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Project Summary

Behavior and Assimilation of Organic and Inorganic Priority Pollutants Codisposed with Municipal Refuse

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Research was undertaken to demonstrate and evaluate the capacity of landfill systems to assimilate and attenuate inorganic and organic priority pollutants loadings codisposed with municipal refuse and to determine the fate and effect of the codisposed pollutants as landfill stabilization progressed under conditions of single-pass leaching and leachate recycle.

The results from the study of 10 simulated landfill columns demonstrated that the columns employing leachate recycle achieved waste stabilization more rapidly and completely and exhibited greater assimilation and attenuation of the codisposed priority pollutants than did the single-pass columns.

This Project Summary was developed by EPA's Risk Reduction Engineering Laboratory, Cincinnati, OH, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Effective management of increasing amounts of solid waste has become a priority societal challenge. Of all available solid waste management options, disposal in landfills is most frequently employed, primarily because of associated economic advantages and tradition. Moreover, regardless of the emphasis on other solid waste management alternatives, the land will continue to serve as a final waste receptor, whether for combustion ash, discards from recycling initiatives, or regulated hazardous waste.

Landfills are currently designed and operated to minimize potential nuisances and adverse health and environmental impacts by controlling disposal methods and by managing leachate and gas generation. One of two fundamental leachate management strategies can be employed; one strives to limit rainfall infiltration and provides single-pass leaching with leachate collection, removal, and separate treatment before ultimate discharge; the second involves controlled rainfall infiltration, leachate collection, and *in situ* recirculation or recycle before ultimate discharge. The former strategy is characteristic of the more conventional or traditional approaches, whereas the latter leachate recirculation technique is a more recent innovation that essentially converts the landfill into a controlled anaerobic bioreactor with accelerated waste conversion and stabilization in a more predictable and cost-effective manner. In either case, the gases generated from waste stabilization consist primarily of methane and carbon dioxide, but greater opportunities for controlled energy recovery and use of the methane are afforded when the temporal and spatial dimensions of landfill development are planned to regulate the progress of waste stabilization. Therefore, accelerated stabilization can result from *in situ* leachate recirculation in controlled landfills, with enhanced opportunities for recovery and use gas as a useful energy source.

Because most landfills essentially exist as anaerobic biological waste stabilization processes during most of their active lives, the same fundamentals that apply to separate anaerobic treatment processes also

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apply to landfills, although effective retention times and opportunities for use and conversion of less available substrates in these separate treatment systems are different from those provided by the landfills of today. Therefore, the purpose of this research was to employ this analogy to demonstrate the comparative capacities of both single-pass leaching and leachate recycle for waste stabilization and concomitant assimilation and attenuation of both organic and inorganic priority pollutants when codisposed with municipal refuse in simulated landfills.

Construction, Loading, and Operation of the Simulated Landfills

The construction and operational features of the five pairs of simulated landfills with single-pass leaching and with leachate recycle are illustrated in Figure 1. All five pairs received equal quantities of shredded municipal refuse, with one pair serving as controls and the other column pairs receiving organic and inorganic priority pollutants at the test loadings indicated in Table 1. The corresponding combined loadings to each of the simulated landfills are indicated in Table 2. After loading, moisture was added incrementally to the simulated landfills to initiate leaching and waste stabilization; an average of 350 L for the recycle columns and 1430 L for the single-pass column over the 1428-day operational period. The moisture added to the former recycle columns was restricted to the amount necessary to maintain leachate recirculation, whereas that added to the latter single-pass columns was equivalent to local rainfall infiltration rates averaged over the experimental period.

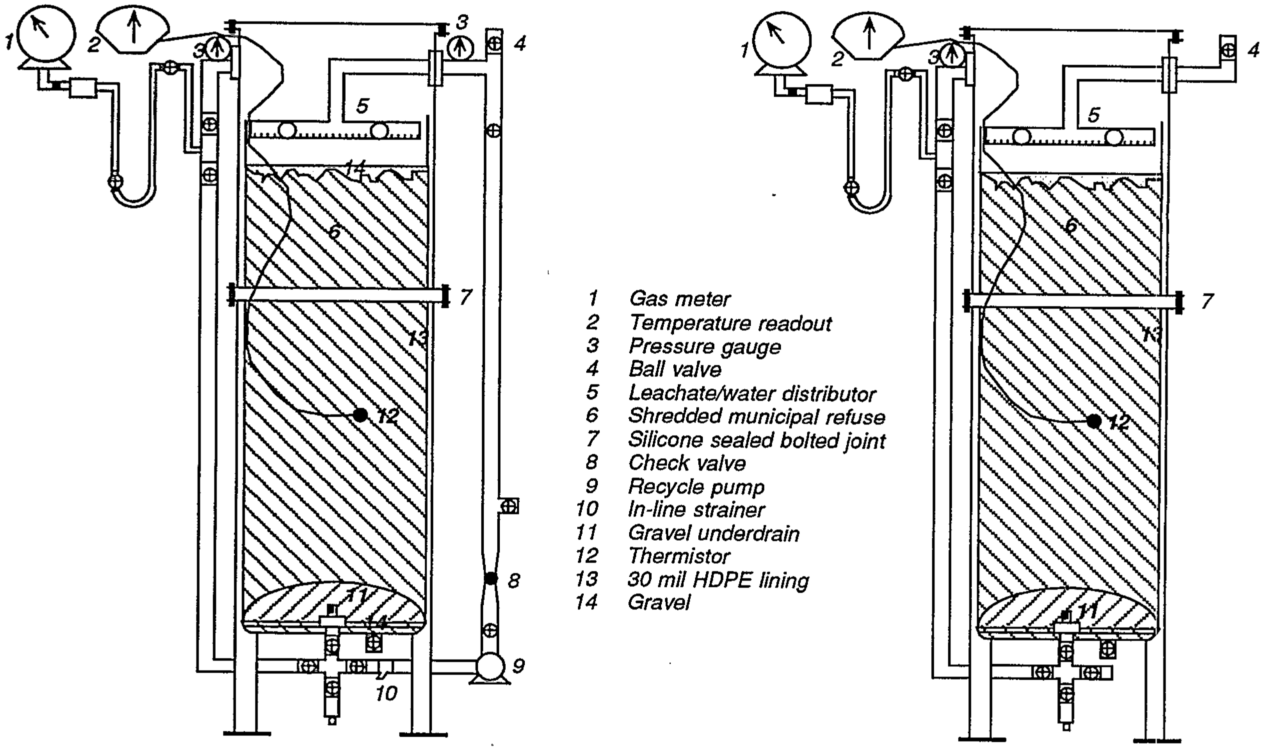
The initial moisture additions were made intentionally to establish and prolong the acid formation phase of landfill stabilization until the effects of aggressive leachate generation could be ascertained. Thereafter, incremental anaerobic digester sludge (a total of 111 L) and pH neutralization (Na_2CO_3) were added over a 232-day period to induce methane fermentation. On completion of the methane fermentation phase, the simulated landfill operations were ended and the columns were disassembled for inspection and retrieval of waste matrix samples for analysis.

Presentation and Discussion of Results

Leachate samples from each of the 10 simulated landfill columns were routinely collected and analyzed for pH, total and individual volatile acids, alkalinity, COD, TOC, ORP, chloride, ammonia, nitrogen, sulfate, sulfide, Na, K,

Ca, Mg, Fe, Cd, Cr, Pb, Mn, Ni, Hg, and the organic priority pollutants or their conversion products. Similarly, gas samples from each column were analyzed for CO_2, O_2, N_2, H_2, and CH_4, and for the volatile organic priority pollutants or their conversion products. Ambient temperature throughout the experimental period (10.3°C to 31.1°C) were also recorded.

Selected results for cumulative gas production (Figure 2) and its composition (Table 3), leachate pH (Figure 3), and total volatile acids (Figure 4) indicate the dramatic differences between performance



Recycled simulated landfill column

Single-pass simulated landfill column

Figure 1. Construction and operational features of simulated landfills.

Table 1. *Simulated Landfill Column Loadings and Operation*

Column number	Column identity*	Operation	Organics	Inorganics
1	CR	Recycle	No	No
2	CS	Single-Pass	No	No
3	OS	Single-Pass	Yes	No
4	OLS	Single-Pass	Yes	Low
5	OMS	Single-Pass	Yes	Medium
6	OR	Recycle	Yes	No
7	OLR	Recycle	Yes	Low
8	OHS	Single-Pass	Yes	High
9	OMR	Recycle	Yes	Medium
10	OHR	Recycle	Yes	High

* CR Control, Recycle
CS Control, Single-Pass
OS Organics, Single-Pass
OLS Organics, Low Inorganics, Single-Pass
OMS Organics, Medium Inorganics, Single-Pass
OR Organics, Recycle
OLR Organics, Low Inorganics, Recycle
OHS Organics, High Inorganics, Single-Pass
OMR Organics, Medium Inorganics, Recycle
OHR Organics, High Inorganics, Recycle

with leachate recycle and single-pass leaching, the discrete separation of periods of active acid formation and methane fermentation (~ day 800), and the comparative effects of the organic and inorganic priority pollutant loadings. Elevated total volatile acids (TVA) concentrations and low pH and gas production were indicative of the acid formation phase of landfill stabilization, whereas reduced TVA concentrations and elevated gas production and pH were indicative of the methane fermentation phase. Moreover, more gas resulted from leachate recycle operations, where convertible substrate was retained within the landfill columns, than resulted from the single-pass leaching operations, where substrate was washed out and wasted with an equivalent loss in potential gas (and energy) yield. Similarly, the recycle columns were less affected by the priority pollutant loadings, with retardation of stabilization more related to heavy metal loadings than to organic priority pollutant additions.

The differences in the effects of priority pollutants were determined to be a function of sufficiency and intensity of potential attenuating mechanisms. For example, the reducing conditions prevailing during methane fermentation provided a favorable chemical environment for microbially mediated reduction of sulfates to sulfides and the resultant removal of many of the heavy metals as sparingly soluble sulfides, as exemplified by leachate cadmium reductions (Figure 5); or for reductive dehalogenation as exemplified by leachate dibromomethane (Figure 6); or trichloroethylene (Figure 7) reductions with accumulations of conversion products (Br and vinyl chloride), respectively. The magnitude of conversion was greater with leachate recycle than with single-pass leaching, largely because of the enhanced opportunities for microbial acclimation with the extended contact times (~ 350 days) of the former as contrasted with the greater inhibition and washout effects of the latter.

In the final analysis, in addition to the more efficient and accelerated waste stabilization provided by leachate recycle in contrast to single-pass leaching, the *in situ* conversion and transformation of the organic and inorganic priority pollutants was more rapid and complete. Although varying quantities of the organic priority

Table 2. *Shredded Municipal Refuse, Organic and Inorganic Priority Pollutants, and Sawdust Loadings (in g) for Each Simulated Landfill Column*

	Column number and type*									
Constituent	1CR	2CS	3OS	4OLS	5OMS	6OR	7OLR	8OHS	9OMR	10OHR
Trichloroethene	—	—	120	120	120	120	120	120	120	120
Dibromomethane	—	—	120	120	120	120	120	120	120	120
2-Nitrophenol	—	—	120	120	120	120	120	120	120	120
1,4-Dichlorobenzene	—	—	120	120	120	120	120	120	120	120
Nitrobenzene	—	—	120	120	120	120	120	120	120	120
Naphthalene	—	—	120	120	120	120	120	120	120	120
1,2,4-Trichlorobenzene	—	—	120	120	120	120	120	120	120	120
2,4-Dichlorophenol	—	—	120	120	120	120	120	120	120	120
Hexachlorobenzene	—	—	120	120	120	120	120	120	120	120
Lindane	—	—	120	120	120	120	120	120	120	120
Bis-2-ethylhexylphthalate	—	—	30	120	120	120	120	120	120	120
Dieldrin	—	—	30	30	30	30	30	30	30	30
Cadmium	—	—	—	35	70	—	35	140	70	140
Chromium	—	—	—	45	90	—	45	180	90	180
Mercury	—	—	—	20	40	—	20	80	40	80
Nickel	—	—	—	75	150	—	75	300	150	300
Lead	—	—	—	105	210	—	105	420	210	420
Zinc	—	—	—	135	270	—	135	540	270	540
Sawdust	6,000	6,000	6,000	6,000	6,000	6,000	6,000	6,000	6,000	6,000
Shredded Municipal Refuse+, kg	381	381	381	381	381	381	381	381	381	381

* C = Control, R = Recycle, O = Organic pollutants, L = Low metals, M = Medium metals, H = High metals, S = Single-pass.
\+ As placed refuse, in kg.

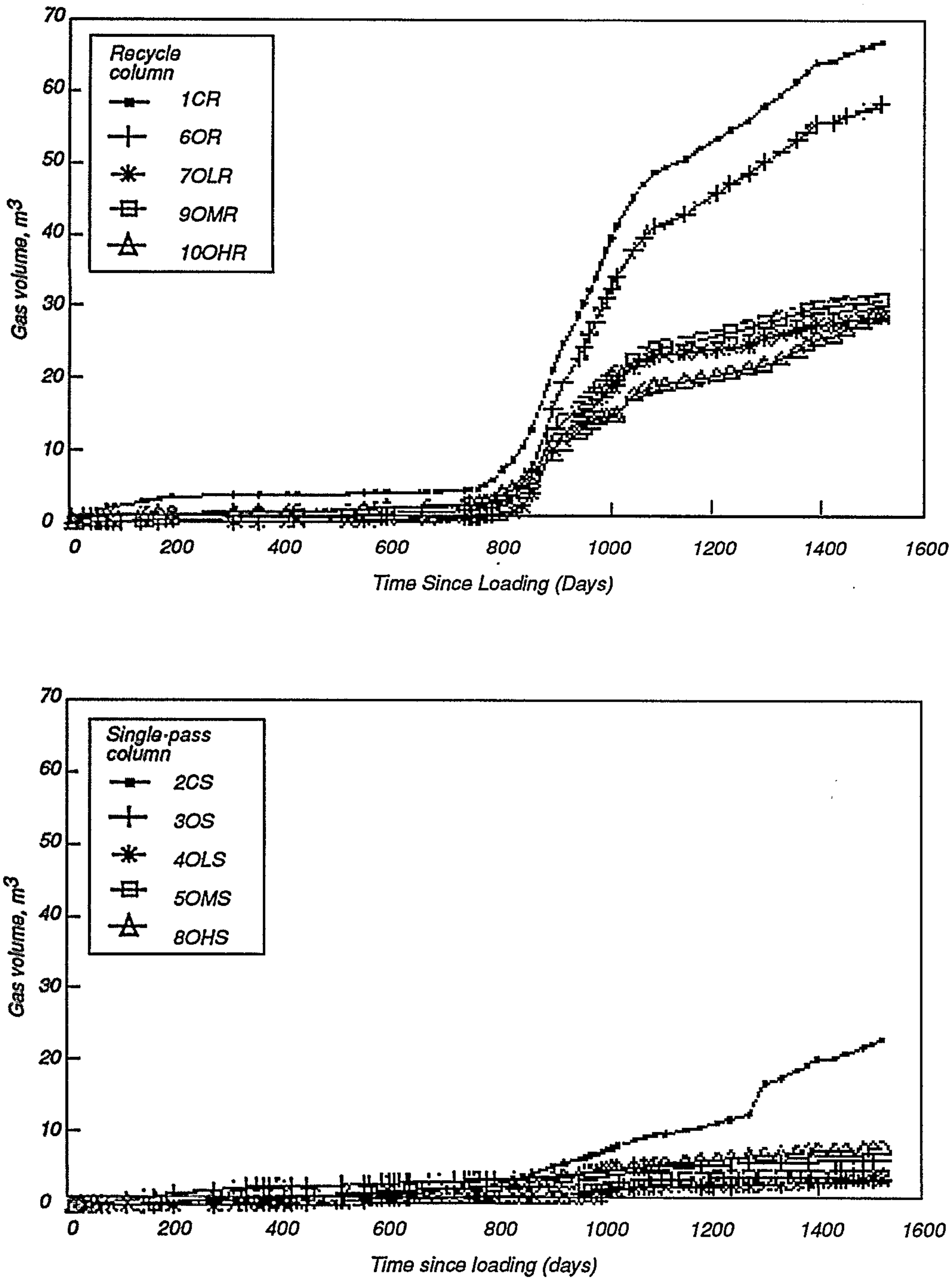


Figure 2. Cumulative gas production during simulated landfill investigations.

pollutants were leached, retained, or transformed (Tables 4 and 5), most of the inorganic heavy metals were either wasted with the discarded leachate (single-pass columns) during acid formation, or removed mainly by precipitation and matrix capture during methane fermentation. Indeed, little of the original heavy metal loadings was detected in the leachates from any columns at the end of the experimental period, with the recycle columns serving as effective reservoirs for capture and storage of the heavy metals.

Summary and Conclusions

Ten simulated landfill columns were operated in pairs with either single-pass leaching or leachate recycle through organic and inorganic priority pollutants that had been codisposed with shredded municipal refuse. The results demonstrated that the fate and effect of the codisposed priority pollutants and the progress of landfill stabilization were affected by the leachate management and loading technique employed. The columns employing leachate recycle achieved waste stabilization more rapidly and completely, as evidenced by trends in gas and leachate characteristics, and also exhibited greater assimilation and attenuation of the codisposed priority pollutants than did the single-pass columns. Furthermore, although the overall gas production and quality was reduced in the columns receiving loadings of inorganic and/or organic priority pollutants, these loading effects were more severe for the single-pass than for the leachate recycle columns.

Conservative leachate constituents, such as chloride and sodium, could be used to reflect the effects of single-pass or leachate recycle operations. Although these constituents were retained within the leachate of the recycle columns at relatively constant concentrations, they were removed from the single- pass columns primarily by washout. This washout from the single-pass columns served to reduce leachate concentration profiles and lessened opportunities for complete waste stabilization and/or effective assimilation/attenuation of priority pollutant loadings. Operations with leachate recycle did not inhibit stabilization of the readily degradable waste fractions, although some retardation was evident at higher priority pollutant loadings; results with single-pass leaching did, however, inhibit both waste stabilization and attenuation processes, mainly because of washout of essential nutrients and elimination of potential *in situ* attenuating mechanisms. These microbially mediated mechanisms were expressed for the leachate recycle columns principally by abiotic and biotic transformation and sorption of the organic priority pollutants within the waste matrix, or by precipitation, sorption, ion-exchange, filtration, and matrix capture of the inorganic priority pollutants. Therefore, results of these investigations have firmly established the efficacy of controlled landfill systems with leachate containment, collection, and recycle for accelerated *in situ* stabilization of both nonhazardous and hazardous solid waste constituents.

Table 3. Comparison of Gas Composition Composition During Simulated Landfill Investigations

Column identity	Project day when N_2 becomes small (<5%)	Project day when CH_4 appears (>1%)	Average gas percentage during the methane fermentation phase (project days 910-1428)		
			CO_2	N_2	CH_4
Recycle:					
1CR	826	700	44.02	2.77	52.81
6OR	868	742	45.17	0.67	54.14
7OLR	854	728	46.07	0.71	53.20
9OMR	868	728	45.42	0.61	53.96
10OHR	868	714	43.54	0.95	55.44
Single-pass:					
2CS	896	238	45.37	0.79	53.74
3OS	-	700	42.96	21.95	34.36
4OLS	-	714	39.47	31.23	28.73
5OMS	-	714	38.21	37.74	23.77
8OHS	-	700	45.81	18.88	34.26

Based on the extensive database developed during the course of the investigations, it could be concluded that:

1. Controlled leachate containment, collection, and recirculation offers opportunities for more rapid and complete stabilization of landfilled municipal solid wastes, including attenuation of codisposed organic and inorganic priority pollutants, than does the single-pass leaching more commonly associated with traditional landfill practices.
2. Loadings of codisposed priority pollutants in the form of heavy metals and/or selected classes of toxic organic substances can retard the sequential phases of landfill stabilization. Loading effects will, however, more severely affect leachate and gas characteristics during single-pass leaching than during leachate recycle operations.
3. Leachate and gas characteristics, described by various physical and chemical indicator parameters, can be used to reflect the progress of waste conversion in terms of longevity and intensity of the acid formation and methane fermentation phases of landfill stabilization.
4. A threshold inhibition level for waste conversion, equivalent to the highest inorganic priority pollutant loading, was established with leachate recycle operations, whereas with single-pass leaching, inhibition was exhibited at the lowest priority pollutant loading. When extrapolated to practice, however, these effects would be a function of site-specific conditions, including the waste loading and operational techniques employed.
5. Landfills possess a finite capacity to attenuate hazardous and nonhazard-ous organic and inorganic waste constituents through a wide array of biological and physicochemical mechanisms. These mechanisms principally include reduction, pre-cipitation, and matrix capture for heavy metals, and biotic or abiotic transformation with matrix interaction through sorption for organic priority pollutants.
6. Controlled landfill systems, designed and operated as anaerobic bioreactors with leachate containment, collection, and recycle, enhance predictability and opportunities for effective management, thereby minimizing potential adverse health and environmental effects, while encouraging innovation and associated regulatory and public acceptance.

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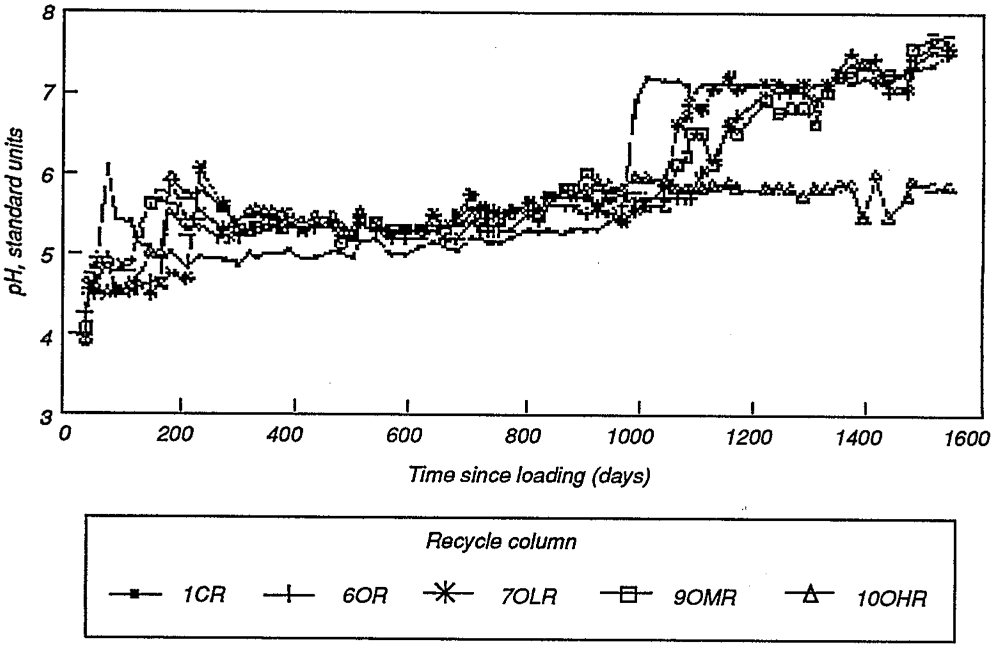


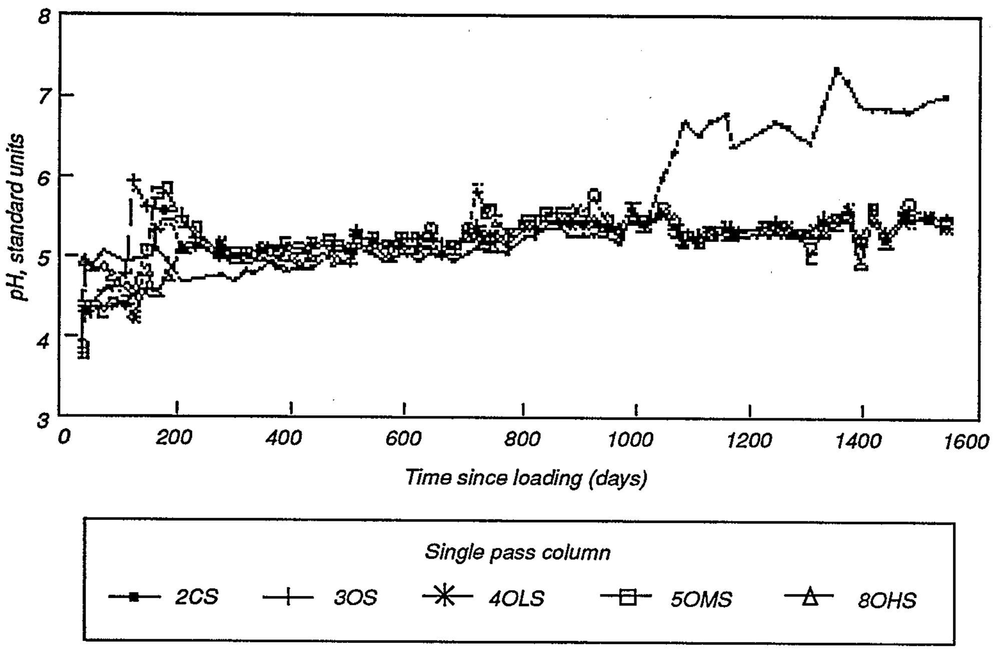
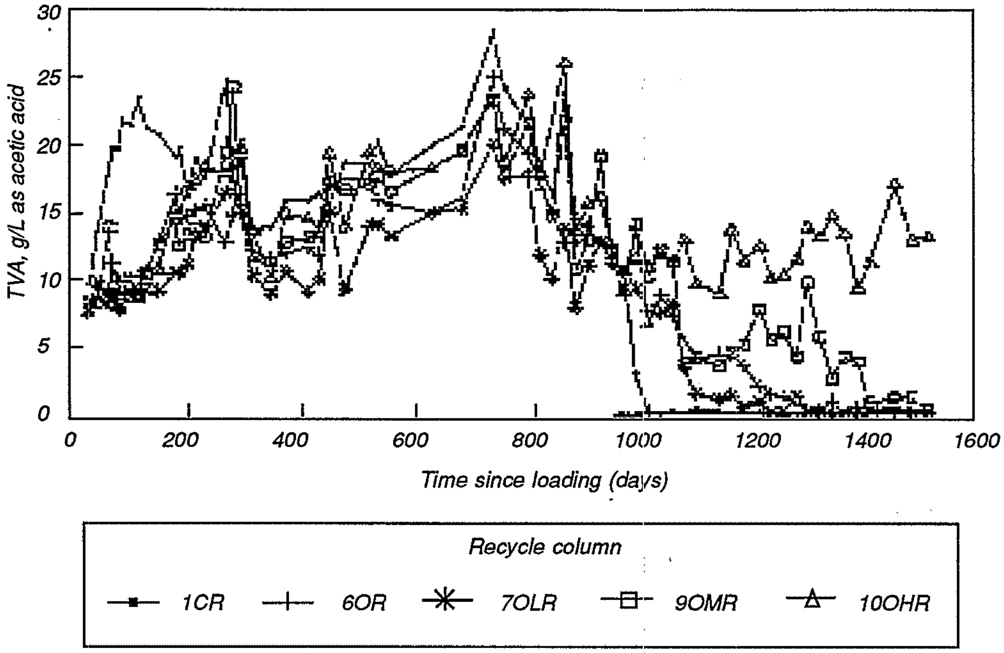


Figure 3. ***Leachate pH during simulated landfill investigations.***



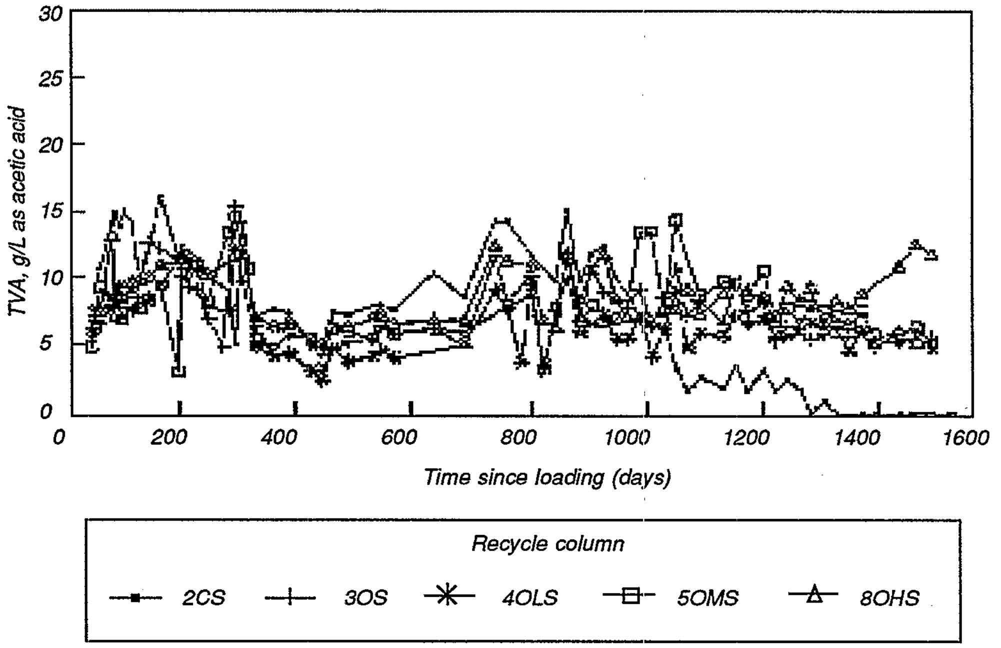


Figure 4. *Leachate total volatile acids during simulated landfill investigations.*

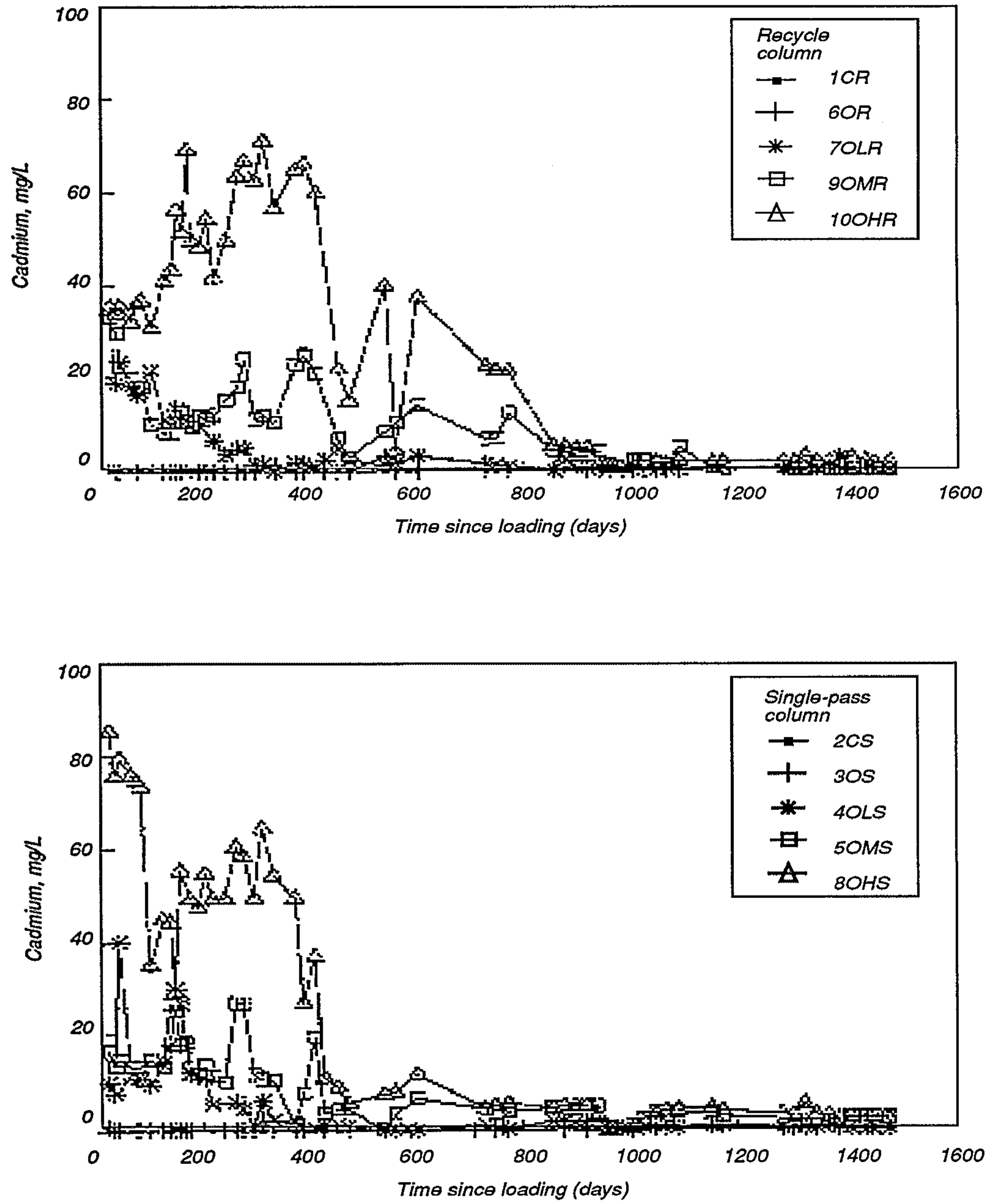


Figure 5. Leachate cadmium during simulated landfill investigations.

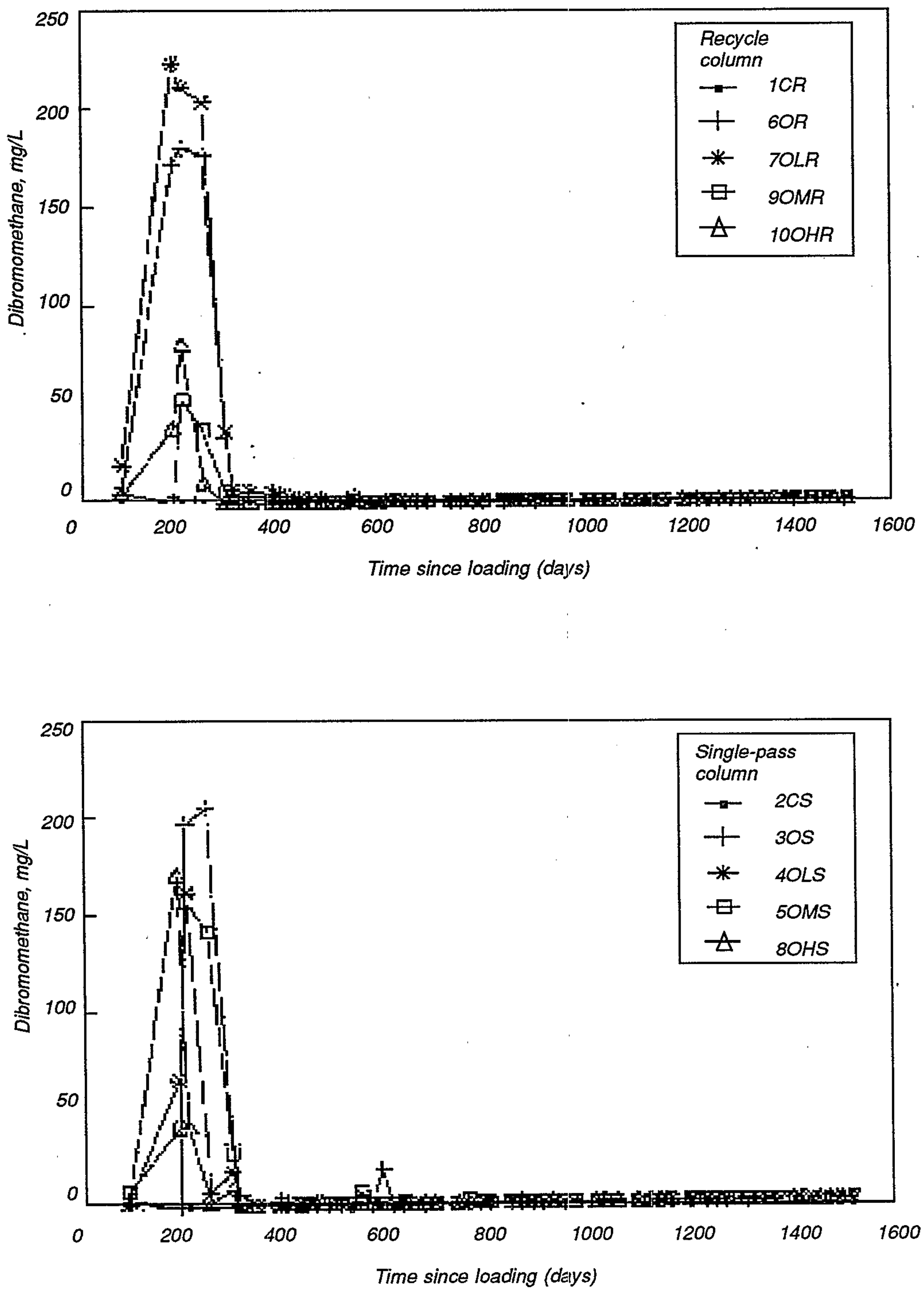


Figure 6. *Leachate dibromomethane during simulated landfill investigations.*

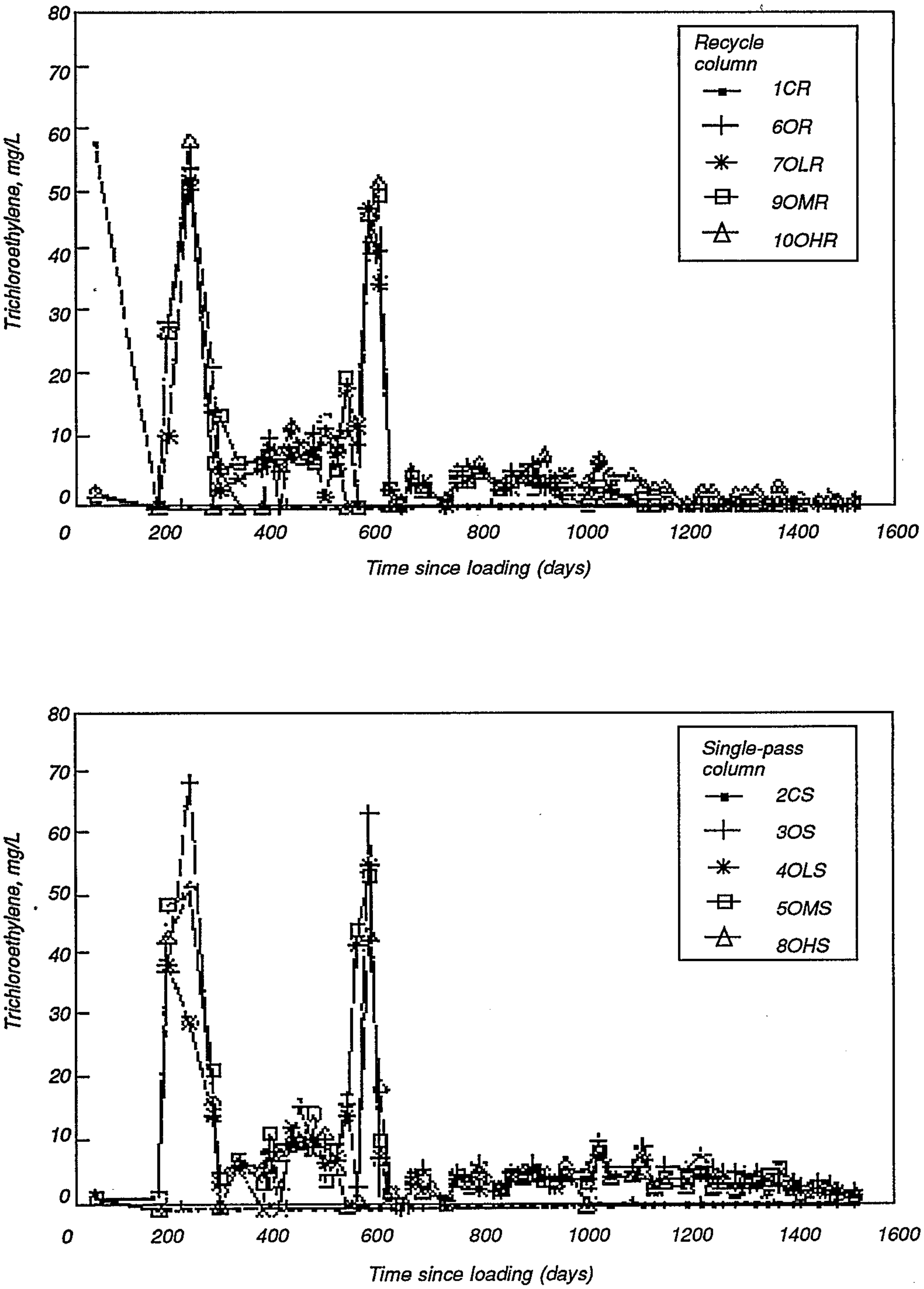


Figure 7. Leachate trichloroethylene during simulated landfill investigations.

Table 4. *Mass Balance Summary on Organic Priority Pollutants for the Single-Pass Simulated Landfill Columns, Percent*

Compound	Leached	Retained	Transformed†
Dibromomethane (DBM)	14.1 (6.1-27.4)*	0	85.9 (72.6-93.9)
Trichloroethene (TCE)	10.7 (7.77-14.58)	0	89.3 (85.42-92.23)
Nitrobenzene (NB)	0.75 (0.02-2.31)	0	99.25 (97.69-99.98)
2-Nitrophenol (NP)	0.31 (0.03-1.16)	0	99.69 (98.84-99.97)
2,4-Dichlorophenol (DCP)	10.9 (8.66-11.81)	15.4 (0.74-25.2)	73.6 (57.71-87.75)
1,4-Dichlorobenzene (DCB)	3.8 (2.53-5.98)	48.4 (30.96-68.63)	47.8 (28.55-81.27)
Naphthalene (NAP)	1.2 (1.04-1.34)	46.8 (17.48-59.53)	52.0 (39.13-81.27)
Lindane (LIN)	0	52.2 (0-100)	47.8 (0-100)
1,2,4-Trichlorobenzene (TCB)	0.17 (0.08-0.32)	39.7 (6.67-60.0)	60.1 (42.00-93.01)
Dieldrin (DIEL)	0	0	100
Hexachlorobenzene (HCB)	0	57.1 (0-96.67)	42.9 (3.33-100)
Bis(2-ethylhexyl)phthalate (BEHP)	0	0	100

*Ranges in parentheses.
†Mass not accounted for in the leachate or recovered from the waste.

Table 5. *Mass Balance Summary on Organic Priority Pollutants for the Recycle Simulated Landfill Columns, Percent*

Compound	Leached	Retained	Transformed†
Dibromomethane (DBM)	1.71 (0.12-2.66)*	0	98.29 (97.34-99.88)
Trichloroethene (TCE)	0.57 (0.40-0.83)	0	99.43 (99.04-99.60)
Nitrobenzene (NB)	0.07 (0.02-0.10)	0	99.93 (99.90-99.98)
2-Nitrophenol (NP)	0.03 (0.01-0.04)	0	99.97 (99.96-99.99)
2,4-Dichlorophenol (DCP)	2.55 (0.41-8.73)	25.17 (6.50-41.99)	72.29 (41.99-94.39)
1,4-Dichlorobenzene (DCB)	1.20 (0.21-3.90)	35.37 (18.99-48.89)	63.44 (50.79-80.80)
Naphthalene (NAP)	0.41 (0.09-1.32)	48.28 (21.75-63.31)	53.00 (21.2-78.16)
Lindane (LIN)	0	66.29 (33.75-93.17)	33.71 (6.83-66.25)
1,2,4-Trichlorobenzene(TCB)	0.05 (0.0-0.17)	38.15 (32.58-43.75)	61.81 (37.42-67.40)
Dieldrin (DIEL)	0	0	100
Hexachlorobenzene (HCB)	0	86.31 (46.42-100)	13.69 (0.53.58)
Bis(2-ethylhexyl)phthalate (BEHP)	0	0	100

*Ranges in parentheses.
†Mass not accounted for in the leachate or recovered from the waste.

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Robert E. Landreth *is the EPA Project Officer (see below).*
The complete report, entitled "Behavior and Assimilation of Organic and Inorganic Priority Pollutants Codisposed with Municipal Refuse," (Order No. PB93-222198AS; Cost: $36.50, subject to change) will be available only from:
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