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Research and Development

Project Summary

Biodegradative Analysis of Municipal Solid Waste in Laboratory-Scale Landfills

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The objective of this research was to characterize the anaerobic biodegradability of the major biodegradable components of municipal solid waste (MSW). Tests were conducted in quadruplicate in 2-L reactors operated to obtain maximum yields. Measured methane (CH₄) yields for grass, leaves, branches, food waste, coated paper, old newsprint, old corrugated containers, and office paper were 144.4, 30.6, 62.6, 300.7, 84.4, 74.3, 152.3, and 217.3 mL CH₄/dry g, respectively. While there was a general trend of increasing CH₄ vield with increasing cellulose plus hemicellulose (carbohydrate) content, many confounding factors precluded establishment of a quantitative relationship. Similarly, the degree of lignification of a particular component was not a good predictor of the extent of biodegradation.

In parallel with the decomposition experiments, leachate from the decomposition of each refuse constituent was analyzed for toxicity using a modified anaerobic toxicity assay. Leachate toxicity was not found in association with the decomposition of any refuse component other than food waste. However, substantial toxicity was measured in leachate from the food waste reactors. This toxicity was consistent with the behavior of the reactors but could not be simulated with high concentrations of carboxylic acids and sodium. The toxicity associated with food waste leachate is not likely to inhibit anaerobic decomposition in U.S. landfills due to the relatively low concentration of food waste in MSW.

Most probable number (MPN) tests were conducted to identify the components of refuse that carry refuse-decomposing microorganisms into landfills and to evaluate the significance of two typical cover soils as carriers of refuse-decomposing microbes. Grass, leaves, and branches were the major identifiable contributors of refuse-decomposing microbes to landfills, while the cover soils tested did not typically contain populations with the activities required for refuse methanogenesis.

This Project Summary was developed by EPA's Air Pollution Prevention and Control Division of the National Risk Management Research Laboratory, Research Triangle Park, NC, 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

Approximately 62% of the municipal solid waste (MSW) generated in the U.S. is disposed of by burial in a sanitary land-fill. The production of methane (CH₄) from sanitary landfills is well documented, and there are about 119 landfill gas recovery projects currently (January 1997) in operation in the U.S. and Canada. While the production of CH₄ from landfills is well established, there is large uncertainty involved in estimating the amount and rate of CH₄ production. This uncertainty is increasing as the composition of the MSW buried changes due to increased recycling.

Development of integrated solid waste management programs, which include recycling and in some cases, combustion, have led to a decrease in the use of landfills. However, there is a limit to the types of waste that can be recycled, and combustion has not been the solid waste management alternative of choice for many communities. Thus, landfills will be a significant part of MSW management for the foreseeable future.

Both CH_4 and carbon dioxide CO_2 are greenhouse gases that contribute to global climate change. CH_4 traps about 20 times more infrared energy than CO_2 on a volume basis. Consequently, although landfill gas contains approximately equal proportions of CH_4 and CO_2 , CH_4 is more significant with respect to atmospheric climate change. Data on the amount of CH_4 that can be expected from refuse already buried, as well as CH_4 that will result from the decomposition of refuse buried in the future, are needed to better assess the impact of landfills on global climate change.

The overall objective of this research was to develop information on the anaerobic decomposition of refuse that will improve our ability to assess the impact of sanitary landfills on global CH₄ accumulation. Three sets of experiments were conducted to meet this objective: (1) measurement of the CH₄ potential of the major biodegradable components of MSW; (2) assessment of whether leachate toxicity, associated with whole refuse or some individual constituent, inhibits the onset or rate of CH₄ production; and (3) identification of solid waste constituents that carry the anaerobic bacteria required for refuse methanogenesis. The results of each set of experiments are summarized separately.

Experiment 1: Measurement of the CH₄ Potential of the Major Biodegradable Components of MSW

The anaerobic biodegradability of the major biodegradable components of MSW was characterized by measurement of their CH₄ yield and the biodegradation of cellulose and hemicellulose. The components that were tested were grass, leaves, branches, food waste, and four types of paper-newsprint (ONP), old corrugated containers (OCC), office paper (OFF), and coated paper (CP). These are the most common types of paper in MSW and also represent the range of biodegradability that could be expected from paper. At one extreme, newsprint contains all of the lignin of wood pulp. At the other extreme, office paper has had almost all of the lignin removed. The decomposition of

mixed residential refuse was also characterized.

Tests were conducted in 2-L laboratory reactors in quadruplicate. Each refuse component was seeded with well-decomposed refuse to initiate refuse-decomposition. CH_4 yield data have been corrected for the background CH_4 produced from the seed. In the case of food waste, two sets of reactors were tested. In the first (F) series, there was insufficient seed, 30% by volume, and the reactors remained inhibited. A second set of food waste reactors (SF) was then initiated with 70% seed by volume, and these reactors produced measurable CH_4 .

The test conditions were designed to measure the maximum CH_4 production potential of each component. This included shredding, incubation at about 40°C, and leachate recycle and neutralization. All reactors were monitored until they were no longer producing measurable CH_4 , except for the old corrugated container reactors in which the CH_4 yield increased by less than 2% over the final 80 days of monitoring. At the termination of the monitoring period, reactors were destructively sampled for analysis of the residual solids.

The CH₄ yield, solids composition, and extent of cellulose and hemicellulose decomposition for each MSW component and mixed MSW are presented in Table 1. As summarized in Table 1, there was substantial variation in the range of CH₄ yields (30.6 to 300.7 mL/dry g) and the extent of decomposition (28 to 94%) among the components tested. In previous research with mixed refuse, carbohydrates accounted for 91% of the stoichiometric CH, potential of MSW. Carbohydrates were the major organic compounds analyzed in the waste components tested here, and the relationship between carbohydrate concentration and CH₄ yield is presented in Figure 1. While the data in Figure 1 show a relationship, the relatively low correlation coefficient ($r^2 = 0.49$) and failure of the regression line to pass through zero, suggest that factors in addition to carbohydrate concentration influence CH₄ yield.

Lignin decreases carbohydrate bioavailability and is expected to confound the relationship presented in Figure 1. The components with the lowest yields are the two sets of seed reactors and leaves. These are also the components with the lowest carbohydrate to lignin [(C+H)/Li)] ratio. The (C+H)/Li ratio is a measure of the degree of lignification. Values of 3 to 4 have been reported for fresh refuse, and lower values are associated with decomposed refuse. There is a general trend of more extensive cellulose biodegradation (MC decreasing) in the less lignified substrates [(C+H)/Li increasing] e.g., food and office paper ($r^2 = 0.28$). However, the quantitative relationship is weak because the office paper (C + H)/Li is well above any of the other components tested. The trend towards increased cellulose loss with a decreasing degree of lignification is most definite among the four paper components.

There is not a linear relationship between (C+H)/Li and the extent of decomposition ($r^2 = 0.02$). However, it is interesting to note that grass, which is highly lignified, underwent nearly complete decomposition as measured by either MC or the extent of decomposition (Table 1). This suggests that the lignin concentration does not always reflect the degree to which lignin inhibits cellulose bioavailability. Apparently, the lignins in grass are not as restrictive to microorganisms as the lignin in other components such as branches. This result is consistent with a report that stated, ". . .the chemistry of grass lignocellulose varies considerably from that of wood."

The solids decomposition (MC and MH) and CH₄ yield data document the biodegradability of even the most lignified substrates, leaves and branches, as well as all other components of MSW tested. The absence of good linear relationships is likely because a number of factors influence CH₄ production and solids decomposition. The biodegradation of newsprint measured here is in contrast to reports on the excavation of readable newsprint that had been buried in landfills decades earlier: however, these reported data did not represent average values, but rather observations during an archaeological excavation. The presence of readable newsprint that had not undergone biodegradation may be due to its isolation from other factors required for biodegradation such as bacteria, moisture, and nutrients. The biodegradability of a newspaper buried in a bag that did not break during waste compaction would differ from the biodegradability of newsprint exposed to other refuse components.

Based on the CH_4 yields presented in Table 1, a model was constructed to estimate CH_4 yields based on assumed compositions of buried refuse. These results are summarized in Table 2. The actual methane yield per wet kg of refuse buried decreases by only 10% between the base case (64.9 L CH_4 /wet kg) and the case with the most recycling (58.6 L CH_4 /wet kg). However, the appropriate way to evaluate changes in methane yield is to calculate the change in methane potential

Reactor	Yield mL CH₄/	Cellulose	Hemi- cellulose	Lignin	MOb	MUb	Extent of
Seed sd	25.5 5.7	23.4	4.7	22.5	0.18 0.02	0.36 0.03	21.8
Seed-2⁴ sd	5.8 0.6	18.3	3.7	22.1	0.34 0.01	0.69 0.11	6.3
Grass sd	144.4 15.5	26.5	10.2	28.4	0.19 0.01	0.42 0.06	94.3
Raleigh grass sd	127.6 21.8	25.6	14.8	21.6			75.5
Leaves sd	30.6° 8.6	15.3	10.5	43.8	0.43 0.05	0.68 0.10	28.3
Branch sd	62.6° 13.3	35.4	18.4	32.6	0.52 0.05	0.59 0.02	27.8
Food		46.1	6.2	8.3			
Second Food	300.7°	55.4	7.2	11.4	0.24	0.58	84.1
sa	10.6				0.02	0.04	
ONP sd	74.33 6.802	48.5	9	23.9	0.73 0.05	0.46 0.06	31.1
OCC sd	152.3 6.7	57.3	9.9	20.8	0.36 0.01	0.38 0.01	54.4
OFF sd	217.3 14.96	87.4	8.4	2.3	0.02 0	0.09 0.01	54.6
CP sd	84.4 8.1	42.3	9.4	15	0.54 0.01	0.58 0.06	39.2
MSW sd	92.0 ^e 4.1	28.8	9	23.1	0.25 0.03	0.22 0.05	58.4

Table 1. CH₄ Yield and Initial and Final Solids Composition Data Summary^a

a Data represent the average for each reactor set. Standard deviations (sd) are presented below the average where data are the average of all reactors in a set.

^b The ratio of the cellulose (MC) or hemicellulose (MH) recovered from a reactor divided by the mass added initially.

• The measured CH₄ yield divided by the yield calculated by assuming conversion of 100% of the cellulose and hemicellulose (and protein in the case of food waste) to CH₄ and CO₂.

^d Seed used for second set of food waste reactors.

• Yield data for the leaf reactors exclude L2, data for the branch reactors exclude B4, and data for the second food and MSW reactors were corrected for leakage.

based on the yield multiplied by the mass landfilled. Using this calculation, the potential reduction in methane production is 25.5% and 38% for the recycling cases based on national averages and local recycling rates, respectively. Thus, these data suggest that recycling can have a substantial impact on the volume of methane available for recovery over the decomposition period.

Where CH_4 is released to the atmosphere, recycling clearly reduces the amount of CH_4 released from landfills. However, at landfills where there is an active program to compare the relative benefits of recycling and energy recovery. Given the CH_4 potential data for individual constituents measured here, this analysis could be done on a component-specific basis because the results may be different for two different types of paper or between yard waste and paper.

The calculated composite CH₄ yields in Table 2 range from 58.6 - 64.9 L CH₄/wet kg of MSW. These values are low relative to landfill gas models that generally assume a yield of 62.3 to 112.2 L/kg. This is surprising in that the CH₄ yields measured here were measured under optimal conditions and should be considerably higher than values assumed for field conditions. There are two potential explanations for this discrepancy. The first explanation is that the assumed waste composition is in error. The data presented in an EPA report represent an estimate of MSW generation and exclude a number of wastes that are buried in landfills. Some of these other wastes have high CH₄ yields (wastewater treatment plant sludge and agricultural and food preparation wastes), while others have little or no CH₄ potential (water treatment plant sludge and construction and demolition debris). A second explanation for the discrepancy in yield calculations pertains to the assumptions used by the landfill gas models. The range of values used, 62.3 to 112.2 L/kg, is based on field measurements and an estimate of the mass of waste buried in a landfill. While this mass is accurately known in newer landfills where all waste received is weighed, this mass represents only an estimate at older facilities and errors of 20 to 30% would not appear to be unreasonable. Thus, the values assumed in practise may be inaccurate.



Figure 1. CH₄ yield vs. carbohydrate concentration.

Table 2. Calculated CH, Yield Based on Measured Yields and Assumed MSW Composition

Case	Yield (L CH₄/ wet kg)	Methane Reductionª (%)	Recycle Rate (%)
Base Case-No Recycling	64.9	na	na
Recycling at National Average	59.9	25.5	19.4
Recycling-Typical Local Program	58.6	38.0	30.9

^a Calculated from the CH₄ yield multiplied by the mass buried after recycling relative to the CH₄ yield and mass buried in the base case.

Experiment 2: Measurement of the Anaerobic Toxicity of Leachate Associated with the Decomposition of Individual Refuse Components

The anaerobic toxicity of leachate associated with the decomposition of each refuse component tested above was measured in parallel with the decomposition experiments. Leachate was sampled three times from each reactor. For food waste, four samples were collected from the F reactors, but no samples were collected from the SF reactors. Six samples were collected from the MSW reactors. The initial strategy was to sample each reactor twice during the acid phase and twice during the decelerated CH_4 production phase. However, except for the first set of food reactors (F), the acid phase was very brief. As a result, only one sample was collected from most reactor sets during the acid phase.

Leachate toxicity was evaluated using a modified anaerobic toxicity assay (ATA). The ATA included anaerobic medium, ground refuse as a carbon source, and an inoculum. The inoculum was a methanogenic consortium enriched from decomposed refuse with ground refuse as a carbon source. CH₄ production from the ground refuse was measured in triplicate in the presence and absence of leachate. Leachate was tested at final concentrations in the ATA of 25 and 80% of its original strength. Two sets of controls were also inoculated. Controls containing inoculum and medium but no refuse were used to measure background CH₄ production from the inoculum. Controls containing inoculum, medium, and ground refuse were used to compare CH_4 production in the presence and absence of leachate.

Leachate toxicity was not measured in association with the decomposition of any refuse component other than food waste. However, leachate associated with the food waste reactors containing 30% seed and 70% food waste (F) exhibited substantial toxicity, and this toxicity was generally consistent with the behavior of the reactors.

The toxicity of the food waste leachate could not be simulated with synthetic leachate containing high concentrations of carboxylic acids and sodium. ATAs with 20, 5, 15, and 12 g/L of acetate, propionate, butyrate, and sodium, respectively, suggested that high concentrations of butyric acid and sodium inhibited the onset of CH₄ production, but that refuse microorganisms could acclimate to these concentrations within 5 to 10 days under the conditions of the ATA. The corresponding concentrations of undissociated acetic, propionic, and butyric acids were 113, 27, and 96.8 mg/L, respectively. Comparison of carboxylic acid concentration data from the S and SF reactors series indicated that the refuse ecosystem was able to tolerate and recover from 142, 35, 24, and 305 mg/L of undissociated acetic, propionic, i-butyric, and butyric acids, respectively. These concentrations of undissociated, carboxylic acids are higher than concentrations reported to be inhibitory in previous research with anaerobic digesters.

Experiment 3: Identification of Solid Waste Constituents that Carry the Anaerobic Bacteria Required for Refuse Methanogenesis

The objective of part of this study was to identify the components of refuse that carry refuse-decomposing microorganisms into landfills. A second objective was to evaluate the significance of two typical cover soils as carriers of refuse-decomposing microbes. Refuse buried in a sanitary landfill is typically covered with 15 cm of soil daily. Recently, geotextile sheets and foams have been proposed as alternatives to soil to minimize the volume of soil in a landfill. While soil may contribute refuse-decomposing microorganisms to landfills, the proposed alternatives almost certainly do not.

The total anaerobic population and the subpopulations of cellulolytic, hemicellulolytic, hydrogen-producing acetogenic (based on butyrate catabolism) bacteria and acetate- and hydrogen $(H_2)/CO_2$ -utilizing methanogenic bacteria were enumerated by the most probable number (MPN) technique on several waste components: grass, branches, leaves, food waste, whole refuse, and landfill cover soil. For each component, the objective was to enumerate microbial populations on a representative sample in the form in which it would typically enter a landfill. Although paper represents 37.6% of refuse, it was not tested because it is likely populated with bacteria originating in wet components of refuse.

Microbial enumerations were performed by MPN tests. Thus, it was necessary to form a liquid inoculum from solid samples. The technique used here was similar to a technique developed previously to process smaller samples. In the laboratory, refuse samples were placed in a 113-L plastic garbage can which had been wiped with ethanol and purged with sterile argon. A measured volume of filter sterilized anaerobic phosphate buffer (23.7 mM, pH 7.2) was then added to a sample to form a slurry. The sample was then stirred by hand (covered with arm length gloves). Next, four samples were removed using a 1-L beaker, and the liquid was poured into a sterile, 4-L, argon-purged, Erlenmeyer flask. The liquid in this flask served as the inoculum for MPN enumerations. Inocula

were serially diluted in phosphate buffer (23.7 mM, pH 7.2). For soil, 250 to 300 g of each sample was added directly to a nitrogen-purged flask, 2.5 L of sodium pyrophosphate (0.1%, pH 7) was added, and the slurry was shaken for 2 minutes. The slurry was then allowed to settle for 1 minute after which a liquid sample was removed for use as an inoculum.

Microbial populations on each waste component and whole refuse are reported in Table 3. Total anaerobic and hemicellulolytic populations were present on all components tested, while the presence of cellulolytic, acetogenic, and methanogenic bacteria was more limited. Thus, identification of the waste components that are the major contributors of cellulolytic, acetogenic, and methanogenic bacteria is evaluated here. Yard waste (grass, leaves, and branches) most consistently carried the microorganisms required for refuse methanogenesis. Surprisingly, food waste did not carry either cellulolytic or methanogenic bacteria, and one of two food waste samples contained only one acetogen per gram. Populations of cellulolytic, acetogenic, and methanogenic bacteria were generally lower in the mixed refuse samples compared to the grass, leaves, and branch samples.

Grass, leaves, and branches were the major identifiable contributors of refusedecomposing microbes to landfills. About 9% of the refuse stream is characterized as "miscellaneous" and contains many different items. In addition to diapers and pet wastes, there may be other components in the miscellaneous fraction that carry refuse-decomposing microbes. However, their presence is small, and they are likely to be poorly distributed. The importance of vard waste should be considered as solid waste management programs are implemented. Where there is interest in CH₄ recovery from landfills, banning yard waste from landfills may be self-defeating. Unless, of course, the landfill is receiving substantial volumes of other wastes known to carry refuse-decomposing microbes. The cover soils tested did not typically contain populations with the activities required for refuse methanogenesis. Thus, efforts to develop lower volume alternatives to cover soil will not adversely impact the input of refuse-decomposing microbes to landfills.

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Table 3.	Anaerobic Microbial	Populations	on Refuse	Components	(Most	Probable	Number-	-log ₁₀
	cells/dry g) ^a						- 10	

Trophic Group	Total Anaerobes	Hemicellu- lolytic	Cellulolytic	Acetogen	Methanogen Acetate	Methanogen H ₂ /CO ₂
Grass (April 92)	9.8	7.9	1.4	0.7	1.6	1.8
Grass (July 92)	9.8	9.5	b	1.8	Ь	1.3
Branches	6.5	4.2	2.5	1.3	1.1	0.8
Leaves (Nov. 91)	5.8	4.1	1.0	<0.4	1.0	0.7
Leaves (Nov. 92)	6.9	4.4	1.8	4.4	3.0	3.8
Food (Mar. 92)	>8.0°	5.3	<-0.4	<-0.1	<-0.1	<-0.1
Food (Aug. 92)	9.4	6.2	<-0.4	0	<0	<0
Refuse (July 92)	9.3	6.6	0.4	0.2	3.6	5.0
Refuse (Sept. 92)	8.4	6.3	<-0.2	<-0.1	<-0.1	0.8

^a Data reported as less than a number indicate that no positive tubes were detected. The number reported assumes

one positive tube in the first dilution.

^b MPN results code was anomalous and not reported.

^c All tubes were positive at the highest dilution tested.

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The complete report, entitled "Biodegradative Analysis of Municipal Solid Waste in Laboratory-Scale Landfills," (Order No. PB97-189674; Cost: \$35.00, subject to change) will be available only from

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