L2) 80 R=0 D0 90 J=1,JJ IF (A(I,J).EQ.0) GO TO 90 K=K+1 IDT(K)=IDX(J) IDT(K)=IDX(J) IDT(K)=IDX(J) IDT(K)=A(I,J) 90 CONTINUE 100 WRITE(6,690) IDY(I),C(I),GC(I),GK(I),(NAHE(IDT(J)),IAT(J),J=1,K) 000 C C 001PUTS PERCENTAGES ENTRY OUTPC(TOL) JJ=NNN WRITE (6,730) DO 110 J=1,JJ WRITE (6,710) NAME(IDX(J)) WRITE (6,710) NAME(IDX(J)) 000 WRITE (6,710) NAME(IDX(J)) V=T(J) IF(NN(3).EQ.0) GO TO 140 I1=NN(1)+NN(2)+1 12=NN(1)+NN(2)+N(3) 000 V=V-A(I,J)*C(I) 150 CONTINUE 160 CONTINUE 172-NN(1)+NN(2) 160 CONTINUE 172-NN(1)+NN(2)	0003580 0003590 0003600 0003610 0003620 0003630 0003630 0003650 0003650 0003660 0003670 0003700 0003710 0003710 0003720 0003740 0003750 0003760
WRITE (6,600)         000           WRITE (6,680) (TYPE(N),N=L1,L2)         000           80         K=0         000           D0 90 J=1,JJ         000           IF (A(I,J).EQ.0) GO TO 90         000           K=K+1         000           IDT(K)=IDX(J)         000           IDT(K)=IDX(J)         000           IAT(K)=A(I,J)         000           90         CONTINUE         000           100         WRITE (6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K)         000           C         000         000           C         000         000           DO 110 J=1,JJ         000           MRITE (6,730)         000           DO 110 J=1,JJ         000           WRITE (6,710) NAME(IDX(J))         000           WRITE (6,710) NAME(IDX(J))         000           WRITE (6,710) NAME(IDX(J))         000           V=T(J)         000           IF(NN(3).EQ.0) GO TO 140         000           II=NN(1)+NN(2)+NN(3)         000           DO 150 I=I1,I2         000           V=V=A(I,J)*C(I)         000           I40         CONTINUE         000           II=1         000 <td< td=""><td>0003590 0003600 0003610 0003620 0003630 0003650 0003650 0003660 0003670 0003680 00033700 0003710 0003710 0003720 0003740 0003750 0003760</td></td<>	0003590 0003600 0003610 0003620 0003630 0003650 0003650 0003660 0003670 0003680 00033700 0003710 0003710 0003720 0003740 0003750 0003760
WRITE (6,680) (TYPE(N),N=L1,L2)       000         80       K=0       000         D0 90 J=1,JJ       000         IF (A(I,J).EQ.0) GO TO 90       000         K=K+1       000         IDT (K)=IDX (J)       000         IAT (K)=A(I,J)       000         90       CONTINUE       000         100       WRITE (6,690) IDY (I), C(I), GC (I), GK (I), (NAME (IDT (J)), IAT (J), J=1, K)       000         000       COUTPUTS PERCENTAGES       000         ENTRY OUTPC (TOL)       000       000         JJ=NNN       000       000         WRITE (6,730)       000       000         WRITE (6,600)       000       000         WRITE (6,710) NAME (IDX (J))       000       000         V=T (J)       000       000         IF (NN (3).EQ.0) GO TO 140       000       000         II =NN (1) +NN (2) +1       000       000         I2=NN (1) +NN (2) +1       000       000         I2=NN (1) +NN (2) +1       000       000         I2=NN (1) +NN (2) +1       000       000         I40       CONTINUE       000         I40       CONTINUE       000         I2=NN (1) +NN (2)       0	0003600 0003610 0003620 0003630 0003650 0003650 0003660 0003670 0003690 0003700 0003710 0003720 0003720 0003740 0003750 0003760
80       R=0       000         DO 90 J=1, JJ       000         IF (A(I, J).EQ.0) GO TO 90       000         RK+1       000         IDT(K)=IDX(J)       000         IAT(K)=A(I, J)       000         90       CONTINUE       000         100       WRITE(6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K)       000         000       RETURN       000         C       000       000         C UUTPUTS PERCENTAGES       000         ENTRY OUTPC(TOL)       000         JJ=NNN       000         WRITE (6,730)       000         DO 110 J=1,JJ       000         WRITE (6,600)       000         WRITE (6,710) NAME(IDX(J))       000         V=T(J)       000         IF(NN(3).EQ.0) GO TO 140       000         I1=NN(1)+NN(2)+NN(3)       000         DO 150 I=11,I2       000         V=V-A(I,J)*C(I)       000         140       000         IF(V.EQ.0) GO TO 110       000         II=NN(1)+NN(2)       000         II=1       000         II=1,1       000         II       000       000         IF(	0003610 0003620 0003630 0003650 0003650 0003670 0003670 0003690 0003700 0003710 0003720 0003730 0003740 0003750 0003760
DO 90 J=1, JJ IF (A(I, J).EQ.0) GO TO 90 K=K+1 OOC IDT(K)=IDX(J) IAT(K)=A(I,J) 90 CONTINUE 100 WRITE(6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 000 C C OUTPUTS PERCENTAGES ENTRY OUTPC(TOL) JJ=NNN WRITE (6,730) DO 110 J=1,JJ WRITE (6,710) NAME(IDX(J)) WRITE (6,710) NAME(IDX(J)) V=T(J) IF(NN(3).EQ.0) GO TO 140 II=NN(1)+NN(2)+1 I2=NN(1)+NN(2)+1 I2=NN(1)+NN(2) V=V-A(I,J)*C(I) 150 CONTINUE 140 CONTINUE 140 CONTINUE 140 CONTINUE 141 CONTINUE 141 CONTINUE 142 CONTINUE 144 CONTINUE 145 CONTINUE 146 CONTINUE 147 CONTINUE 147 CONTINUE 148 CONTINUE 149 CONTINUE 140 CONT	0003620 0003630 0003640 0003650 0003650 0003670 0003670 0003700 0003710 0003720 0003730 0003740 0003750 0003760
IF (A(I,J).EQ.0) GO TO 90 K=K+1 IDT(K)=IDX(J) IAT(K)=A(I,J) 90 CONTINUE 100 WRITE(6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K) 000 C C OUTPUTS PERCENTAGES ENTRY OUTPC(TOL) JJ=NNN WRITE (6,730) DO 110 J=1,JJ 000 WRITE (6,710) NAME(IDX(J)) V=T(J) IF(NN(3).EQ.0) GO TO 140 11=NN(1)+NN(2)+1 12=NN(1)+NN(2)+NN(3) DO 150 I=11,I2 V=V-A(I,J)*C(I) 150 CONTINUE 140 CONTINUE 140 CONTINUE 12=NN(1)+NN(2) DD 160 IL=1,2 DD 160 IL=1,2 DD 160 TO 170 000 000 000 000 000 000 000	00003630 00003640 00003650 00003670 00003670 00003690 00003700 00003710 00003720 00003730 00003740 00003750 00003760
K=K+1       000         IDT (K) = IDX (J)       000         IAT (K) = A(I, J)       000         90       CONTINUE       000         100       WRITE (6,690) IDY (I), C (I), GC (I), GK (I), (NAME (IDT (J)), IAT (J), J=1, K)       000         C       000       000         C       000       000         JUNN       000       000         WRITE (6,730)       000         DO 110 J=1, JJ       000         WRITE (6,710) NAME (IDX (J))       000         WRITE (6,710) NAME (IDX (J))       000         WRITE (6,710) NAME (IDX (J))       000         V=T (J)       000         IF (NN (3).EQ.0) GO TO 140       000         I1=NN (1)+NN (2)+1       000         I2=NN (1)+NN (2)+1       000         I2=NN (1)+NN (2)+1       000         I0       150 I=I1, I2       000         V=V=A (I, J) *C (I)       000         140       CONTINUE       000         IF (V.EQ.0) GO TO 110       000         I1=1       000         I2=NN (1)+NN (2)       000         DO 160 LL=1, 2       000         IF (LL.EQ.1) GO TO 170       000	00003640 00003650 00003670 00003680 00003690 00003700 00003710 00003720 00003730 00003740 00003750 00003760
IDT (K) = IDX (J) IAT (K) = A(I, J) 90 CONTINUE 100 WRITE (6,690) IDY (I), C(I), GC (I), GK (I), (NAME (IDT (J)), IAT (J), J=1, K) 000 RETURN 000 C C 0UTPUTS PERCENTAGES 000 WRITE (6,730) D0 110 J=1, JJ 000 WRITE (6,600) WRITE (6,600) WRITE (6,600) WRITE (6,710) NAME (IDX (J)) V=T (J) IF (NN (3).EQ.0) GO TO 140 11=NN (1)+NN (2)+1 12=NN (1)+NN (2)+1 12=NN (1)+NN (3) D0 150 CONTINUE 140 CONTINUE 150 CONTINUE 140 CONTINUE 150 CONTINUE 1	0003650 0003660 0003670 0003690 0003700 0003710 0003720 0003730 0003740 0003750 0003760 0003770
LAT (K) = A (I, J) 90 CONTINUE 100 WRITE (6,690) IDY (I), C (I), GC (I), GK (I), (NAME (IDT (J)), IAT (J), J=1, K) 000 C C 0000 C 0000 UTPUTS PERCENTAGES ENTRY OUTPC (TOL) JJ=NNN WRITE (6,730) DO 110 J=1, JJ WRITE (6,600) WRITE (6,600) WRITE (6,610) NAME (IDX (J)) V=T (J) IF (NN (3). EQ. 0) GO TO 140 11=NN (1)+NN (2)+1 12=NN (1)+NN (2)+1 12=NN (1)+NN (3) DO 150 I=11, I2 V=V-A (I, J) *C (I) 150 CONTINUE 140 CONTINUE 140 CONTINUE 140 CONTINUE 150 CON	00003660 0003670 0003680 00003690 00003700 00003710 00003720 00003730 00003740 00003750 00003760
90       CONTINUE       000         100       WRITE(6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K)       000         000       RETURN       000         00       000       000         C       000         C UTPUTS PERCENTAGES       000         DJ=NNN       000         WRITE (6,730)       000         D0 110 J=1,JJ       000         WRITE (6,600)       000         WRITE (6,710) NAME(IDX(J))       000         WRITE (6,710) NAME(IDX(J))       000         V=T(J)       000         IF (NN (3).EQ.0) GO TO 140       000         I1=NN (1)+NN (2)+1       000         DO 150 I=I1,I2       000         V=V-A(I,J)*C(I)       000         150 CONTINUE       000         IF(N (1)+NN (2)       000         IF (V.EQ.0) GO TO 110       000         If (V.EQ.0) GO TO 110       000         If (-1)       000         I2=NN (1)+NN (2)       000         DO 160 LL=1,2       000         IF (LL.EQ.1) GO TO 170       000	00003670 00003680 00003690 00003700 00003710 00003720 00003730 00003740 00003750 00003760 00003770
90       CONTINUE       000         100       WRITE(6,690) IDY(I),C(I),GC(I),GK(I),(NAME(IDT(J)),IAT(J),J=1,K)       000         000       RETURN       000         00       000       000         C       000         C UTPUTS PERCENTAGES       000         DJ=NNN       000         WRITE (6,730)       000         D0 110 J=1,JJ       000         WRITE (6,600)       000         WRITE (6,710) NAME(IDX(J))       000         WRITE (6,710) NAME(IDX(J))       000         V=T(J)       000         IF (NN (3).EQ.0) GO TO 140       000         I1=NN (1)+NN (2)+1       000         DO 150 I=I1,I2       000         V=V-A(I,J)*C(I)       000         150 CONTINUE       000         IF(N (1)+NN (2)       000         IF (V.EQ.0) GO TO 110       000         If (V.EQ.0) GO TO 110       000         If (-1)       000         I2=NN (1)+NN (2)       000         DO 160 LL=1,2       000         IF (LL.EQ.1) GO TO 170       000	00003680 0003690 0003700 0003710 00003720 00003730 00003740 00003750 00003760 00003770
100       WRITE(6,690)       IDY(I), C(I), GC(I), GK(I), (NAME(IDT(J)), IAT(J), J=1, K)       000         RETURN       000         C       000         C       000         ENTRY OUTPC(TOL)       000         JJ=NNN       000         WRITE (6,730)       000         D0 110 J=1,JJ       000         WRITE (6,710)       NAME(IDX(J))         WRITE (6,710)       000         WRITE (6,710)       000         WRITE (6,710)       000         WRITE (6,710)       000         I1=NN(1)+NN(2)+1       000         I2=NN(1)+NN(2)+1       000         I2=NN(1)+NN(2)       000         D0 150 I=I1, I2       000         V=V-A(I, J)*C(I)       000         140       CONTINUE       000         I2=NN(1)+NN(2)       000         I1=1       000         I2=NN(1)+NN(2)       000         I1=1       000         I2=NN(1)+NN(2)       000         D0 160 LL=1,2       000         IF (LL.EQ.1)       000	00003680 0003690 0003700 0003710 00003720 00003730 00003740 00003750 00003760 00003770
RETURN       000         C       000         C OUTPUTS PERCENTAGES       000         JJ=NNN       000         JJ=NNN       000         WRITE (6,730)       000         DO 110 J=1, JJ       000         WRITE (6,600)       000         WRITE (6,600)       000         WRITE (6,710) NAME (IDX(J))       000         V=T(J)       000         If (NN (3) .EQ.0) GO TO 140       000         I1=NN (1) +NN (2) +1       000         DO 150 I=I1, I2       000         V=V-A(I,J)*C(I)       000         150 CONTINUE       000         If (V.EQ.0) GO TO 110       000         II=1       000         I2=NN (1) +NN (2)       000         II=1       000 <td>00003690 0003700 0003710 00003720 00003730 00003740 00003750 00003760 00003770</td>	00003690 0003700 0003710 00003720 00003730 00003740 00003750 00003760 00003770
C 0000 C 0UTPUTS PERCENTAGES 0000 ENTRY 0UTPC (TOL) 0000 JJ=NNN 0000 WRITE (6,730) 0000 WRITE (6,730) 0000 WRITE (6,600) 0000 WRITE (6,710) NAME (IDX (J)) 0000 WRITE (6,710) NAME (IDX (J)) 0000 IF (NN (3).EQ.0) GO TO 140 0000 II=NN (1)+NN (2)+1 0000 I1=NN (1)+NN (2)+1 0000 I2=NN (1)+NN (2)+NN (3) 0000 DO 150 I=I1,I2 0000 V=V-A(I,J)*C(I) 0000 I50 CONTINUE 0000 I40 CONTINUE 0000 IF (V.EQ.0) GO TO 110 0000 II=1 0000 II=1 0000 II=1 0000 II=1 0000 II=1 0000 II=1 0000 II=1 0000 II=1 00000 II=1 0000 II=1 00000 II=1 0000 II=1 00000 II=1 00000 II=1 00000 II=1 00000 II=1 000000 II=1 000000 II=1 000000000 II=1 0000000000000000000000000000	00003700 00003710 00003720 00003730 00003740 00003750 00003760 00003770
C OUTPUTS PERCENTAGES 000 ENTRY OUTPC (TOL) 000 JJ=NNN 000 WRITE (6,730) 000 WRITE (6,730) 000 WRITE (6,600) 000 WRITE (6,710) NAME (IDX(J)) 000 WRITE (6,710) NAME (IDX(J)) 000 IF (NN (3) .EQ.0) GO TO 140 000 II=NN (1) +NN (2) +1 000 I2=NN (1) +NN (2) +1 000 DO 150 I=I1, I2 000 V=V-A (I, J) *C (I) 000 I50 CONTINUE 000 I40 CONTINUE 000 IF (V.EQ.0) GO TO 110 000 II=1 0000 II=1 0000 II=1 0000 II=1	00003710 00003720 00003730 00003740 00003750 00003760 00003770
ENTRY OUTPC (TOL) 000 JJ=NNN 000 WRITE (6,730) 000 WRITE (6,600) 000 WRITE (6,600) 000 WRITE (6,710) NAME (IDX(J)) 000 V=T(J) 000 IF (NN(3).EQ.0) GO TO 140 000 I1=NN(1)+NN(2)+1 000 I2=NN(1)+NN(2)+1 000 DO 150 I=I1,I2 000 V=V-A(I,J)*C(I) 000 150 CONTINUE 000 IF(V.EQ.0) GO TO 110 000 IF(V.EQ.0) GO TO 110 000 II=1 000 II=	00003720 00003730 00003740 00003750 00003760 00003770
JJ=NNN       000         WRITE (6,730)       000         DO 110 J=1,JJ       000         WRITE (6,600)       000         WRITE (6,710) NAME (IDX(J))       000         V=T(J)       000         IF (NN (3).EQ.0) GO TO 140       000         II=NN (1) +NN (2) +1       000         DO 150 I=I1,I2       000         V=V-A(I,J) *C(I)       000         150 CONTINUE       000         IF (V.EQ.0) GO TO 110       000         II=1       000         III=1       000         III	00003730 00003740 00003750 00003760 00003770
WRITE (6,730)       000         DO 110 J=1, JJ       000         WRITE (6,600)       000         WRITE (6,710) NAME (IDX(J))       000         If (NN(3).EQ.0) GO TO 140       000         IF (V.EQ.0) GO TO 110       000         IF (V.EQ.0) GO TO 110       000         II=1       000         IZ=NN(1)+NN(2)       000         DO 160 LL=1,2       000         IF (LL.EQ.1) GO TO 170       000	00003740 00003750 00003760 00003770
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	00003750 00003760 00003770
WRITE (6,600)       000         WRITE (6,710) NAME(IDX(J))       000         V=T(J)       000         IF(NN(3).EQ.0) GO TO 140       000         I1=NN(1)+NN(2)+1       000         I2=NN(1)+NN(2)+NN(3)       000         DO 150 I=I1,I2       000         V=V-A(I,J)*C(I)       000         150 CONTINUE       000         140 CONTINUE       000         IF(V.EQ.0) GO TO 110       000         II=1       000         001       000         I2=NN(1)+NN(2)       000         IF(V.EQ.0) GO TO 110       000         II=1       000 <td< td=""><td>00003760</td></td<>	00003760
WRITE $(6,710)$ NAME $(IDX(J))$ 000         V=T(J)       000         IF (NN (3) .EQ.0) GO TO 140       000         I1=NN (1) +NN (2) +1       000         I2=NN (1) +NN (2) +NN (3)       000         DO 150 I=I1, I2       000         V=V-A(I, J) *C(I)       000         150 CONTINUE       000         140 CONTINUE       000         IF (V.EQ.0) GO TO 110       000         II=1       000         I2=NN (1) +NN (2)       000         II=1       000         I2=NN (1) +NN (2)       000         II=1       000         III=1       000	00003770
$ \begin{array}{c} v = T (J) & 000 \\ IF (NN (3) . EQ. 0) GO TO 140 & 000 \\ II = NN (1) + NN (2) + 1 & 000 \\ I2 = NN (1) + NN (2) + NN (3) & 000 \\ DO 150 I = I1, I2 & 000 \\ v = v - A (I, J) * C (I) & 000 \\ 150 CONTINUE & 000 \\ 140 CONTINUE & 000 \\ IF (v. EQ. 0) GO TO 110 & 000 \\ II = 1 & 000 \\ I1 = 1 & 000 \\ I2 = NN (1) + NN (2) & 000 \\ DO 160 LL = 1, 2 & 000 \\ IF (LL. EQ. 1) GO TO 170 & 000 \\ \end{array} $	
$ \begin{array}{c} v = T (J) & 000 \\ IF (NN (3) . EQ. 0) GO TO 140 & 000 \\ II = NN (1) + NN (2) + 1 & 000 \\ I2 = NN (1) + NN (2) + NN (3) & 000 \\ DO 150 I = I1, I2 & 000 \\ v = v - A (I, J) * C (I) & 000 \\ 150 CONTINUE & 000 \\ 140 CONTINUE & 000 \\ IF (v. EQ. 0) GO TO 110 & 000 \\ II = 1 & 000 \\ I1 = 1 & 000 \\ I2 = NN (1) + NN (2) & 000 \\ DO 160 LL = 1, 2 & 000 \\ IF (LL. EQ. 1) GO TO 170 & 000 \\ \end{array} $	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0003790
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0003800
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	0003810
V=V-A(I,J)*C(I)       000         150       CONTINUE       000         140       CONTINUE       000         IF(V.EQ.0)       GO TO 110       000         II=1       000       000         I2=NN(1)+NN(2)       000       000         DO 160       LL=1,2       000         IF (LL.EQ.1)       GO TO 170       000	
150       CONTINUE       000         140       CONTINUE       000         IF(V.EQ.0)       GO TO 110       000         II=1       000         I2=NN(1)+NN(2)       000         D0       160       LL=1,2         IF (LL.EQ.1)       GO TO 170       000	0003820
140       CONTINUE       000         IF(V.EQ.0) GO TO 110       000         I1=1       000         I2=NN(1)+NN(2)       000         D0 160 LL=1,2       000         IF (LL.EQ.1) GO TO 170       000	0003830
IF (V.EQ.0) GO TO 110       000         I1=1       000         I2=NN(1)+NN(2)       000         DO 160 LL=1,2       000         IF (LL.EQ.1) GO TO 170       000	0003840
I1=1       000         I2=NN(1)+NN(2)       000         D0 160 LL=1,2       000         IF (LL.EQ.1) GO TO 170       000	0003850
I2=NN (1)+NN (2)       000         D0 160 LL=1,2       000         IF (LL.EQ.1) GO TO 170       000	0003855
DO 160 LL=1,2 000 IF (LL.EQ.1) GO TO 170 000	0003860
IF (LL.EQ.1) GO TO 170 000	0003870
IF (LL.EQ.1) GO TO 170 000	0003880
	0003890
	0003900
	0003910
	00003920
	00003930
	00003940
	0003950
PC-PC*100 000	0003960
L-0 000	0003970
	0003980
	0003980
	0003990
	00003990 00004000
	00003990 00004000 00004010
	00003990 00004000 00004010 00004020
	00003990 00004000 00004010 00004020 00004030
WRITE (6,720) PC, IDY(I), (NAME(IDT(M)), IAT(M), M=1, L) 000	00003990 00004000 00004010 00004020
	00003990 00004000 00004010 00004020 00004030
120 CONTINUE 000	00003990 00004000 00004010 00004020 00004030 00004040
120 CONTINUE 000 160 CONTINUE 000	00003990 00004000 00004010 00004020 00004030 00004040 00004050

		107
	RETURN	00004080
С		00004090
600	FORMAT('0')	00004100
610	FORMAT('0',' ID',10X,'X',4X,'LOGX',10X,'T',5X,'COMPONENTS')	00004110
620	FORMAT ('0', 15, 2X, 1PE9.2, 2X, 0PF6.2, 2X, 1PE9.2, 5X, A4)	00004120
630	FORMAT('0',' ID',4X,'LOGK',5X,'SPECIES: TYPE ',7A4)	00004130
640	FORMAT('0', 15, 2X, 0PF6.2, 5X, 4(A4, 1X, 13, 4X))	00004140
650	FORMAT('1',' OUTPUT DATA: ITERATIONS = ',I3)	00004150
660	FORMAT('0',' ID',10X,'X',4X,'LOGX',10X,'T',10X,'Y',5X,	00004160
	1'SPECIES')	00004170
670	FORMAT('0', 15, 2X, 1PE9.2, 2X, 0PF6.2, 2(2X, 1PE9.2), 5X, A4)	00004180
680	FORMAT('0',' ID',10X,'C',4X,'LOGC',4X,'LOGK',5X,	00004190
000	1'SPECIES: TYPE ',7A4)	00004200
(00		00004210
690	FORMAT('0', I5, 2X, 1PE9.2, 2(2X, 0PF6.2), 5X, 4(A4, 1X, I3, 4X))	
700	FORMAT('1',' INPUT DATA')	00004220
710	FORMAT ('0',A4)	00004230
720	FORMAT ('0', 7X, F6.1, 5X, 'PERCENT BOUND IN SPECIES #', 15, 3X, 4(A4, 1	X,00004240
	113,4%))	00004250
730	FORMAT ('1', ' PERCENTAGE DISTRIBUTION OF COMPONENTS')	00004260
	END	00004270
	SUBROUTINE SIMQ(Z,Y,N,NXDIM)	00004280
_	DIMENSION Z (NXDIM, NXDIM), Y (NXDIM)	00004290
С	PROVISION FOR N=1	00004300
	IF(N.NE.1) GO TO 50	00004310
	Y(1) = Y(1) / Z(1, 1)	00004320
	RETURN	00004330
50	CONTINUE	00004340
С	ELEMENT OF ELIMINATION	00004350
	N1=N-1	00004360
	DO 10 M-1,N1	00004370
	ZMAX=0.	00004380
	IMAX=0	00004390
с	FIND MAX OF COLUMN	00004400
L		00004410
	DO 20 I-H,N	
	IF (ABS (Z(I,M)).LE.ZMAX) GO TO 20	00004420
	IMAX=I	00004430
	ZMAX=ABS(Z(I,M))	00004440
20	CONTINUE	00004450
С	ERROR RETURN	00004460
	IF(IMAX.NE.0) GO TO 30	00004470
	CALL ERROR (8)	00004480
30	CONTINUE	00004490
C	ROW INTERCHANGE	00004500
C	IF (IMAX.EQ.M) GO TO 35	00004510
	V=Y (M)	00004520
	Y(M) = Y(IMAX)	00004530
	Y (IMAX) -V	00004540
	DO 40 J-M,N	00004550
	V=Z (M, J)	00004560
	Z(M, J) = Z(IMAX, J)	00004570
	Z(IMAX, J)=V	00004580
40	CONTINUE	00004590
35	CONTINUE	00004600
C		00004610
L	DIAGONALIZE	
	M1=M+1	00004620
	DO 70 I=M1,N	00004630
	V=Z(I,M)/Z(M,M)	00004640
	Y(I) = Y(I) - V + Y(M)	00004650
	DO 70 J=M,N	00004660
	Z(I,J)=Z(I,J)-V*Z(M,J)	00004670

		100
70	CONTINUE	00004680
10	CONTINUE	00004690
C	BACK SUBSTITUTE	00004700
	Y(N) = Y(N) / Z(N, N)	00004710
	N1=N-1	00004720
	DO 100 K=1,N1	00004730
	I=N-K	00004740
	II=I+1	00004750
	DO 90 J=I1.N	00004760
90	Y(I) = Y(I) - Y(J) * Z(I, J)	00004770
100	Y(I) = Y(I) / Z(I, I)	00004780
100	RETURN	00004780
	END	00004800
	SUBROUTINE SOLID	00004810
	COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN (6), NNN	00004820
	COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),	00004830
-	1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400)	00004840
С		00004850
	IF(NN(3)+NN(4).EQ.0) RETURN	00004860
	LL = NN(3) + NN(4)	00004870
	IO=NN(1)+NN(2)+NN(3)+NN(4)+1	00004880
	JO=NNN+1	00004890
	DO 601 L=1,LL	00004900
	10=10-1	00004910
	J0=J0-1	00004920
C FI	ND JEXC FOR IO	00004930
	D0 602 $J=1, J0$	00004940
	IF (ABS(A(I0, J0-(J-1))).GT.0.001) GO TO 603	00004950
602	CONTINUE	. 00004960
(02	CALL ERROR (6)	00004970
603	JEXC=JO-(J-1)	00004980
	CALL EXCOL (JEXC, JO)	00004990
	NXS=J0-1	00005000
C 140	NCS=IO-1	00005010
C MU	DIFY A, B, T	00005020
	DO 604 I=1,NCS	00005030
604	DO 604 J=1,NXS $A(T, T) = A(T, T) =$	00005040
004	A(I, J) = A(I, J) - A(I0, J) * A(I, J0) / A(I0, J0)	00005050 00005060
605	DO 605 J=1,NXS T(I) = T(I) = A(IO, I) + T(IO) (A(IO, IO))	
605	T(J) = T(J) - A(I0, J) + T(J0) / A(I0, J0)	00005070
606	D0 606 I=1,NCS GK(I)=GK(I)-A(I,J0)*GK(I0)/A(I0,J0)	00005080
606		00005090 00005100
601	CONTINUE	
С		00005110
	RETURN	00005120
	END	00005130
	SUBROUTINE SOLIDX (KK)	00005140
	COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN (6), NNN	00005150
	COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),	00005160
~	1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400)	00005170
С	<b>FT</b> = 0	00005180
~	KK=0	00005190
С	TE (NTV (2) + NTV (1) TO (2) CO TO (20)	00005200
	IF (NN (3) +NN (4).EQ.0) GO TO 470	00005210
	LL = NN(3) + NN(4)	00005220
	II = NN(1) + NN(2)	00005230
	10 = NN(1) + NN(2) + 1	00005240
	JO=NNN-NN(3)-NN(4)+1	00005250
СМ	JJ=NNN OLE BALANCE MINUS SOLIDS	00005260 00005270

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.

DO 440 J=J0,JJ 00005280 Y(J) = -T(J)00005290 DO 440 I=1,II 00005300 Y(J) = Y(J) + A(I, J) \* C(I)00005310 440 CONTINUE 00005320 C AMOUNT OF SOLIDS 00005330 DO 460 L=1,LL 00005340 C(10) = -Y(J0) / A(10, J0)00005350 GC(I0) = ALOG10(ABS(C(I0)))00005360 DO 450 K=J0,JJ 00005370 Y(K) = Y(K) + A(I0, K) \* C(I0)00005380 450 CONTINUE 00005390 C UNMODIFY A, B, T, GX, X 00005400 00005410 NXS=J0-1 NCS=10-1 00005420 V=GK(10) 00005430 DO 571 J=1,NXS 00005440 571 00005450 V=V+A(IO, J)\*GX(J)00005460 GX(J0) = -V/A(I0, J0)X(J0) = 10.\*\*(GX(J0))00005470 DO 61 I=1,NCS 00005480 00005490 DO 61 J=1,NXS 61 A(I,J)=A(I,J)+A(I0,J)\*A(I,J0)/A(I0,J0)00005500 DO 62 J=1,NXS 00005510 62 T(J) = T(J) + A(I0, J) \* T(J0) / A(I0, J0)00005520 00005530 DO 63 I=1,NCS GK(I) = GK(I) + A(I, J0) + GK(I0) / A(I0, J0)63 00005540 10=10+1 00005550 J0=J0+1 00005560 460 CONTINUE 00005570 470 CONTINUE 00005580 C SOLUBILITY PRODUCTS 00005590 IF(NN(5)+NN(6).EQ.0) GO TO 220 00005600 10=NN(1)+NN(2)+NN(3)+NN(4)+100005610 II=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)+NN(6)00005620 JJ=NNN 00005630 DO 210 I=I0,II 00005640 00005650 V=GK(I)DO 200 J=1, JJ 00005660 200 00005670 V=V+A(I,J)\*GX(J)GC(I) = V00005680 C(I)=10.\*\*V 00005690 210 00005700 CONTINUE 220 CONTINUE 00005710 С 00005720 CHECK FOR DISSOLUTION 00005730 С 00005740 С 00005750 IF (NN (4).EQ.0) GO TO 45 00005760 IMIN=0 00005770 VMIN=0 10=NN(1)+NN(2)+NN(3)+100005780 II=NN(1)+NN(2)+NN(3)+NN(4)00005790 DO 44 I=I0,II 00005800 IF(C(I).GT.VMIN) GO TO 44 00005810 00005820 VMIN=C(I) IMIN=I 00005830 00005840 44 CONTINUE IF(IMIN.EQ.0) GO TO 45 00005850 WRITE(6,610) ITER, IDY(IMIN) 00005860 FORMAT('0',' ITERATIONS= ',13,': SOLID ',15,' DISSOLVES') 610 00005870

00005880 CALL EXROW(IMIN, II) NN(5) = NN(5) + 100005890 NN(4) = NN(4) - 100005900 KK=-1 00005910 RETURN 00005920 45 CONTINUE 00005930 С 00005940 C CHECK FOR PRECIPITATION 00005950 С 00005960 IF (NN(5).EQ.0) GO TO 22 00005970 00005980 VMAX=0 IMAX=0 00005990 10=NN(1)+NN(2)+NN(3)+NN(4)+100006000 II=NN(1)+NN(2)+NN(3)+NN(4)+NN(5)00006010 DO 47 I=I0,II 00006020 IF (GC(I).LT. VMAX) GO TO 47 00006030 VMAX=GC(I) 00006040 IMAX=I 00006050 47 CONTINUE 00006060 IF(IMAX.EQ.0) GO TO 22 00006070 WRITE(6,600) ITER, IDY(IMAX) 00006080 600 FORMAT('0',' ITERATIONS= ', I3, ': SOLID ', I5, ' PRECIPITATES') 00006090 CALL EXROW(IMAX, IO) 00006100 NN(4) = NN(4) + 100006110 NN(5) = NN(5) - 100006120 KK=1 00006130 22 CONTINUE 00006140 RETURN 00006150 END 00006160 SUBROUTINE SOLVE 00006170 COMMON/PARM/NXDIM, NYDIM, ITMAX, ITER, EPS, NN(6), NNN 00006180 COMMON/VAR/GX(30), X(30), T(30), Y(30), Z(30, 30),00006190 1C(400), GC(400), GK(400), A(400, 30), IDX(30), IDY(400)00006200 С 00006210 NC=NN(1)+NN(2)00006220 NX=NNN-NN(3)-NN(4)00006230 1000 CONTINUE 00006240 C COMPLEXES 00006250 DO 2 I=1,NC 00006260 00006270 V=GK(I)DO 3 J=1,NX 00006280 3 V=V+A(I,J)\*GX(J)00006290 GC(I) = V00006300 C(I) = 10.\*\*GC(I)00006310 00006320 2 CONTINUE C MOLE BALANCE 00006330 DO 201 J=1,NX 00006340 V = -T(J)00006350 DO 200 I=1.NC 00006360 200 V=V+A(I,J)\*C(I)00006370 Y(J) = V00006380 201 CONTINUE 00006390 C COMPUTE Z 00006400 DO 300 I=1,NX 00006410 DO 300 J=1,NX 00006420 300 Z(I,J)=000006430 DO 400 J=1,NX 00006440 DO 400 I=1,NC 00006450 00006460 DO 400 K=1,NX 400 Z(J,K) = Z(J,K) + A(I,J) \* A(I,K) \* C(I) / X(K)00006470

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C CON	IVERGENCE TEST	00006480
	DO 800 J=1,NX	00006490
	VMAX=ABS(T(J))	00006500
	DO 810 I=1,NC	00006510
	IF(ABS(A(I,J)*C(I)).LT.VMAX) GO TO 810	00006520
	VMAX=ABS(A(I,J)*C(I))	00006530
810	CONTINUE	00006540
	IF (ABS(Y(J))/VMAX.GT.EPS) GO TO 840	00006550
800	CONTINUE	00006560
	RETURN	00006570
840 ·	ITER=ITER+1	00006580
	IF(ITER.GT.ITMAX) CALL ERROR(7)	00006590
C ITE	ERATE	00006600
	CALL SIMQ(Z,Y,NX,NXDIM)	00006610
	DO 500 J=1,NX	00006620
	$\mathbf{X}(\mathbf{J}) = \mathbf{X}(\mathbf{J}) - \mathbf{Y}(\mathbf{J})$	00006630
	IF(X(J).GT.O.) GO TO 501	00006640
	X(J) = (X(J) + Y(J)) / 10.	00006650
501	GX(J) = ALOG1O(X(J))	00006660
500	CONTINUE	00006670
	GO TO 1000	00006680
	END	00006690

APPENDIX C

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#### APPENDIX C

#### Potential Migratory Activity Levels of Co-58, Sr-85, and Ce-141, for the Loading Levels Utilized in this Study, in the Event of Leachate Migration into the Surrounding Environment

- Radionuclides Considered Co-58; Half Life = 70.8 days Sr-85; Half Life = 64.8 days Ce-141; Half Life = 32.5 days

Assumptions:

- 1. Co-58, Sr-85, and Ce-141 are codisposed with municipal refuse at levels of 0.91 mCi, 1.09 mCi, and 5.78 mCi, respectively. Thus, the loading levels of Co-58, Sr-85, and Ce-141 utilized in this study, on a dry weight of refuse basis, were 1.6 x  $10^{-7}$  Ci/kg, 2.0 x  $10^{-7}$  Ci/kg, and 10.6 x  $10^{-7}$  Ci/kg, respectively.
- 2. During the period of investigation, the maximum levels of Co-58, Sr-85, and Ce-141 present in leachate from the landfill with provisions for leachate recycle (Cell B), expressed as a percent of the initial loading levels, were 12.7%, 15.7%, and 0.03%, respectively. The levels of Co-58, Sr-85, and Ce-141 present in the leachate from the landfill operated in a single pass mode were lower than corresponding levels in leachate from Cell B. Thus, the maximum levels of Co-58, Sr-85, and Ce-141 present in leachate, from a landfill receiving both municipal refuse and these radionuclides at the aforementioned loading levels, are assumed as 13%, 16%, and 0.03%, respectively.

3. Infiltration into the landfill is limited to a maximum height of 0.3 m (1 ft) above the bottom layer of the landfill.

4. The as-placed porosity of the refuse within the landfill is 0.4.

In accordance with 10 CFR Part 20, Appendix B, the maximum concentrations of Co-58, Sr-85, and Ce-141 in water above the natural background are 1.0 x  $10^{-3}$  Ci/m<sup>3</sup>, 3 x  $10^{-3}$  Ci/m<sup>3</sup>, and 3 x  $10^{-3}$  Ci/m<sup>3</sup>, respectively.

The maximum levels of Co-58, Sr-85, and Ce-141 present in the leachate are 13%, 16%, and 0.03% of the initial loading levels. Therefore, the maximum levels of Co-58, Sr-85, and Ce-141 present in the leachate are 0.12 mCi, 0.17 mCi, and 0.002 mCi, respectively. Assuming, leachate in the landfill is limited to a 0.3 m (1 ft) head over the bottom layer and an as-placed porosity value of 0.4 for the refuse, the volume of leachate contained in the landfill with provisions for leachate recycle (Cell B) is 1.11 m<sup>3</sup>. The actual volume of leachate in Cell B was  $1.32 \text{ m}^3$ .

Assuming, in the event of a liner failure the entire leachate from Cell B migrated into the surrounding environment and neglecting any dilution, the migratory activity levels of the radionuclides will be:

> Co-58:  $\frac{0.12}{1.32} \frac{\text{mCi}}{\text{m}^3} = 0.09 \text{ mCi/m}^3$ Sr-85:  $\frac{0.17}{1.32} \frac{\text{mCi}}{\text{m}^3} = 0.13 \text{ mCi/m}^3$ Ce-141:  $\frac{0.002}{1.32} \frac{\text{mCi}}{\text{m}^3} = 0.0015 \text{ mCi/m}^3$

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These values of 0.09 mCi/m<sup>3</sup>, 0.13 mCi/m<sup>3</sup>, and 0.0015 mCi/m<sup>3</sup> for Co-58, Sr-85, and Ce-141, respectively, are lower than the maximum permissible concentrations of Co-58 (1 mCi/m<sup>3</sup>), Sr-85 (3 mCi/m<sup>3</sup>), and Ce-141 (3 mCi/m<sup>3</sup>) in water above the natural background.

APPENDIX D

Sampling Date	Time Since Leachate Production Began, Days
7/27/82	0
8/19/82	13
8/16/82	20
8/24/82	28
8/30/82	34
9/07/82	41
9/14/82	48
9/21/82	55
9/27/82	61
10/04/82	68
10/11/82	75
10/18/82	82
10/20/82 11/02/82	90
11/08/82	97 103
11/15/82	103
11/22/82	110
11/29/82	124
12/07/82	132
12/14/82	139
12/27/82	152
1/07/83	163
1/12/83	168
1/18/83	174
1/25/83	181
2/08/83	196
2/17/83	205
3/03/83	219
3/15/83	231
3/26/83	241
4/08/83	254
4/14/83	260
4/20/83	266
4/29/83 5/14/83	275
7/13/83	290 350
7/23/83	350
1,25,05	500
	continued

# Table D-1. Sampling Date and Corresponding Day Since Leachate Generation Was Initiated

# Table D-1. Continued...

Sampling Date	Time Since Leachate Production Began, Days	
8/01/83 8/09/83 8/20/83 9/01/83 9/15/83 9/27/83 9/30/83 10/15/83 10/29/83 11/12/83	369 377 388 400 414 426 429 444 458 472	
11/22/83 12/07/83 12/26/83 1/19/84 2/04/84 2/18/84 3/15/84 4/15/84 5/15/84 6/15/84 7/15/84 8/15/84 10/15/84 11/15/84 12/15/84 1/15/85 2/15/85	482 497 516 540 556 570 594 625 655 686 716 747 778 808 839 869 900 931	

Time Since	Ambient Te	emperature*	Internal 1	Cemperature
Leachate Production Began,	Maximum	Minimum	Cell A	Cell B
Days	°C	°C	°C	°C
97	25.0	14.4	12	12
103	18.8	2.7	10	10
110	13.3	1.1	10	10
117	20.5	13.8	14	14
124	19.4	10.0	12	12
132	16.6	2.7	5	6
139	10.0	-3.3	4	4
152	21.6	14.4	13	13
163	15.5	2.7	7	7
168	1.1	-3.3	4	4
174	2.7	~5.5	3	3 5
181	10.0	-0.5	5	5
196	10.0	-3.3	3 5 3 8	3
205	17.7	6.6		8
219	23.8	8.3	10	10
231	23.3	8.8	14	14
241	12.7	1.6	14	14
254	19.4	15.0	14	15
260	18.3	15.0	18	18
266	14.4	-1.1	18	18
275	27.2	13.8	25	25
290	29.4	16.6	29	30
350	34.4	21.6	33	35
360	37.2	25.0	37	39
369	30.0	21.1	33	35
377	33.3	22.2	33	37
388	37.2	23.8	38	39
400	25.0	20.5	30	32
414	23.8	15.5	27	28
426	22.7	11.6	22	23
429	23.3	12.7	22	22
444	21.6	8.3	21	22
458	24.4	7.2	23	23
472	11.1	0.0	20	21
482	22.7	5.5	20	22

Table D-2. Maximum and Minimum Ambient Temperatures and Temperatures Within Cell A (Single Pass) and Cell B (Leachate Recycle)

.

Time Since	Ambient Te	emperature*	Internal	Temperature
Leachate Production	Maximum	Minimum	Cell A	Cell B
Began, Days	• <sub>C</sub>	oc	ο <sub>C</sub>	٥ <sub>C</sub>
	ŭ	ŭ	<u>~</u>	
497	7.7	-1.1	18	18
516	-8.3	-17.7	7	8
540	2.7	-6.6	5	5
556	13.8	4.4	10	13
570	21.1	5.5	20	23
594	26.1	8.3	25	26
625	22.2	8.8	22	23
655	23.3	11.1	22	25
686	32.7	21.1	30	33
716	30.0	22.2	30	33
747	31.1	21.1	32	34
778	28.3	17.7	30	30
808	26.1	18.3	28	28
839	20.5	3.3	22	22
869	20.0	9.4	20	21
900	3.8	-3.3	10	12
931	6.6	-4.4	10	11
970	22.2	8.3	18	18

Table D-2. Continued...

\*Obtained from the National Oceanic and Atmospheric Administration (NOAA), Asheville, NC

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Time Since	Moisture Cont	ent of Refuse
Leachate		ed weight of refuse)
Production		
Began,	Cell A	Cell B
Days	%	%
0	65.3	60.3
14	63.4	64.4
21	62.1	64.4
28	66.1	68.7
49	67.2	70.9
56	69.2 69.0	73.9
61 68	68.8	73.9 73.9
75	68.6	73.9
. 82	68.4	73.9
90	68.2	73.9
97	67.1	73.8
110	67.3	73.8
117	67.1	73.8
124	66.7	73.8
132	66.1	73.8
139	65.4	73.8
152	65.5	73.7
174	63.8	73.7
181	62.6	73.7
196	62.2	73.7
205	61.9	73.7 .
219	61.7	73.7
241	61.8	73.7
260	61.5	73.6
290	61.4	73.6
350	61.3	73.5
369	61.3	73.5
388	61.2	73.4
414	61.1	73.4
444	61.0	73.4
472	60.9	73.3
497	60.8	73.2
540	60.7	73.1

Table D-3.	Variations in the Moisture Content of the
	Municipal Refuse Within Cell A (Single Pass)
	and Cell B (Leachate Recycle)

#### Table D-3. Continued...

	tent of Refuse ed weight of refuse)
Cell A	Cell B
¢∕	%
60.6	73.0
60.5	73.0
60.5	73.0
60.3	72.9
60.2	72.8
60.1	72.7
60.0	72.7
59.8	72.5
	(Based on as-place Cell A % 60.6 60.5 60.5 60.3 60.2 60.1 60.0

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Time Since Leachate	Chemical O	xygen Demand
Production Began,	Cell A	Cell B
Days	mg/L	mg/L
0	15,100	13,975
13	26,956	19,468
20	28,224	16,600
28	24,120	19,620
34	26,880	17,920
48	35,276	34,138
55	40,256	38,327
61	35,526	35,200
68	26,800	21,760
75	21,280	22,080
82	19,000	25,087
90	22,450	21,185
97	21,240	18,540
103	19,228	23,859
110	18,685	24,809
117	8,333	22,333
124	6,630	24,751
132	4,397	23,874
139	2,541	25,856
152	3,680	20,210
163	4,530	18,877
181	3,811	23,762
196	3,674	24,530
205	3,661	16,744
212	3,240	23,650
219	3,716	18,583
231	2,888	21,066
241	1,803	23,644
254	2,564	21,544
260	3,716	20,654
266	2,231	18,583
279	2,894	23,500
283	3,324	17,540
350	2,656	7,614
360	1,562	3,593
369	1,708	3,978

Table D-4. Chemical Oxygen Demand (COD) of Leachate Samples From Cell A (Single Pass) and Cell B (Leachate Recycle)

Table D-4. Continued...

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Time Since	Chemical Ox	ygen Demand
Leachate Production	Cell A	Cell B
Began, Days	mg/L	mg/L
377	1,532	3,548
388	1,640	3,785
400	1,551	1,689
414	1,482	1,689
426	1,448	1,655
444	1,812	2,202
458	1,525	2,116
472	1,680	2,634
482	1,834	2,864
497	1,538	2,402
516	1,437	2,216
540	1,290	2,450
556	1,063	2,553
570	1,276	2,340
594	1,137	1,962
625	1,261	1,872
655	1,098	1,921
686	1,100	1,831
716	1,061	1,771
747	1,256	1,662
778	1,324	1,692
808	1,231	1,620
839	1,251	1,738
869	1,267	1,644
900	1,452	1,623
931	1,441	1,457
970	1,349	1,522
	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	,

Time Since	Biochemical	Oxygen Demand
Leachate Production Began,	Cell A	Cell B
Days	mg/L	mg/L
0	6,849	7,025
13	12,120	13,317
20	14,066	9,904
28	15,215	12,200
34	15,502	11,913
48	15,000	14,003
55	22,200	19,400
61	12,000	12,900
68	15,900	15,000
75	12,360	13,980
82	12,300	12,900
90	12,100	13,725
97	12,060	11,700
103	13,700	18,000
117	7,800	15,000
124	6,000	15,600
132	2,700	14,100
139	2,350	17,700
152	2,150	15,650
181	2,950	18,650
196	3,530	14,500
205	3,515	10,550
219	3,010	12,500
241	1,700	12,500
254	2,307	14,649
266	1,940	13,937
283	3,024	13,865
350	2,100	4,900
360	1,020	1,860
369	1,308	2,090
377	958	1,740
388	1,100	1,980
400	1,012	1,210
414	924	1,026
426	725	980
444	980	1,216

Table D-5. Biochemical Oxygen Demand (BOD<sub>5</sub>) of Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

continued...

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Table D-5. Continued...

Time Since Leachate	Biochemical (	Dxygen Demand
Production	Cell A	Cell B
Began, Days		
Days	mg/L	mg/L
458	815	1,370
472	905	1,495
482	925	1,612
497	705	1,465
540	625	1,250
556	465	1,165
625	255	374
686	265	352
747	339	415
778	370	389
808	356	437
839	350	364
869	380	386
900	428	421
931	373	313
970	370	342

Time Since	Total Orga	nic Carbon
Leachate Production Began,	Cell A	Cell B
Days	mg/L	mg/L
0	5,300	4,500
13	7,367	6,722
20	7,081	5,704
28	6,978	5,771
34	7,081	5,274
48	8,580	7,020
55	8,372	8,450
61	7,359	7,827
68	8,220	9,150
75	7,020	7,430
82	7,150	8,300
90	10,200	8,870
97	8,670	9,340
103	8,210	9,280
110	6,270	8,490
117	5,800	9,220
124	5,050	10,050
132	4,425	9,660
152	3,022	10,000
181	2,250	8,600
196	2,500	7,500
205	2,032	6,300
212	2,500	7,750
219	1,825	7,926
231	1,219	6,703
241	1,117	8,536
254	1,315	7,653
260	1,865	6,715
266	1,121	6,423
279	1,560	7,983
283	1,710	6,121
350	1,790	1,950
360	1,366	1,566
369	1,564	1,790
377	1,350	1,540
388	1,450	1,790

Table D-6. Total Organic Carbon (TOC) of Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

Time Since	Total Orga	nic Carbon
Leachate Production Began,	Cell A	Cell B
Days	mg/L	mg/L
400	797	980
426	907	943
444	1,095	1,275
458	975	1,065
472	1,165	1,238
482	1,210	1,289
497	853	1,306
540	709	1,029
556	829	1,302
570	1,050	1,159
594	807	853
625	741	668
655	712	835
686	733	732
716	818	656
747	800	665
778	757	691
808	733	720
839	711	698
869	768	650
900	800	615
931	775	610
970	767	609

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Time Since Leachate			Volatil	e Fatty Acids	2	
Production Began,	Acetic	Propionic	Isobutyric	Butyric	Valeric	Total, expressed as
Days	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L Acetic Acid
0	3,931	1,482	232	5,303	1,984	10,073
13	2,842	844	922	2,022	1,190	6,233
28	4,479	1,342	624	,3,822	1,552	9,518
34	4,034	1,883	607	3,465	1,396	9,408
41	15,854	1,803	521	3,376	1,556	20,892
48	7,689	1,549	501	3,288	1,419	12,402
55	5,275	3,147	527	3,168	1,464	11,288
61	6,465	1,946	324	3,150	1,290	11,182
68	5,761	2,912	587	2,239	1,483	11,542
75	4,773	1,552	183	2,083	954	8,248
82	6,180	1,835	238	1,887	1,070	9,849
90	4,780	1,700	235	1,887	1,017	8,203
103	3,218	970	236	1,326	844	5,565
110	2,216	1,523	203	547	467	4,237
117	1,133	2,063	275	347	394	3,461
124	891	1,735	352	300	128	2,818
134	681	738	71	206	156	1,560
142	150	569	61	32	204	794
153	245	1,248	51	67	228	1,472
163	77	841	N.D.*	N.D.	N.D.	759
174	161	112	97	61	38	405
181	139	494	78	36	42	643
205	134	390	N.D.	43	287	650
212	130	398	N.D.	N.D.	N.D.	453

### Table D-7. Individual and Total Volatile Fatty Acids Present in Leachate Samples from Cell A (Single Pass)

# Table D-7. Continued...

Time Since Leachate	Volatile Fatty Acids					
Production Began,	Acetic	Propionic	Isobutyric	Butyric	Valeric	Total, expressed as
Days	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L Acetic Acid
210	110	266	2.11	7	5	hoh
219	110	366	14.	1	5	424
231	63	283	N.D.	N.D.	N.D.	292
254	178	57	33	42	14	295
266	105	50	22	31	16	183
290	150	65	31	30	19	256
350	27	30	10	11	14	75
360	N.D.	33	3	N.D.	N.D.	30
369	31	33	5	N.D.	N.D.	62
377	21	21	16	17	10	64
388	37	38	10	10	10	66
400	N.D.	4	N.D.	1	N.D.	3
426	N.D.	8	N.D.	N.D.	N.D.	6
444	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
458	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
472	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

Time Since Leachate			Volatile	e Fatty Acids	3	
Production Began,	Acetic	Propionic	Isobutyric	Butyric	Valeric	Total, expressed as
Days	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L Acetic Acid
0	3,071	1,144	143	4,029	1,243	7,573
13	3,045	1,233	958	2,312	1,446	7,124
28	3,595	1,097	339	1,983	875	6,582
34	3,829	1,216	649	2,345	1,105	7,506
41	5,603	815	544	2,724	1,858	9,584
48	4,025	1,436	518	2,552	1,144	7,955
55	4,296	3,089	533	2,739	1,218	9,747
61	4,501	1,319	302	2,494	919	8,017
68	6,301	3,630	348	3,903	1,216	12,857
75	3,943	2,414	241	2,292	774	8,082
82	5,163	1,818	199	2,420	698	8,833
90	3,050	1,285	192	1,734	643	5,783
103	4,309	1,193	169	1,957	719	7,149
110	2,313	1,109	153	1,508	497	4,637
117	2,508	1,013	466	2,631	965	6,008
124	2,114	1,731	512	2,998	829	6,398
-134	3,404	2,086	547	2,786	903	7,899
142	1,356	2,046	627	3,861	891	6,599
153	2,854	1,725	424	2,964	1,183	7,258
163	1,859	1,334	295	2,849	832	5,574
174	1,926	1,592	667	2,369	768	5,727
181	1,913	1,856	372	1,752	276	5,029
205	2,470	965	223	1,205	308	4,408
212	2,605	1,774	364	1,926	83	5,654

### Table D-8. Individual and Total Volatile Fatty Acids Present in Leachate Samples from Cell B (Leachate Recycle)

Time Since Leachate			Volatile	e Fatty Acids	3	
Production Began,	Acetic	Propionic	Isobutyric	Butyric	Valeric	Total, expressed as
Days	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L Acetic Acid
				_		
219	4,144	1,324	314	1,839	2,419	8,109
231	4,426	1,381	329	1,805	771	7,457
254	4,066	1,330	320	1,809	906	7,127
266	3,569	1,201	225	1,700	865	6,364
290	3,961	1,301	325	2,000	953	7,161
350	181	306	23	32	106	530
360	347	670	34	19	104	988
369	225	356	19	31	78	594
377	355	482	33	22	90	831
388	350	550	30	19	96	886
400	9	1	N.D.*	N.D.	N.D.	10
426	1	N.D.	N.D.	N.D.	N.D.	1
444	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
462	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.

Table D-8. Continued...

\*N.D. - None Detectable

Time Since	BOD5	COD
Leachate Production Began, Days	Cell A	Cell B
0		0.50
0 13	0.45 0.45	0.50
20	0.50	0.68 0.59
28	0.63	0.66
34	0.03	0.66
48	0.43	0.41
55	0.55	0.50
61	0.34	0.36
68	0.59	0.68
75	0.58	0.63
82	0.64	0.51
90	0.53	0.64
97	0.56	0.63
103	0.71	0.75
117	0.93	0.67
124	0.90	0.63
132	0.61	0.59
139	0.92	0.68
152	0.58	0.77
181	0.77	0.78
196	0.96	0.59
205	0.95	0.63
219	0.81	0.67
241	0.94	0.53
254	0.89	0.68
266	0.87	0.75
283	0.91	0.79
350	0.79	0.91
360	0.65	0.51
369	0.76	0.52
377	0.62	0.49
388	0.67	0.52
400	0.65	0.71
414	0.62	0.61
426	0.50	0.59

Table D-9.	Biochemical Oxygen Demand to Chemical Oxygen Demand
	(BOD <sub>5</sub> /COD) Ratio of Leachate Samples from Cell A
	(Single Pass) and Cell B (Leachate Recycle)

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Time Since	BOD <sub>5</sub> /	(COD
Leachate Production Began, Days	Cell A	Cell B
444 458 472 482 497 540 556 625 686 747 778 808 839 869 900 931	0.54 0.53 0.54 0.50 0.45 0.48 0.44 0.20 0.24 0.27 0.28 0.29 0.28 0.29 0.28 0.30 0.29 0.26	0.55 0.65 0.56 0.56 0.61 0.51 0.45 0.20 0.19 0.25 0.23 0.27 0.21 0.23 0.21
970	0.27	0.22

# Table D-9. Continued...

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'ime Since Leachate	COD/TOC		
Production Began, Days	Cell A	Cell B	
0	2.85	2.63	
13	3.65	2.89	
20 28	3.98	2.91	
20 34	3.45	3.40	
48	3.79	3.40	
55	4.11 4.80	4.86	
55 61	4.80	4.53	
68	3.26	4.49 2.38	
75	3.03	2.30	
82	2.65	3.02	
90	2.20	2.39	
97	2.44	2.09	
103	2.34	2.57	
110	2.98	2.92	
117	1.43	2.42	
124	1.31	2.46	
132	1.09	2.47	
152	1.21	2.02	
181	1.69	2.76	
196	1.47	3.27	
205	1.80	2.66	
212	1.30	3.05	
219	2.04	2.34	
231	2.37	3.14	
241	1.61	2.77	
254	1.95	2.82	
260	1.99	3.08	
266	1.99	2.89	
279	1.86	2.94	
283	1.94	2.87	
350	1.48	3.90	
360	1.14	2.29	
369	1.09	2.22	
377	1.13	2.30	

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Table D-10.	Chemical Oxygen Demand to Total Organic Carbon (COD/TOC)
	Ratio of Leachate Samples from Cell A (Single Pass) and
	Cell B (Leachate Recycle)

Time Since	COD/TOC		
Leachate Production Began, Days	Cell A	Cell B	
388 400 426 444 458 472 482 497 540 556 570 594 625 655 686 716 747 778 808 839	1.13 $1.95$ $1.60$ $1.65$ $1.56$ $1.44$ $1.52$ $1.05$ $1.82$ $1.19$ $1.21$ $1.40$ $1.70$ $1.54$ $1.54$ $1.54$ $1.54$ $1.54$ $1.57$ $1.75$ $1.68$ $1.76$	2.11 1.72 1.75 1.73 1.99 2.12 2.22 1.84 2.38 1.96 2.02 2.30 2.80 2.30 2.50 2.70 2.50 2.45 2.25 2.49	
869 900 931 970	1.65 1.82 1.86 1.76	2.53 2.64 2.39 2.46	

Table D-10. Continued...

Time Since	Conductivity	
Leachate Production Began,	Cell A	Cell B
Days	μmhos	µmhos
13	10,500	9,200
20	7,700	6,200
28	10,400	9,000
34	10,200	8,200
41	6,400	5,400
48	6,800	6,400
55	8,950	8,400
61	10,600	9,400
68	10,900	10,000
75	10,200	10,000
82		8,800
90	9,250	
	10,000	9,800
97	10,700	10,600
103	9,800	9,700
110	9,300	9,600
117	9,200	9,900
124	8,900	10,700
132	9,400	10,300
139	10,600	11,500
152	10,900	12,600
163	9,800	12,500
168	11,200	11,500
181	10,200	12,000
196	9,800	11,500
205	9,900	11,600
219	9,800	11,500
241	9,500	11,200
254	9,900	11,200
260	10,200	11,500
266	9,800	10,700
275	9,600	10,900
283	9,900	10,600
350	11,500	12,000
360	11,200	12,100
369	10,800	11,100
377	10,400	9,600

Table D-11. Conductivity of Leachate from Cell A (Single Pass) and Cell B (Leachate Recycle)

Time Since	etivity	
Leachate Production Began,	Cell A	Cell B
Days	µmhos	µmhos
388	10,100	9,400
400	8,500	7,900
414	8,100	7,300
426	7,200	6,100
429	6,100	5,400
444	6,200	6,200
458	5,400	4,900
472	4,320	4,110
482	4,450	3,210
497	3,650	3,150
516	3,450	2,050
540	2,950	2,910
556	3,255	2,450
570	2,855	2,350
594	2,655	2,710
625	2,455	2,650
655	2,855	2,720
686	2,950	2,790
716	2,855	2,400
747	2,750	2,010
778	2,610	1,850
808	2,420	1,965
839	2,250	2,150
869	2,550	2,215
900	2,605	1,985
931	2,250	1,995
970	2,150	1,855

Table D-11. Continued...

Time Since	p	рН		ORP	
Leachate Production	Cell A	Cell B	Cell A	Cell	
Began,					
Days			E <sub>C</sub> , mV	Е <sub>с</sub> , т	
0	5.20	5.40	300	240	
13	5:00	5.20	210	200	
20	5.10	5.20	-20	-10	
28	5.80	5.60	10	20	
34	5.80	5.60	-30	-10	
41	5.30	5.00	-50	-20	
48	5.10	5.20	-80	-40	
55	5.60	5.50	-150	-90	
61	5.10	5.00	-155	-90	
68	5.01	5.01	-40	-10	
75	5.06	5.07	-270	-298	
82	5.16	5.10	-139	-104	
90	5.42	5.38	-178	-57	
97	5.28	5.19	-178	-140	
103	5.45	5.20	-220	-80	
110	5.80	5.15	-215	-105	
117	6.71	5.24	-238	-88	
124	6.75	5.17	-200	-30	
132	7.11	5.34	-209	-57	
139	7.14	5.40	-218	-75	
152	6.89	5.36	-246	-127	
163	6.86	5.36	-210	-101	
168	6.93	5.43	-209	-78	
174	6.81	5.36	-215	-60	
181	6.87	5.28	~245	-45	
196	6.87	5.41	-245	-67	
205	7.20	5.68	-239	-70	
219	7.09	5.76	-250	-107	
241	6.86	5.45	-210	-95	
254	6.90	5.60	-225	-97	
260	6.80	5.60	-220	-105	
266	6.80	5.60	-225	-110	
275	6.83	5.58	-220	-107	
283	6.88	5.55	-210	-103	

Table D-12. pH and Oxidation-Reduction Potential (ORP) of Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle

Time Since Leachate	p	Н	OF	RP
Production Began,	Cell A	Cell B	Cell A	Cell B
Days -			E <sub>C</sub> , mV	E <sub>C</sub> , mV
350	7.03	6.98	-224	-231
360	7.15	7.03	-205	-235
369	7.10	6.95	-210	-231
377	7.09	6.90	-192	-206
388	7.10	6.98	-205	-232
400	7.10	7.01	-221	-225
414	7.05	7.03	-216	-240
426	7.12	7.10	-225	-236
444	7.10	7.06	-200	-224
458	7.09	7.14	-190	-215
472	7.11	7.13	-205	-225
482	7.06	7.08	-198	-240
497	7.01	7.12	-201	-231
516	7.10	7.20	-183	-215
540	7.16	7.18	-180	-210
556	7.14	7.15	-175	-210
570	7.12	7.21	-143	-230
594	7.11	7.26	-160	-210
625	7.16	7.23	-152	-203
655	7.18	7.21	-159	-199
686	7.12	7.28	-140	-210
716	7.21	7.34	-145	-205
747	7.15	7.21	-130	-188
778	7.10	7.15	-136	-198
808	7.16	7.21	-148	-189
839	7.09	7.21	-135	-195
869	7.17	7.25	-136	-188
900	7.16	7.21	-145	-182
931	7.11	7.23	-142	-197
970	7.10	7.27	-139	-191

Time Since Leachate Production	Organic Carbon P Volatile A	
Began,	Cell A	Cell B
Days	CEIL X	CETT D
Duj 0		
0	0.76	0.67
13	0.36	0.43
28	0.54	0.45
34	0.53	0.57
48	0.58	0.45
55	0.54	0.46
61	0.61	0.42
68	0.56	0.56
75	0.47	0.44
82	0.55	0.43
90	0.32	0.26
103	0.27	0.31
110	0.27	0.22
117	0.23	0.26
124	0.22	0.25
132	0.15	0.32
152	0.19	0.29
181	0.11	0.23
205	0.13	0.28
212	0.07	0.29
219	0.09	0.41
231	0.09	0.44
254	0.09	0.37
266	0.07	0.40
283	0.06	0.47
350	0.02	0.11
360	0.01	0.25
369	0.02	0.13
377	0.02	0.21
388	0.02	0.20
400	-	0.01
426	-	-

Table D-13. Ratio of the Organic Carbon Present in Total Volatile Acid to the Total Organic Carbon (TOC) Content of Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

Time Since		0-11				Cell	D	
Leachate Production Began,	Iron	Cell Manganese	Zinc	Nickel	Iron	Manganese	Zinc	Nickel
Days	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
13	140	45	39	3.5	83	35	26	4.0
34	143	44	20	3:5	20	35	25	3.2
41	208	42	16	3.7	270	39	23	3.5
55	395	51	9	-	395	50	21	3.7
68	300	51	8.4	3.5	466	46	19	2.9
82	150	51	4.5	2.9	480	49	19	2.6
96	110	7	1.0	1.8	450	51	8.5	2.9
110	50	9	1.0	2.4	550	49	10.0	4.0
124	32	8	1.5	2.0	475	56	12.0	3.5
139	50	12	1.5	2.6	575	58	10.0	4.0
152	120	29	1.1	4.0	625	46	8.0	3.5
168	120	10	1.3	3.2	450	74	9.0	6.6
181	70	37	0.6	6.2	285	79	11.0	7.3
205	120	27	0.9	4.0	450	59	7.5	4.6
219	70	16	0.4	3.7	285	57	6.0	3.2
241	59	17	0.5	3.4	260	54	7.0	3.1
260	· 55	15	0.4	3.3	275	50	6.0	3.5
275	60	17	0.5	3.4	255	52	7.0	3.2
350	13	-	-	-	38	17	0.6	2.7
360	18	~	-	2.2	38	19	0.5	2.7
369	16	22	0.15	2.2	47	18	0.6	3.2
377	12	21	0.25	3.3	32	17	0.4	3.3
388	10	16	0.15	2.7	21	18	0.5	2.2
400	8	12	0.2	2.1	25	16	0.3	2.1

Table D-14. Iron, Manganese, Zinc, and Nickel Concentrations in Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

continued...

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Time Since							_	
Leachate	Iron	Cell	A Zinc	Nickel	Inon	Cell	Zinc	Nickel
Production Began,	11.011	Manganese	21110	NICKET	Iron	Manganese	2100	Nickel
Days	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
106	6	10	0.1	2 2	16	2.11	0.1	2.1
426	6	10	0.1	2.3	15	14	0.4	2.1
444	5	8	0.2	1.5	12	8	0.2	0.6
472	6.5	5.4	0.1	0.5	11	6	0.1	0.5
516	7.6	6.3	N.D.*	N.D.	12.6	5.3	0.2	0.4
540	6.5	5.1	N.D.	N.D.	13	6.2	0.1	0.2
570	7.9	6.7	N.D.	N.D.	11.4	4.3	0.1	N.D.
594	10.1	5.2	N.D.	N.D.	10.7	3.6	0.2	N.D.
625	12	4.2	N.D.	N.D.	15.2	2.6	0.2	N.D.
655	10	3.2	N.D.	N.D.	18.6	2.8	N.D.	N.D.
686	8	4.6	N.D.	N.D.	17.6	3.1	N.D.	N.D.
716	7.5	5.1	N.D.	N.D.	12	3.7	N.D.	N.D.
747	7.9	4.8	N.D.	N.D.	8.5	4.1	N.D.	N.D.
778	8.5	4.9	N.D.	N.D.	8.2	4.0	N.D.	N.D.
808	6.7	4.5	N.D.	N.D.	8.9	4.1	N.D.	N.D.
839	7.2	4.1	N.D.	N.D.	9.1	3.2	N.D.	N.D.
869	8.4	3.6	N.D.	N.D.	10.2	2.9	N.D.	N.D.
900	6.1	2.7	N.D.	N.D.	8.1	2.1	N.D.	N.D.
931	7.2	3.6	N.D.	N.D.	7.5	3.1	N.D.	N.D.
970	8.1	2.1	N.D.	N.D.	8.6	2.0	N.D.	N.D.
510	0.1	<i>C</i> .1	N. D.	N.D.	0.0	2.0	N.D.	N. D.

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Table D-14. Continued...

\*N.D. - None Detectable

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Leachate Production Began,	Chloride			
Degan,		(C <sub>t</sub> /C <sub>o</sub> )*	Chloride	(C <sub>t</sub> /C <sub>o</sub> )**
Days	mg/L		mg/L	
13	667	1.00	470	1.00
34	581	.87	445	0.94
41	544	0:81	421	0.89
55	519	0.77	372	0.79
68	507	0.75	372	. 0.79
82	534	0.80	604	1.28
96	719	1.07	654	
110	704	1.05	713	1.39 1.52
124	654	0.98	654	1.39
139	587	0.88	528	1.12
152	555	0.83	565	1.20
168	515	0.77	540	1.15
181	489	0.73	526	1.12
205	469	0.70	549	1.16
219	459	0.69	519	1.10
231	458	0.69	528	1.10
254	425	0.64	501	1.06
275	401	0.60	535	
283	415	0.62	516	1.13
350	360	0.54	491	1.10
369	315	0.47	491 453	1.05
388	320	0.48	486	0.96 1.03
414	301	0.45	501	1.05
429	285	0.43	466	0.99
444	266	0.40	400	0.91
472	200	0.34	420	0.85
497	254	0.38	401 408	
540	221		408	0.87
570	245	0.33		0.89
594	245 201	0.37	429	0.91
625	201 211	0.30	406	0.86
686		0.32	385	0.82
716	206	0.31	366	0.78
747	216 180	0.32	345	0.74
778	196	0.27 0.29	341 328	0.73 0.70

Table D-15. Chloride Concentrations in Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

Time Since Leachate	Cell	l A	Cell	LB
Production Began,	Chloride	(C <sub>t</sub> /C <sub>o</sub> )*	Chloride	(C <sub>t</sub> /C <sub>o</sub> )**
Days	mg/L		mg/L	
808	175	0.26	331	0.70
839	178	0.27	321	0.68
869	179	0.27	315	0.67
900	180	0.27	309	0.66
931	172	0.26	301	0.64
970	165	0.25	300	0.63

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\*C<sub>0</sub> = 667 mg/L \*\*C<sub>0</sub> = 470 mg/L

Sample				Counts P	er Minute in	Different G	PC Fractions			
-	1	2	3	4	• 5	6	7	8	9	10
	(OmL-5mL)	(5mL-10mL)	(10mL-15mL)	(15mL-20mL)	(20mL-25mL)	(25mL-30mL)	(30mL~35mL)	(35mL-40mL)	(40mL~45mL)	(45mL-50mL)
Blank (pH=6.80)	N.D.*	N.D.	N.D.	N.D.	N.D.	N.D.	2,991	2,991	679	113
A~2HA (pH=7.00)	N.D.	23	540	85	444	9,455	10,819	3,492	375	N.D.
A-2HA (pH=5.50)	N.D.	64	190	N.D.	406	5,088	10,198	4,686	1,092	173
A-2HA (pH=8.50)	N.D.	462	125	142	6,408	9,348	2,105	203	N.D.	N.D.
Leachate (pH=7.27)	N.D.	N.D.	N.D.	N.D.	1,632	7,434	4,910	360	N.D.	N.D.

## Table D-16. Radioactivity in the Different GPC Fractions of Equilibrated Samples

\*N.D. - None Detectable.

GPC Fractions	Sample Bla →	nk <sup>*</sup> A-2HA pH=7.0	A-2HA pH=5.5	А-2НА рН=8.5	Leachate <sup>**</sup> pH=7.27
1	-	-	-	-	-
2	-	0.1	0.3	2.5	-
3	-	2.1	0.7	0.7	-
4	-	0.3	-	0.8	-
5		1.8	1.9	34.1	11.3
6	-	37.5	23.2	49.7	59.9
7	36.	5 42.9	46.6	11.2	34.3
8	50.	2 13.8	21.4	1.0	2.5
9	11.	4 1.5	4.9	-	-
10	1.	9 –	-	-	-

Table D-17.	Percent Relative Radioactivity	in Different
	GPC Fractions of Equilibrated	Samples

\*Deionized Water \*\*Leachate from Cell B; Day 970

Time Since Leachate		Cobalt
Production Began,	Cell A	Cell B
Days	mg/L	mg/L
13	0.90	0.78
20	0.42	0.71
34	0.56	0.65
50	0.51	0.55
61	0.52	0.56
82	0.47	0.91
96	0.34	0.99
110	0.27	1.01
124	0.29	0.97
139	0.41	0.82
152	0.54	0.88
168	0.38	0.85
181	0.61	0.82
205	0.42	0.85
241	0.42	0.72
260	0.68	0.68
275	0.52	0.73
350	0.48	0.63
360	0.23	0.55
369	0.15	0.46
377.	0.15 N.D.*	0.48
388	N.D.*	0.45
400	N.D.	0.43
426	N.D.	0.26
24 24 24	N.D.	0.07
472	N.D.	0.12
516	0.05	0.15
540	N.D.	0.13
570	N.D.	0.11
594	N.D.	0.15
625	N.D.	0.15
655	N.D.	0.23
686	N.D.	0.27
716	N.D.	0.29
747	N.D.	0.40

Table D-18. Cobalt Concentrations in Leachate from Cell A (Single Pass) and Cell B (Leachate Recycle)

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## Table D-18. Continued...

Time Since Leachate	Cobalt		
Production Began,	Cell A	Cell B	
Days	mg/L	mg/L	
778	N.D.	0.26	
808	N.D.	0.20	
839	N.D.	0.30	
869	N.D.	0.22	
900	N.D.	0.21	
931	N.D.	0.21	
970	N.D.	0.20	

\*None Detectable

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Time Since Leachate	Correcti	on Factor, [Cl <sup>-</sup> ] <sub>t</sub> /[Cl <sup>-</sup> ] <sub>max</sub>
Production	Cell A	Cell B
Began,		
Days	mg/L	mg/L
13	0.02	0.65
34	0.93 0.81	0.65
41	0.76	0.62
55	0.70	0.58
68	0.72	0.52
82		0.52
96	0.75	0.84
110	1.00	0.91
124	0.98	1.00
139	0.91	0.91
	0.82	0.74
152	0.77	0.79
168	0.68	0.76
181	0.68	0.74
205	0.65	0.76
219	0.64	0.72
231	0.64	0.74
254	0.59	0.69
275	0.56	0.74
283	0.58	0.72
350	0.44	0.69
369	0.44	0.63
388	0.45	0.68
414	0.42	0.68
429	0.40	0.65
444	0.37	0.60
472	0.32	0.57
497	0.35	0.57
540	0.31	0.58
570	0.35	0.60
594	0.28	0.56
625	0.30	0.54
686	0.29	0.51
716	0.30	0.49
747	0.25	0.48
778	0.27	0.46
808	0.24	0.46

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Table D-19. Chloride Based Correction Factor (C.F.) for Dilution

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continued...

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Time Since Leachate	Correction	Factor, [C1 <sup>-</sup> ] <sub>t</sub> /[C1 <sup>-</sup> ] <sub>max</sub>
Production	Cell A	Cell B
Began,		
Days	mg/L	mg/L
0.5.0		
839	0.25	0.44
869	0.25	0.44
900	0.25	0.43
931	0.24	0.42
970	0.23	0.41

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Time Since Leachate	Cell	A	Cell	Cell B	
Production Began, Days	COD "As-Measured" mg/L	Corrected COD mg/L	COD "As-Measured"	Corrected COD	
Days	mg/ L	mg/L	mg/L	mg/L	
13	26,956	28,984	19,468	29,950	
34	26,880	33,185	17,920	28,903	
48	35,276	46,415	34,138	58,858	
55	40,256	55,911	38,327	73,705	
68	26,800	38,285	21,760	73,625	
82	19,000	25,333	25,087	30,158	
96	21,240	21,240	18,540	23,340	
110	18,685	19,066	24,809	19,066	
124	6,630	7,285	24,751	27,198	
139	2,541	3,099	25,856	34,940	
152	3,680	4,779	20,210	25,582	
163	4,530	6,661	18,877	24,838	
181	3,811	5,604	23,762	32,110	
205	3,661	5,632	16,744	22,031	
219	3,716	5,806	18,583	25,809	
231	2,888	4,512	21,066	28,467	
254	2,564	4,345	21,544	31,223	
283	3,324	5,731	17,540	24,361	
350	2,656	5,312	7,614	11,034	
369	1,708	3,881	3,978	6,314	
388	1,640	3,644	3,785	5,566	
414	1,482	3,528	1,689	2,483	
426	1,448	3,620	1,655	2,546	
444	1,812	4,897	2,202	3,670	
472	1,680	5,250	2,634	4,621	
497	1,538	4,394	2,402	4,214	
540	1,290	4,161	2,450	4,224	
570	1,276	3,646	2,340	3,900	
594	1,137	4,060	1,962	3,503	
625	1,261	4,203	1,872	3,466	
686	1,100	3,793	1,831	3,590	
716	1,061	3,536	1,771	3,614	
747	1,256	5,024	1,662	3,462	
778	1,324	4,903	1,692	3,678	

Table D-20. "As-Measured" COD and "Dilution-Effect-Corrected" COD of Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

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Time Since Leachate	Cell	Cell A		Cell B		
Production	COD	Corrected	COD	Corrected		
Began,	"As-Measured"	COD	"As-Measured"	COD		
Days	.mg/L	mg/L	mg/L	mg/L		
808	1,231	5,129	1,620	3,521		
839	1,251	5,004	1,738	3,950		
869	1,267	5,068	1,644	3,736		
900	1,452	5,808	1,623	3,774		
931	1,441	6,004	1,451	3,454		
970	1,342	5,834	1,522	3,712		

Time Since Leachate	Ion	ic Strength (I)
Production	Cell A	Cell B
Began,		
Days	М	M
13	0.168	0.147
20	0:123	0.099
28	0.166	0.144
34	0.163	0.131
41	0.102	0.086
48	0.108	0.102
55	0.143	0.134
61	0.169	0.150
68	0.174	0.160
75	0.163	0.160
82	0.148	0.140
90	0.160	0.156
97	0.171	0.169
103	0.156	0.155
110	0.148	0.153
117	0.147	0.158
124	0.142	0.171
132	0.150	0.164
139	0.169	0.184
152	0.174	0.201
163	0.156	0.200
168	0.179	0.184
181	0.163	0.192
196	0.156	0.184
205	0.158	0.185
219	0.156	0.184
241	0.152	0.179
254	0.158	0.179
260	0.163	0.184
266	0.156	0.171
275	0.153	0.174
283	0.158	0.169
350	0.184	0.192
360	0.179	0.193
369	0.172	0.177

Table D-21. Ionic Strength (I) of Leachate from Cell A (Single Pass) and Cell B (Leachate Recycle)

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continued...

Time Since Leachate	I	onic Strength (I)
Production	Cell A .	Cell B
Began,		
Days	М	Μ
377	0.166	0.153
388	0.161	0.150
400	0.136	0.126
414	0.129	0.116
426	0.115	0.097
429	0.097	0.086
444	0.099	0.099
458	0.086	0.078
472	0.069	0.065
482	0.071	0.051
497	0.058	0.050
516	0.055	0.032
540	0.047	0.046
556	0.052	0.039
570	0.045	0.037
594	0.042	0.043
625	0.039	0.042
655	0.045	0.043
686	0.047	0.044
716	0.045	0.038
747	0.044	0.032
778	0.041	0.029
808	0.038	0.031
839	0.036	0.034
869	0.040	0.035
900	0.041	0.031
931	0.036	0.031
970	0.034	0.029

Time Since Leachate		Ac	tivity Coef:	ficient, (Y	<sub>i</sub> )	
Production		Cell A			Cell B	
Began, Days	Z=1	Z=2	Z=3	Z=1	Z=2	Z=3
13	0.75	0.33	0.08	0.76	0.34	0.09
20	0.77	0.35	0.09	0.78	0.38	0.11
28	0.75	0.33	0.08	0.76	0.34	0.09
34	0.75	0.33	0.08	0.77	0.35	0.09
41	0.78	0.37	0.11	0.79	0.39	0.12
48	0.78	0.37	0.11	0.78	0.37	0.11
55	0.76	0.34	0.09	0.77	0.35	0.09
61	0.75	0.33	0.08	0.76	0.34	0.08
68	0.75	0.32	0.08	0.76	0.33	0.08
75	0.75	0.33	0.08	0.76	0.33	0.08
82	0.76	0.34	0.09	0.76	0.34	0.09
90	0.76	0.33	0.08	0.76	0.33	0.08
97	0.75	0.32	0.08	0.75	0.32	0.08
103	0.76	0.34	0.09	0.76	0.34	0.09
110	0.76	0.34	0.09	0.76	0.33	0.08
117	0.76	0.34	0.09	0.76	0.34	0.09
124	0.76	0.34	0.09	0.75	0.32	0.08
132	0.76	0.34	0.08	0.75	0.33	0.08
139	0.75	0.33	0.08	0.75	0.32	0.07
152	0.75	0.32	0.08	0.75	0.31	0.07
163	0.76	0.34	0.09	0.75	0.31	0.07
168	0.75	0.32	0.08	0.75	0.32	0.07
181	0.75	0.33	0.08	0.75	0.32	0.07
196	0.76	0.34	0.09	0.75	0.32	0.07
205	0.76	0.34	0.09	0.75	0.32	0.07
219	0.76	0.34	0.09	0.75	0.32	0.07
241	0.76	0.33	0.08	0.75	0.32	0.08
254	0.76	0.34	0.09	0.75	0.32	0.08
260	0.75	0.33	0.08	0.75	0.32	0.07
266	0.76	0.34	0.09	0.75	0.32	0.08
275	0.76	0.33	0.08	0.75	0.32	0.08
283	0.76	0.34	0.09	0.75	0.35	0.08
350	0.75	0.32	0.07	0.75	0.32	0.07
360	0.75	0.32	0.08	0.75	0.32	0.07

Table D-22. Activity Coefficient (Y<sub>i</sub>) for Monovalent, Divalent, and Trivalent Ions in Leachate Samples from Cell A (Single Pass) and Cell B (Recycle)

Table D-22. Continued...

Time Since

Activity	Coefficient,	$(\gamma_i)$
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Leachate					1,	
Production Began, Days		Cell A			Cell B	
	Z=1	Z=2	Z=3	Z=1	Z=2	Z=3
369	0.75	0.32	0.08	0.75	0.32	0.08
377	0.75	0.33	0.08	0.76	0.33	0.08
388	0.76	0.33	0.08	0.76	0.34	0.08
400	0.76	0.35	0.09	0.77	0.36	0.10
414	0.77	0.35	0.10	0.78	0.36	0.10
426	0.78	0.36	0.10	0.78	0.38	0.11
429	0.78	0.38	0.11	0.79	0.39	0.12
444	0.78	0.38	0.11	0.78	0.38	0.11
458	0.79	0.39	0.12	0.80	0.41	0.13
472	0.81	0.42	0.14	0.81	0.43	0.15
482	0.80	0.42	0.14	0.82	0.46	0.17
497	0.82	0.44	0.16	0.82	0.46	0.18
516	0.82	0.45	0.17	0.85	0.52	0.23
540	0.83	0.47	0.18	0.83	0.47	0.18
556	0.82	0.46	0.17	0.84	0.49	0.21
570	0.83	0.48	0.19	0.84	0.50	0.21
594	0.83	0.48	0.20	0.83	0.48	0.20
625	0.84	0.49	0.21	0.83	0.48	0.20
655	0.83	0.48	0.19	0.83	0.48	0.20
686	0.83	0.47	0.18	0.83	0.48	0.19
716	0.83	0.48	0.19	0.84	0.49	0.21
747	0.83	0.48	0.19	0.85	0.52	0.23
778	0.83	0.49	0.20	0.85	0.53	0.24
808	0.84	0.50	0.21	0.85	0.52	0.23
839	0.84	0.51	0.21	0.84	0.51	0.22
869	0.84	0.49	0.20	0.84	0.51	0.22
900	0.84	0.49	0.20	0.85	0.52	0.23
931	0.84	0.51	0.21	0.85	0.52	0.23
970	0.84	0.51	0.22	0.85	0.53	0.24

Time Since Leachate	Tota	al Sulfide (S <sub>T</sub> )
Production Began,	Cell A	Cell B
Days	mg/L	mg/L
. 27	N.D.*	N.D.
43	0.23	N.D.
64	0.31	N.D.
110	0.15	
134	0.23	N.D.
152		N.D.
	0.30	N.D.
168	0.21	N.D.
183	0.34	N.D.
198	0.36	N.D.
205	0.23	N.D.
219	0.32	N.D.
241	0.38	N.D.
260	0.29	N.D.
283	0.85	N.D.
350	0.81	0.56
369	0.15	0.45
377	0.45	0.65
388	0.65	0.95
400	0.61	1.50
426	0.43	3.25
444	0.23	4.50
472	0.15	2.25
516	0.28	1.95
540	0.31	1.65
570	0.26	1.25
594	0.21	1.15
625	0.17	0.85
655	0.19	0.95
686	0.22	0.65
716	0.19	0.55
747	0.11	0.25
778	0.10	0.15
808	N.D.	0.11
839	N.D.	0.13

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Table D-23. Total Sulfide Concentrations in Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

## Table D-23. Continued...

Time Since Leachate	Total Sulfide $(S_T)$		
Production Began,	Cell A	Cell B	
Days	mg/L	mg/L	
869	N.D.	N.D.	
900 931	N.D. N.D.	N.D. N.D.	
970	N.D.	N.D.	

Time Since Leachate	Total Alkalinity*		
Production	Cell A	Cell B	
Began,			
Days	mg/L as CaCO <sub>3</sub>	mg/L as CaCO <sub>3</sub>	
13	4,717	3.547	
20	5,559	4,380	
28	8,500	10,000	
34	16,900	12,700	
55	8,466	7,042	
61	9,169	7,127	
68	9,504	9,750	
75	6,598	6,384	
82	7,485	6,889	
97	4,229	5,910	
103	4,500	7,800	
110	4,350	4,850	
117	4,200	6,500	
124	4,100	6,350	
132	4,500	6,800	
139	4,300	7,150	
152	4,750	7,500	
163	4,440	6,500	
168	4,800	6,100	
181	5,000	6,750	
196	4,600	6,250	
205	4,800	6,600	
219	4,700	6,670	
241	4,300	6,120	
260	4,100	5,255	
266	4,200	5,735	
283	4,100	5,825	
350	3,450	3,650	
360	4,050	4,595	
377	3,583	3,666	
400	3,683	3,253	
414	3,215	3,050	
426	2,950	3,151	
429	2,655	3,156	
444	2,450	2,985	

Table D-24. Total Alkalinity of Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

Table D-24. Continued...

Time Since Leachate	Total Alkalinity*			
Production Began,	Cell A	Cell B		
Days	mg/L as CaCO <sub>3</sub>	mg/L as CaCO <sub>3</sub>		
h= 0	0.150			
458	2,150	2,465		
472	2,250	2,215		
482	2,200	2,015		
497	2,106	2,050		
516	2,195	1,985		
540	2,000	1,865		
556	2,050	1,915		
570	2,065	1,975		
594	1,965	1,915		
625	1,985	1,995		
655	2,125	1,925		
686	2,225	1,985		
716	2,067	1,915		
747	2,095	1,929		
778	2,165	1,967		
808	2,087	1,927		
839	2,015	1,815		
869	2,070	1,867		
900	2,055	1,895		
931	2,035	1,950		
970	1,990	1,825		

\*Titrated to pH=3.5

Time Since	Stro	ntium
Leachate Production Began,	Cell A	Cell B
Days	mg/L	mg/L
13	4.5	5.0
34	6.5	9.8
50	5.8	12.3
61	5.4	11.8
82	5.2	16.2
96	5.9	13.9
110	5.9	14.9
114	6.0	15.6
139	5.4	17.3
152	5.1	18.2
168	5.7	18.6
181	5.1	18.1
205	4.2	17.9
241	4.3	18.6
260	4.2	17.3
275	4.3	17.2
350	3.8	7.9
377	3.7	3.9
400	3.5	3.7
472	3.1	2.9
540	3.2	2.5
570	2.8	2.1
594	2.5	2.5
625	2.3	2.6
655	2.1	2.1
686	2.3	1.9
716	2.2	1.8
747	2.3	1.9
778	2.1	2.0
808	2.1	1.8
839	2.1	1.7
869	2.0	1.7
900	1.9	1.7
931	1.8	1.6
970	1.7	1.5

Table D-25. Strontium Concentrations in Leachate from Cell A (Single Pass) and Cell B (Leachate Recycle)

Time Since Leachate	Total Inorganic Carbon	
roduction Began,	Cell A	Cell B
Days	mg/L	mg/L
117*	200	N.D.
124	100	N.D.
132	300	N.D.
152	250	N.D.
181	300	N.D.
196	450	N.D.
205	400	N.D.
212	526	N.D.
219	415	N.D.
231	470	N.D.
241	575	N.D.
254	625	N.D.
260	650	100
266	620	126
273	585	250
283	500	115
350	615	450
360	525	500
369	490	575
377	515	515
388	525	565
400	495	522
414	450	587
426	425	606
429	415	585
444	410	479
458	395	450
472	350	415
482	385	395
497	295	401
516	265	358
540	301	373
556	315	395
570	310	315
594	327	287

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Table D-26. Total Inorganic Carbon (TIC) Present in Leachate Samples from Cell A (Single Pass) and Cell B (Leachate Recycle)

continued.....

Table D-26. Continued...

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Time Since	Total Inorganic Carbon	
Leachate Production Began,	Cell A	Cell B
Days	mg/L	mg/L
625	310	315
655	327	287
686	287	215
716	265	295
747	292	263
778	311	257
808	301	287
839	265	299
869	295	216
900	327	192
931	343	187
970	301	199

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\*TIC was below detectable limits prior to Day 117.

## REFERENCES

- 1. Carter, M. W., A. A. Moghissi, and B. Kahn, <u>Management of Low-</u> Level Radioactive Waste, Pergamon Press, (1979).
- 2. Gillette, R., "Radiation Spill at Hanford: The Anatomy of an Accident", Science, 181, August, 1973.
- 3. Lipschutz, R. D., <u>Radioactive Waste</u>, Ballinger Publishing Co., (1980.
- 4. National Academy of Science, "The Shallow-Land Burial of Low-Level Radioactivity Contaminated Solid Waste", Washington, D.C., (1976).
- 5. Toste, A. P., L. J. Kirby, and T. R. Pahl, "Organic Geochemical Studies at a Commercial Shallow-Land Disposal Site of Low-Level Nuclear Waste", in <u>Waste Management '84</u>, Vol. 2, Post, R. G., (Ed.), p:557-563 (1984).
- 6. Zehner, H. H., "Preliminary Hydrogeologic Investigation of the Maxey Flats Radioactive Waste Burial Site, Fleming County, Kentucky", in <u>Research Program at Maxey Flats and Consideration of</u> <u>Other Shallow Land Burial Sites</u>, NUREG/CR-1832, PNL-3510, XI-1, (1981).
- 7. Toste, A. P., L. J. Kirby, and T. R. Pahl, "Role of Organics in the Subsurface Migration of Radionuclides in Groundwater", in <u>Geochemical Behavior of Radioactive Waste</u>, Barney, G. S., Navratil, J. D., and W. W. Schulz (Eds.), American Chemical Society Symposium Series; <u>246</u>, p.251-270 (1984).
- Kirby, L. J., A. P. Toste, and C. L. Wilkerson, "Chemical Species of Migrating Radionuclides at a Shallow-Land Low-Level Radioactive Waste Burial Site", in <u>Environmental Migration of Long-Lived</u> <u>Radionuclides</u>, International Atomic Energy Agency, Vienna, Austria p.63 (1982).
- Means, J. L., and C. A. Alexander, "The Environmental Biochemistry of Chelating Agents and Recommendations for the Disposal of Chelated Radioactive Wastes", in <u>Nuclear and Chemical Waste</u> Management, Vol. 2, p:183-196 (1981).

- 10. Rees, T. F., and J. M. Cleveland, "Characterization of Plutonium in Waters at Maxey Flats, Kentucky, and Near the Idaho Chemical Processing Plant, Idaho", in <u>Environmental Migration of Long-Lived</u> <u>Radionuclides</u>, International Atomic Energy Agency, Vienna, <u>Austria</u>, p.41-52 (1982).
- 11. Means, J. L., D. A. Crerar, and J. O. Duguid, "Migration of Radioactive Wastes: Radionuclide Mobilization by Complexation Agents", <u>Science</u>, Vol. 200, p.1477-1481 (1978).
- Toste, A. P., L. J. Kirby, D. E. Robertson, K. H. Abel, and R. W. Perkins, "Characterization of Radionuclide Behavior in Low-Level Waste Sites", <u>IEEE Transactions on Nuclear Science</u>, NS-30 (1), p.580-585 (1983).
- 13. Kirby, L. J. (Ed.), "Radionuclide Distributions and Migration Mechanisms at Shallow-Land Burial Sites: Annual Report of Research Investigations on the Distribution, Migration, and Containment of Radionuclides at Maxey Flats, Kentucky", NUREG/CR-2383, National Technical Information Service, Springfield, Virginia (1982).
- 14. Toste, A. P., L. J. Kirby, W. H. Rickard, and D. E. Robertson, "Radionuclide Characterization, Migration, and Monitoring at a Commercial Low-Level Disposal Site", in <u>Radioactive Waste Manage-</u> <u>ment</u>, Proceedings of an International Conference on Radioactive Waste Management, International Atomic Energy Agency, Vienna, Austria, IAEA-CN-43/470, <u>5</u>, p.213-226 (1984).
- 15. Jordan, E. A., and C. F. Nern, "Low-Level Waste Management: Towards a Stable System", <u>Transactions of American Nuclear</u> <u>Society</u>, Vol. 43, p.96 (1983).
- 16. Lohaus, P. H., and T. C. Johnson, "The NRC Approach to Dealing with Hazardous Substances in Low-Level Radioactive Waste", Transactions of American Nuclear Society, Vol. 44, p.63 (1983).
- 17. Kemper, J. S., E. J. Kohler, R. J. Scholz, "One Utility's Low-Level Waste Management Approach to Being in a Region Without a Burial Ground", <u>Transactions of American Nuclear Society</u>, Vol. 47, p.76 (1984).
- Higginbotham, L. B., and K. C. Jackson, "Licensing of New Low-Level Waste Disposal sites and Relevant State Activities", <u>Trans-</u> actions of American Nuclear Society, Vol. 47, p.63 (1984)
- Day, J. E., "Low-Level Waste Management Strategies Available to Utilities in a Region Without a Burial Gound", <u>Transactions of</u> American Nuclear Society, Vol. 47, p.75 (1984).

- 20. Buchnea, A., "A System Modeling Approach to Evaluating Low-Level Waste Disposal Systems", <u>Transactions of American Nuclear Society</u>, Vol. 44, p.88 (1983).
- Chang, K. C., "Co-disposal of Low-Level Radioactive Waste Within Sanitary Landfills", Doctoral Dissertation, Georgia Institute of Technology, Atlanta, GA, September, 1982.
- 22. Pohland, F. G., "Leachate Recycle as Landfill Management Option", Jour. of Env. Eng. Div., American Society of Civil Enginers, <u>EE6</u>, p.105-1069 (1980).
- Pohland, F. G., "Controlled Landfill Stabilization by Leachate Recycle", <u>Report to U.S. EPA</u>, Grant No. R-803953, p.149, September, 1982.
- 24. Toste, A. P., T. R. Pahl, and R. B. Myers, "Analysis of Organic Chelating Agents in Nuclear Waste and Their Aqueous Leachates", in Proceedings of the 1984 International Chemical Congress of Pacific Basin Societies, Dec. 12-16 (1984).
- 25. Dertein, J. T., "Critical Evaluation of Treatment Processes for Leachates from Landfill Disposal Sites", Special Research Problem Report, Georgia Institute of Technology, Atlanta, GA, p.228 (1983).
- Atomic Industrial Forum, Inc., "A Survey and Evaluation of Handling and Disposing of Solid Low-Level Nuclear Fuel Cycle Wastes", Washington, D.C., (1976).
- 27. U.S. EPA, "Low-Level Radioactive Waste Management", 520/3-79-002, (1979).
- Leventhal, L. and B. C. Christensen, "Laboratory Procedures for Assessment of Chemical/Physical Toxicity Parameters in Low-Level Wastes", <u>Transactions of American Nuclear Society</u>, Vol. 44, p.67 (1983).
- 29. "State-of-the-Art Review of Radioactive Waste Volume Reduction Techniques for Commercial Nuclear Power Plants", Oak Ridge National Laboratory, ORNL/SUB-79/13837/2, April, 1980.
- Anderson, R. L. <u>et al.</u>, "Institutional Radioactive Waste", NUREG/CR-0028, University of Maryland at Baltimore, March, 1978.
- 31. IRG, "Report to the President by the Interagency Review Group on Nuclear Waste Management", Washington, D.C., TIC-28817, October, 1978.

- 32. Kibbey, A. H., H. W. Godbee, and E. L. Compere, "A Review of Solid Radioactive Waste Practices in Light-Water-Cooled Nuclear Reactor Power Plants", NUREG/CR-0144, October; 1978.
- 33. "State-of-the-Art Review of Alternatives to Shallow Land Burial of Low-Level Radioactive Waste", Oak Ridge National Laboratory, ORNL/SUB-79/13837/1, April, 1980.
- 34. Macbeth, P. J., G. Wehmann, B. J. Thames, and D. H. Card, "Evaluation of Alternative Methods for the Disposal of Low-Level Radioactive Waste", NUREG/CR-0680, July, 1979.
- 35. Colombo, P. J., A. J. Weiss, and A. J. Francis, "Evaluation of Isotope Migration-Land Burial, Water Chemistry at Commercially Operated Low-Level Radioactive Waste Disposal Sites, Quarterly Progress Report No. 2, BNL-NUREG-50666, p.20, May, 1977.
- 36. Colombo, P., A. J. Weiss, and A. J. Francis, "Evaluation of Isotope Migration-Land Burial, Water Chemistry at Commercially Operated Low-Level Radioactive Waste Disposal sites, BNL-NUREG-50670, p.21, June, 1977.
- 37. Colombo, P., A. J. Weiss, and A. J. Francis, "Evaluation of Isotope Migration-Land Burial, Water Chemistry at Commercially Operated Low-Level Radioactive Waste Disposal Sites, BNL-NUREG-50695, p.20, September, 1977.
- 38. Colombo, P., A. J. Weiss, and A. J. Francis, "Evaluation of Isotope Migration-Land Burial, Water Chemistry at Commercially Operated Low-Level Radioactive Waste Disposal Sites, BNL-NUREG-50739, p.39, December, 1977.
- 39. Fenimore, J. W., "Borehole Monitoring of Radioactive Waste Trenches", in <u>Management of Low-Level Radioactive Waste</u>, Carter, M. W., A. A. Moghissi, and B. Kahn (Eds.), Pergamon Press, NY, p.1173-1192 (1979).
- 40. Garrett, P. M., "An Evaluation of Low-Level Radioactive Waste Burial Ground Capacities at the Major Department of Energy Reservations", ORNL/NFW-79/14, p.49, January, 1979.
- 41. Giardina, P. A., M. F. DeBonis, J. Eng, and G. L. Meyer, "Summary Report on the Low-Level Radioactive Waste Burial Site, West Valley, NY (1963-1975)", USGS Open File Report, February, 1977.
- 42. Godse, V. B., A. L. Mohan, M. Singh, R. V. Amalraj, and K. T. Thomas, "Characterization of Trombay Soils for Disposal of Radioactive Wastes", <u>Proceedings of a Symposium on Disposal of</u> <u>Radioactive Waste into the Ground, International Atomic Energy</u> <u>Agency</u>, Vienna, Austria, p.301-314, June, 1967.

- 43. Holcomb, W. F., "A Summary of Shallow Land Burial of Radioactive Wastes at Commercial Sites Between 1962 and 1976, with Projections", Nuclear Safety, Vol. 19, No. 1, p.50-59 (1978).
- 44. Kelleher, W. J., "Water Problems at the West-Valley Burial Site", in <u>Management of Low-Level Radioactive Waste</u>, Carter, M. W., A. A. Moghissi, and B. Kahn (Eds.), Pergamon Press, NY, p.843-851 (1979).
- Lomenick, T. R., "Burial Ground Technology", ORNL-5508, p.65-78, September, 1978.
- 46. Lomenick, T. F., and K. E. Cowser, "Land Burial of Solid Waste at Oak Ridge National Laboratory", in <u>Ground Disposal of Radioactive</u> <u>Wastes</u>, Morgan, J. M. Jr., <u>et al.</u> (Eds.), p.635, March, 1962.
- 47. Markham, O. D., "Radioactive Contamination Near the Radioactive Waste Management Complex, Idaho National Engineering Laboratory Site", IDO-12079, p.68, July, 1975.
- 48. Meyer, G. L., "Recent Experience with the Land Burial of Solid Low-Level Radioactive Wastes", <u>Proceedings of an IAEA Symposium on</u> <u>Management of Radioactive Wastes from the Nuclear Fuel Cycle</u>, Vol. 2, p.383-395 (1976).
- 49. Myers, D. A., "Environmental Monitoring Report on the Status of Ground Water Beneath the Hanford Site-Jan-Dec 1977", <u>PNL-2624</u>, p.94, April, 1978.
- 50. Myers, D. A., J. J. Fix, P. J. Blumer, J. R. Raymond, V. L. McGhan, and E. L. Hilty, "Environmental Monitoring Beneath the Hanford Site-Jan-Dec 1975", BNWL-2034, p.27 (1976).
- 51. Stevens, P. R., and G. D. Debuchanane, "Problems in Shallow Land Disposal of Solid Low-Level Radioactive Waste in the United States", <u>Bulletin of the International Association of Engienering</u> Geology, Vol. 14, p.161-171 (1976).
- 52. Pohland, F. G., J. P. Gould, and S. B. Ghosh, "Management of Hazardous Waste by Landfill Codisposal with Municipal Refuse", Hazardous Waste Journal, Vol. 1, No. 2, 1985.
- 53. Tchobanoglous, G., H. Theisen, and R. Eliassen, <u>Solid Wastes:</u> Engineering Principles and Management Issues, McGraw-Hill, NY, 1979.
- 54. Myers, T. E., J. C. Duke Jr., P. G. Malone, and D. W. Thompson, "Gas Production in Sanitary Landfill Simulators", <u>Fifth Annual</u> <u>Research Symposium, Solid and Hazardous Waste Research Division</u>, U.S. EPA, EPA-600/9-79-023a, March, 1979.

- 55. Chian, E.S.K., E. Hammerburg, and F. B. DeWalle, "Effect of Moisture Regimes and Other Factors on Municipal Solid Waste Stabilization", <u>Proceedings of the Third Annual Municipal Solid</u> <u>Waste Research Symposium</u>, EPA-600/9-77-0260a, September, 1977.
- 56. U.S. EPA, "National Survey of Community Solid Waste Practices", Cincinnati, OH (1968).
- 57. Walsh, J. J., and R. N. Kinman, "Leachate and Gas Production Under Controlled Moisture Conditions", EPA-600/8-79-023a, August, 1979.
- 58. "Analysis of Composition of Rubbish in the United States", <u>Solid</u> Waste Management/RRJ, September, 1982.
- 59. Pohland, F. G., and J. P. Gould, "Fate of Heavy Metals During Stabilization of Solid Waste Materials with Leachate Recycle", Final Project Report, EPA Project R-806498, p.219 (1984).
- 60. Pohland, F. G., J. T. Dertier, and S. B. Ghosh, "Leachate and Gas Quality Changes During Landfill Stabilization", <u>Proc. Third Intl.</u> Symp. on Anaerobic Digestion, Cambridge, MA, p.185 (1983).
- Regan, T. M., and M. M. Peters, "Heavy Metals in Digesters: Failure and Cure", Jour. Water Pollution Control Federation, Vol. 39, (1967).
- 62. Lawrence, A. W., and P. L. McCarty, "The Effects of Sulfides on Anaerobic Treatment", <u>Proc. Nineteenth Industrial Waste</u> Conference, Purdue University, Lafayette, IN, (1964).
- 63. Lawrence, A. W., and P. L. McCarty, "The Role of Sulfide in Preventing Heavy Metal Toxicity in Anaerobic Treatment", <u>Jour.</u> Water Pollution Control Federation, Vol. 37, (1965).
- Pohland, F. G., J. P. Gould, R. E. Ramsey, B. J. Spiller, and W. R. Esteves, "Containment of Heavy Metals in Landfills with Leachate Recycle", U.S. EPA-600/9-81-002a, p.181, March, 1981.
- 65. Stumm, W., and J. J. Morgan, <u>Aquatic Chemistry</u>, J. Wiley and Sons, NY.
- 66. Westall, J. C., J. L. Zachary, and F.M.M. Morel, <u>MINEQL: A</u> <u>Computer Program for the Calculation of Chemical Equilibrium</u> <u>Composition of Aqueous Systems</u>, Massachusetts Institute of Technology, EPA Grant No. R-803738, July, 1976.
- 67. Oakley, S. M., C. E. Delphey, K. J. Williamson, and P. O. Nelson, "Kinetics of Trace Metal Partitioning in Model Anoxic Marine Sediments", <u>Water Research</u>, Vol. 14, p.1067-1072 (1980).

- 68. Cabaniss, A. E., M. S. Shuman, and B. J. Collins, "Metal-Organic Binding: A Comparison of Models", Presented at the International Symposium on Complexation of Trace Metals in Natural Waters, Texel, the Netherlands, May 2-6, 1983 (In Press).
- McKnight, D. M., G. L. Feder, E. M. Thurman, R. L. Wershaw, and J. C. Westall", Complexation of Copper by Aquatic Humic Substances from Different Environments", <u>The Science of the Total</u> <u>Environment</u>, Vol. 28, p.65-76 (1983).
- 70. Beveridge, A., and W. F. Pickering, "Influence of Humate-Solute Interactions on Aqueous Heavy Metal Ion Levels", <u>Water, Air, and</u> <u>Soil Pollution</u>, Vol. 14, p.171-185 (1980).
- 71. Buffle, J., F. Greter, and W. Haerdi, "Measurement of Complexation Properties of Humic and Fulvic Acids in Natural Waters with Lead and Copper Ion-Selective Electrodes", <u>Anal. Chem</u>., Vol. 49, p.216-222 (1977).
- 72. Benes, P., E. T. Gjessing, and E. Steinnes, "Interactions Between Humus and Trace Elements in Fresh Water", <u>Water Research</u>, Vol. 10, p.711-716 (1976).
- 73. Perdue, E. M., "Solution Thermochemistry of Humic Substances: Acid-Base Equilibria of River Water Humic Substances", in <u>Chemical</u> <u>Modeling in Aqueous Systems</u>, Jenne, E. A., (Ed.), ACS Symposium Series, No. 93, p.99-113 (1979).
- 74. Rashid, M. A., and J. D. Leonard, "Modifications in the Solubility and Precipitation Behavior of Various Metals as a Result of Their Interactions with Sedimentary Humic Acid", <u>Chemical Geology</u>, Vol. 11, p.89-97 (1973).
- 75. Ganzerli Valentini, M. T., L. Maggi, R. Stella, and G. Ciceri, "Metal-Humic and Fulvic Acid Interactions in Fresh Water Ultrafiltrate Fractions", <u>Chemistry in Ecology</u>, Vol. 1, p.279-291 (1983).
- 76. Whitworth, C. G., Jr., "Metal Ion Complexation and Precipitation by Coal Humic Acid", <u>Diss. Abst. Int. Rt. B-Sci. and Eng</u>., Vol. 43, No. 4, p.207 (1982).
- 77. Knox, K., and P. H. Jones, "Complexation Characteristics of Sanitary Landfill Leachates", <u>Water Research</u>, Vol. 13, No. 9, p.839-846 (1979).
- 78. Chian, E.S.K., and F. B. DeWalle, "Evaluation of Leachate Treatment Volume I: Characterization of Leachate", U.S. EPA, EPA-600/2-77-186a (1977).

- 79. Raveh, A., and Y. Avimelech, "Leaching of Pollutants from Sanitary Landfill Models", Jour. Water Pollution Control Federation, Vol. 51, No. 11, p.2705-2716 (1979).
- Artiola-Fortuny, J., and W. H. Fuller, "Humic Substances in Landfill Leachates: I. Humic Acid Extraction and Identification", Jour. Environmental Quality, Vol. 11, p.663-669 (1982).
- 81. Swanson, J. L., "Organic Complexant-Enhanced Mobility of Toxic Elements in Low-Level Wastes", Presented at the Sixth Annual Participant's Information Meeting DOE Low-Level Waste Management Program (LLWMP), Denver, CO, <u>PNL-SA-12518</u>, September, 1984.
- 82. <u>Standard Methods for the Examination of Water and Wastewater</u>, 13th Ed., APHA, AWWA, WPCF, (1975).
- 83. Cross, W. H., S. R. Harper, S. B. Ghosh, and H. Graven, "Rapid Gas Chromatographic Analysis of Volatile Acids in Anaerobic Treatment Systems", <u>Presented at the 185th National Meeting of ACS</u> (Div. of Env. Chem.), March, 1983.
- 84. U.S. EPA, "Interim Method for the Analysis of Elemental Priority Pollutants in Sludge", (1978).
- Reuter, J. H., "Chemical and Spectroscopic Characterization of Humic Substances Derived from River Swamps in the Flood Plains of Southeastern U.S. Coastal Streams", <u>NTIS</u>, Springfield,, VA, PB81-150245, (1980).
- Perry, R. H., and C. H. Chilton, <u>Chemical Engineering Handbook</u>, 5th Ed. McGraw-Hill Book Company, p.19-84.
- 87. Chian, E.S.K., S. S. Cheng, F. B. DeWalle, and P.P.K. Kuo, "Removal of Organics in Sewage and Secondary Effluent by Reverse Osmosis", <u>Prog. Water Technology</u>, Vol. 9, Pergamon Press, p.761-776 (1977).
- Dehaan, H., "Molecular-Size Distribution of Soluble Humic Compounds from Different Natural Waters", <u>Freshwat. Biol.</u>, Vol. 2, p.235-241 (1974).
- Dawson, H. J., B. F. Hrutfiord, R. J. Zasocki, and F. C. Ugolini, "The Molecular Weight and Origin of Yellow Organic Acids", <u>Soil</u> <u>Sci.</u>, Vol. 132, p.191-199 (1981).
- 90. Eaker, D., and J. Parath, "Sorption Effects in Gel Filtration: I. A Survey of Amino Acid Behavior of Sephadex G-10", <u>Separation</u> <u>Science</u>, Vol. 2, p.507-550 (1967).

- 91. Gjessing, E. T., "Gel and Ultramembrane Filtration of Aquatic Humus: A Comparison of the Two Methods", <u>Hydrologie</u>, Vol. 35, No. 2, p.286-294 (1973).
- 92. Gjessing, E. T., and G. F. Lee, "Fractionation of Organic Matter in Natural Waters on Sephadex Columns", <u>Environ. Sci. Technol.</u>, Vol. 1, p.631-638 (1967).
- 93. Gloor, R., H. Leidner, K. Wuhrmann, and Th. Fleishmann, "Exclusion Chromatography and Carbon Detection: A Tool for Further Characterization of Dissolved Organic Carbon", <u>Water Research</u>, Vol. 15, p.451-462 (1981).
- 94. Goh, K. M., and M. R. Reid, J. Soil Sci., Vol. 26, p.207 (1975).
- 95. Hine, P. T., and D. B. Bursill, "Gel Permeation Chromatography of Humic Acid: Problems Associated with Sephadex Gel", <u>Water</u> Research, Vol. 18, No. 11, p.1461-1465 (1984).
- 96. Liao, W., R. F. Christman, J. D. Johnson, D. S. Millington, and J. R. Hass, "Structural Characterization of Aquatic Humus Material", Environ. Sci. Technol., Vol. 16, p.403-410 (1982).
- 97. Posner, A. M., "Importance of Electrolyte in the Determination of Molecular Weights by 'Sephadex' Gel Filtration, With Especial Reference to Humic Acid", Nature, Vol. 198, p.1161 (1963).
- 98. Snow, M. G., "Study of Brown Organic Acids by Gel Filtration", Water Treatment Exam., Vol. 24, p.297-317 (1975).
- 99. Swift, R. S. and A. M. Posner, "Gel Chromatography of Humic Acid", J. Soil Sci., Vol. 22, p.237 (1971).
- 100. Urano, K., K. Katagiri, and K. Kamato, "Characteristics of Gel Chromatography Using Sephadex Gel for Fractionation of Soluble Organic Pollutants", Water Research, Vol. 14, p.741-745 (1980).
- 101. Wershaw, R. L., and D. J. Pinckney, "The Fractionation of Humic Acids from Natural Water Systems", <u>J. Res. U.S. Geol. Sur</u>., Vol. 1, p.361-366 (1973).
- 102. Larson, R. A., "Dissolved Organic Matter of a Low-Coloured Stream", <u>Freshwater Biology</u>, Vol. 8, p.91-104 (1978).
- 103. Hall, K. J., and G. F. Lee, "Molecular Size and Spectral Characterization of Organic Matter in a Meromictic Lake", <u>Water</u> <u>Research</u>, Vol. 8, p.239-251 (1974).
- 104. Kemp, A.L.W., and H.K.T. Wong, "Molecular-Weight Distribution of Humic Substances From Lake Ontario and Erie Sediments", <u>Chemical</u> Geology, Vol. 14, p.15-22 (1974).

- 105. Tate, K. R., and H. A. Anderson, "Phenolic Hydrolysis Products from Gel Chromatographic Fractions of Soil Humic Acids", <u>J. Soil</u> <u>Sci.</u>, Vol. 29, p.76-83 (1978).
- 106. Plechanov, N., "Studies of Molecular Weight Distribution of Fulvic and Humic Acids by Gel Permeation Chromatography. Examination of the Solute Molecular Composition Using RI, UV, Fluorescence and Weight Measurement as Detection Techniques", Organic Geochem., Vol. 5, No. 3, p.143-149 (1983).
- 107. Thurman, E. M., R. L. Wershaw, R. L. Malcolm, and D. J. Pinckney, "Molecular Size of Aquatic Humic Substances", <u>Organic Geochem.</u>, Vol. 4, p.27-35 (1982).
- 108. Reuter, J. H., and E. M. Perdue, "Calculation of Molecular Weights of Humic Substances from Colligative Data: Application of Aquatic Humus and its Molecular Size Fractions", <u>Geochimica et</u> Cosmochimica Acta, Vol. 45, p.2017-2022 (1981).
- 109. Pohland, F. G., "Sanitary Landfill Stabilization with Leachate Recycle and Residual Treatment", EPA-600/2-75-043, October, 1975.
- 110. Lind, C. J., "Specific Conductance as a Means of Estimating Ionic Strength", U.S. Geologic Survey Professional Paper 700D, p.272-280 (1970).
- 111. Russell, L. L., "Chemical Aspects of Groundwater Recharge with Wastewaters", Ph.D. Thesis, University of California at Berkeley, (1976).
- 112. Davies, C. W., <u>Electrochemistry</u>, George Newnes Limited, London, U.K., p.52-62 (1967).
- 113. "Methods of Chemical Analysis of Water and Wastes", U.S. EPA, 1979.