

NATIONAL COUNCIL FOR AIR AND STREAM IMPROVEMENT

CRITICAL REVIEW OF FOREST PRODUCTS DECOMPOSITION IN MUNICIPAL SOLID WASTE LANDFILLS

TECHNICAL BULLETIN NO. 872 MARCH 2004

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PRESIDENT'S NOTE

NCASI's investigative program addresses a number of technical questions related to the climate change issue, ranging from the science of climate change to the industry's greenhouse gas "footprint." In this report, Dr. Morton Barlaz, Professor and Associate Head of the Civil, Construction and Environmental Engineering Department at North Carolina State University, examines the data that can be used to characterize the carbon sequestration and greenhouse gas emissions associated with forest products that are disposed of in landfills. Dr. Barlaz is one of the leading authorities on the chemical and biological processes in municipal solid waste landfills and is the source of the data used by the U.S. government to estimate carbon sequestration in landfills.

A number of studies have been conducted on the fate of carbon in mixed municipal solid waste, but only a few have examined the contribution of forest products specifically. The studies of forest products demonstrate that the lignin in forest products is essentially nondegradable under the anaerobic conditions that exist in municipal solid waste landfills. In addition, lignin and certain other materials such as coatings tend to protect cellulose and hemicellulose from biological attack, limiting the decomposition of these otherwise degradable components of paper and wood. Using the published data, carbon storage factors can be developed that represent the nondegradable portion of the carbon in paper, paperboard, and wood products in landfills. This report contains a summary of the carbon storage factors which can be used to estimate the carbon sequestration attributable to forest products in municipal solid waste (MSW) landfills.

The rate at which the degradable carbon in municipal solid waste is converted into landfill gas is affected by a number of variables, most notably moisture content, pH, and nutrient availability. The estimated degradation rates for mixed municipal solid waste vary by more than an order of magnitude. The literature provides little basis for identifying degradation rates specific to forest products in landfills.

In his analysis, Dr. Barlaz identifies several opportunities for improving the understanding of the fate of carbon in landfilled forest products. These include developing data on the degradability of wood products, and undertaking a rigorous analysis of the available field data to determine if they can be used to reduce the uncertainty in degradation rates.

Part of the funding for this project was provided by the International Council of Forest and Paper Associations. NCASI gratefully acknowledges this support.

Pm the

Ronald A. Yeske

March 2004



au service de la recherche environnementale pour l'industrie forestière depuis 1943

MOT DU PRESIDENT

Le programme de recherche de NCASI aborde différentes questions techniques liées au dossier du changement climatique allant de la science du changement climatique à l'empreinte « écologique » des émissions de gaz à effet de serre causées par les activités de l'industrie. Dans ce rapport, Dr. Morton Barlaz, professeur et chef adjoint au département de génie civil, construction et environnement à la *North Carolina State University*, examine les données pouvant être utilisées pour caractériser la séquestration du carbone et les émissions de gaz à effet de serre associées aux produits forestiers qui sont envoyés à l'enfouissement. Dr. Barlaz est une autorité reconnue dans la compréhension des processus chimiques et biologiques qui se produisent dans les sites d'enfouissement de déchets solides municipaux et est à l'origine des données qu'utilise le gouvernement américain pour estimer la séquestration du carbone dans les sites d'enfouissement.

Il existe un certain nombre d'études sur le devenir du carbone dans les déchets solides municipaux mixtes, mais il n'y en a que quelques-unes qui ont porté spécifiquement sur la contribution des produits forestiers. Ces études révèlent que la lignine des produits forestiers est essentiellement non dégradable dans les conditions anaérobiques qui prévalent dans les sites d'enfouissement de déchets solides municipaux. De plus, la lignine et certaines autres matières comme les sauces de couchage contribuent à protéger la cellulose et l'hémicellulose d'une attaque biologique limitant ainsi la décomposition de composantes du papier et du bois qui autrement se dégradent. Il est possible d'élaborer des facteurs d'accumulation du carbone qui représentent la portion non dégradable du carbone dans les papiers, cartons et produits du bois qui se retrouvent dans les sites d'enfouissement à l'aide des données déjà publiées. Ce rapport résume les facteurs d'accumulation du carbone qui peuvent être utilisés pour estimer la séquestration du carbone attribuable aux produits forestiers dans les sites d'enfouissement de déchets solides municipaux (DSM).

Le taux de dégradation du carbone dans les déchets solides municipaux pour convertir le carbone en gaz d'enfouissement est influencé par un certain nombre de variables dont les principales sont le taux d'humidité, le pH et la disponibilité des nutriments. Les taux estimés de dégradation des déchets solides municipaux mixtes varient de plus d'un ordre de grandeur. Il y a peu de références dans la documentation existante pour identifier les taux de dégradation caractéristiques des produits forestiers dans les sites d'enfouissement.

Dans son analyse, le Dr. Barlaz met en évidence plusieurs possibilités pour améliorer la compréhension du devenir du carbone dans les produits forestiers envoyés à l'enfouissement, notamment l'élaboration de données sur la dégradabilité des produits du bois et l'analyse rigoureuse des données de terrain disponibles pour déterminer si ces dernières peuvent être utilisées pour diminuer l'incertitude entourant les taux de dégradation.

Ce projet a été financé en partie par l'*International Council of Forest and Paper Associations*. NCASI remercie sincèrement l'ICFPA de cet appui.

Pm Johne

Ronald A. Yeske

Mars 2004

National Council for Air and Stream Improvement

CRITICAL REVIEW OF FOREST PRODUCTS DECOMPOSITION IN MUNICIPAL SOLID WASTE LANDFILLS

TECHNICAL BULLETIN NO. 872 MARCH 2004

ABSTRACT

This report reviews the published information on the factors that affect the decomposition of forest products in municipal solid waste (MSW) landfills and the data available for estimating the fate of carbon in these products.

Estimates of carbon sequestration in, and greenhouse gas emissions from, municipal solid waste (MSW) landfills are usually derived from mathematical models. A variety of models have been developed for this purpose, but the most commonly used models apply first order decay kinetics to the degradable fraction of the carbon and assume that the nondegradable carbon remains in the landfill indefinitely. Two parameters, therefore, are needed to use the models: the nondegradable fraction of the waste, and the first order decay rate for the degradable fraction.

Laboratory studies have been used to determine the ultimate degradability of a number of forest products in a landfill environment. The data can be used to develop carbon storage factors representing the fraction of the carbon in the original product that is nondegradable in the landfill. Carbon storage factors vary from approximately 0.1 to over 0.8 depending on the product. Higher carbon storage factors are associated with wood products and paper/paperboard products that are high in lignin content (e.g., lumber and newsprint) or are coated. Carbon storage factors for a variety of industry products are summarized in this report.

The rate at which the degradable carbon municipal solid waste is converted to landfill gas is determined by a number of factors, most notably moisture content, pH, and nutrient availability. First order decay rates for municipal solid waste vary by more than an order of magnitude. The rates identified in this review range from 0.003 to 0.094 yr⁻¹, although most of the values are in the range of 0.02 to 0.07 yr⁻¹. Well-documented decay rates for individual components of the municipal solid waste stream, including different types of discarded forest products, are not available.

KEYWORDS

anaerobic decomposition, carbon sequestration, greenhouse gases, landfills, methane, municipal solid waste

RELATED NCASI PUBLICATIONS

Calculation Tools for Estimating Greenhouse Gas Emissions from Pulp and Paper Mills, Version 1.0. November 2002. http://www.ncasi.org/pulpandpaperreport.pdf.

Characterizing Carbon Sequestration in Forest Products along the Value Chain. NCASI report to the International Council of Forest and Paper Associations. December 26, 2003. http://www.ncasi.org/publications/carbon sequestration.pdf.

ANALYSE CRITIQUE DE LA DECOMPOSITION DES PRODUITS FORESTIERS DANS LES SITES D'ENFOUISSEMENT DE DECHETS SOLIDES MUNICIPAUX

BULLETIN TECHNIQUE NO. 872 MARS 2004

RÉSUMÉ

Ce rapport est une analyse de l'information qui a été publiée sur les facteurs qui influencent la décomposition des produits forestiers dans les sites d'enfouissement de déchets solides municipaux (DSM) et les données disponibles pour estimer le devenir du carbone dans ces produits.

Les estimations liées à la séquestration du carbone dans les sites d'enfouissement de déchets solides municipaux (DSM) et les gaz à effet de serre qui en découlent proviennent habituellement de modèles mathématiques. On a conçu divers modèles à cette fin, mais ceux qui sont les plus couramment utilisés font appel à une cinétique de décomposition de premier ordre pour la fraction dégradable du carbone et supposent que le carbone non dégradable demeure dans le site d'enfouissement indéfiniment. Par conséquent, deux paramètres sont nécessaires pour utiliser les modèles: la fraction du déchet qui est non dégradable et le taux de décomposition de premier ordre de la fraction dégradable.

Des études de laboratoire ont été utilisées pour déterminer la dégradabilité maximale d'un certain nombre de produits forestiers dans un contexte d'enfouissement. Les données peuvent servir à élaborer des facteurs d'accumulation du carbone qui représentent la fraction du carbone dans le produit original qui est non dégradable dans le site d'enfouissement. Les facteurs d'accumulation du carbone varient d'environ 0,1 à plus de 0,8 selon le produit. Les facteurs les plus élevés sont associés aux produits du bois et aux produits de papier ou de carton dont le taux de lignine est élevé (par ex. le bois d'œuvre et le papier journal) ou qui sont couchés. Les facteurs d'accumulation du carbone de divers produits de l'industrie sont présentés dans ce rapport.

Le taux de dégradation du carbone dans les déchets solides municipaux pour convertir le carbone en gaz d'enfouissement est influencé par un certain nombre de variables dont les principales sont le taux d'humidité, le pH et la disponibilité des nutriments. Les taux de décomposition de premier ordre pour les déchets solides municipaux mixtes varient de plus d'un ordre de grandeur. Dans cette analyse, les taux vont de 0,003 à 0,094 an⁻¹ quoique la majorité des valeurs se situent dans l'intervalle entre 0,02 et 0,07 an⁻¹. Des taux de décomposition bien documentés ne sont pas disponibles pour les composantes individuelles du flux de déchets solides municipaux, incluant les différents types de produits forestiers mis au rebut.

MOTS CLES

Déchets solides municipaux, décomposition anaérobique, gaz à effet de serre, méthane, séquestration du carbone, sites d'enfouissement

AUTRES PUBLICATIONS DE NCASI DANS CE DOMAINE

Calculation Tools for Estimating Greenhouse Gas Emissions from Pulp and Paper Mills, Version 1.0, novembre 2002, http://www.ncasi.org/pulpandpaperreport.pdf.

Characterizing Carbon Sequestration in Forest Products along the Value Chain, Rapport préparé par NCASI pour l'*International Council of Forest and Paper Associations*, 26 décembre 2003, http://www.ncasi.org/publications/carbon_sequestration.pdf.

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CRITICAL REVIEW OF FOREST PRODUCTS DECOMPOSITION IN MUNICIPAL SOLID WASTE LANDFILLS

1.0 INTRODUCTION

The overall objectives of this report are to summarize information on the decomposition of materials produced by the forest products industry that are disposed of in landfills and to identify additional information that is needed to quantify the rate and ultimate extent of decomposition of these materials. An understanding of the rate and extent of paper and wood products decomposition in landfills is required to quantify the associated production of greenhouse gases as well as carbon sequestration. This report begins with a summary of the state of solid waste management in the U.S. and a presentation of waste composition data that documents the large amounts of cellulose buried in U.S. landfills. The second section of this report presents background information on the biological decomposition of refuse in landfills. The third section of the report summarizes literature on the decomposition of municipal solid waste (MSW) in general, and paper and wood products in particular, in both landfills and laboratory- and field-scale simulations. The existing literature is then analyzed to summarize the state of current knowledge and the types of information that are required to generate improved estimates of greenhouse gas emissions and carbon sequestration associated with paper and wood products in landfills. The fourth section of this report summarizes existing information on methane production models. The final section makes recommendations for future work to better quantify greenhouse gas emissions and carbon sequestration attributable to landfilled paper and wood products.

1.1 The State of Solid Waste Management in the U.S.

The most authoritative and consistent source of information on MSW management in the U.S. is the annual waste characterization report published by USEPA (USEPA 2002a). This report contains statistics on the manner in which MSW is managed, the composition of MSW as generated, and the quantities of materials that are recycled. While valuable, the data in this report are limited to wastes defined as MSW by USEPA. The EPA's definition of MSW includes wastes generated in the residential, multifamily, commercial, and institutional sectors but does not include many other wastes that are sometimes buried in landfills including construction and demolition (C&D) waste, water and wastewater treatment sludges, and non-hazardous industrial wastes. Thus, while key information from the USEPA report is summarized in this section, it is not a precise description of all waste generated or buried in landfills in the U.S. Limited waste composition data for other countries are presented in Appendix A.

As illustrated in Figure 1.1, in 2000, 55.3% of MSW was buried in landfills while lesser amounts were recycled, composted, and combusted in waste-to-energy facilities. The relative fraction of MSW buried in landfills has not changed significantly over the past several years although the number of landfills has decreased substantially, from about 8000 in 1988 to about 2000 in 2000. This decrease reflects a trend towards larger regional landfills that receive waste from larger areas.

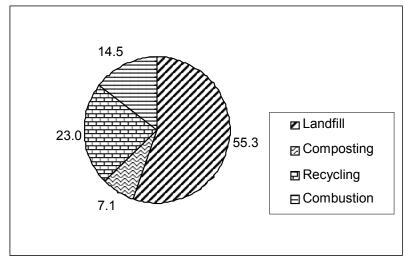


Figure 1.1 Management of MSW in the U.S. (USEPA 2002a)

Historically, landfills have been operated to minimize moisture infiltration, which has the effect of reducing the rate and extent of waste decomposition. More recently, there has been increased interest in the operation of landfills as bioreactors, in which leachate and perhaps other liquids are recirculated through the waste to promote decomposition (Pacey et al. 1999). Potential advantages include:

- a. reduced potential for cracking of the final cover because it is placed after decomposition and settling are almost complete
- b. additional landfill capacity due to rapid refuse decomposition
- c. reduced leachate treatment costs
- d. improved economics of gas recovery as higher amounts are produced over a shorter time
- e. reduced costs for long term monitoring because the landfill stabilizes faster.

Potential problems with this technology include:

- a. increased potential for odor due to increased rates of gas production
- b. leachate breakouts
- c. increased complexity of operation due to the need for liquids management.

As indicated above, C&D waste is not included in USEPA's definition of MSW. This is particularly important because the quantity of C&D waste entering landfills is significant. For example, a recent citation to the Construction Materials Recycling Association suggests that C&D debris accounts for 25 to 45% of the overall North American waste stream (McLendon, Townsend, and Cochran 2002). A survey of the waste entering landfills in Missouri indicated that about 18.5% of the entering mass was C&D waste (Siders 1999). Micales and Skog (1997) estimated that C&D waste comprised 28% of the mass entering landfills in 1993. There has been increased emphasis on the recycling of C&D waste over the past five years, so the quantity buried in landfills is likely decreasing with time. In some states, C&D waste must be buried in segregated landfills. When segregated, C&D waste likely decomposes at a slower rate than when buried in traditional landfills due to reduced levels of moisture and nutrients.

1.2 Composition of Municipal Solid Waste

The composition of MSW as generated and the recycling rates for selected materials are presented in Table 1.1. An estimate of C&D waste composition in Florida is presented in Table 1.2 (McLendon, Townsend, and Cochran 2002). Micales and Skog (1997) estimated that 25% of C&D waste is wood products and that another 25% of C&D waste contains some cellulosic materials. The authors use these numbers to estimate that C&D waste contains 30% wood, which is comparable to the sum of wood and yard waste in Table 1.2.

	Composition	Recovery ^a
Waste Component	(%)	(%)
Total Paper	37.4	5 0 0
Newsprint (ONP)	5.3	58.2
Office Paper (OFF)	3.2	54.1
Magazines (OMG)	0.9	31.9
Corrugated Boxes (OCC)	13.0	70.7
Other Paper	15.0	17.3
Total Metals	7.8	
Aluminum Cans	0.7	54.6
Steel Cans	1.1	58.0
Other Metals	6.0	
Total Plastics	10.7	
PET	0.4	34.9
Milk and Water Bottles		
(HDPE)	0.3	30.4
Other Plastic	10.0	
Total Glass	5.5	
Glass Containers	4.8	26.3
Other Glass	0.7	
Food Waste	11.2	
Yard Waste	12.0	
Rubber and Leather	2.7	
Textiles	4.0	
Wood	5.5	
Other	1.7	
Miscellaneous Inorganic	1.5	
Total	100	

 Table 1.1 Composition of MSW in the U.S. in 2000 (USEPA 2002a)

^a recovery as a % of waste generation

Component	% of Dry Weight			
Asphalt Shingles	3			
Clay/Concrete Shingles	5			
Concrete	30			
Wood	17			
Yard Trash	11			
Land-Clearing Debris	8			
Asphalt Concrete	7			
Metals	8			
Drywall	6			
Cardboard	2			
Miscellaneous	3			

Table 1.2 Estimated Composition of Construction and DemolitionWaste in Florida (McLendon, Townsend, and Cochran 2002)

Another way to characterize the composition of waste is by analysis of its organic components. Cellulose and hemicellulose are the principal biodegradable components of MSW. The other major organic component, lignin, is at best only slowly degradable under methanogenic conditions (Colberg 1988). In addition, lignin interferes with the decomposition of cellulose and hemicellulose by physically impeding microbial access to these degradable carbohydrates. The cellulose, hemicellulose, and lignin concentrations for a series of waste samples collected over the past 14 years are summarized in Table 1.3. In all cases, the samples represent residential refuse as opposed to a mixture of all types of MSW and C&D waste. Thus, these samples do not include the large quantities of office paper or wood waste typically generated in the commercial and construction sectors. The last column in Table 1.3 represents values for the aggregate composition of waste entering a Canadian landfill. These values were calculated from measurement of the cellulose, hemicellulose, and lignin content of individual waste components and a waste composition survey. The composition of the aggregated waste stream includes some inert wastes which dilute the cellulose, hemicellulose, and lignin concentrations. Finally, the analysis of 87 samples of 1 to 3 year-old refuse recovered from a traditional landfill in Kentucky exhibited average cellulose, hemicellulose, and lignin concentrations of 36.8, 8.2 and 18.5%, respectively, with an average cellulose plus hemicellulose to lignin (CH:L) ratio of 2.59 (sd = 1.0) (Barlaz, unpublished data).

Available data on the cellulose, hemicellulose, and lignin concentrations in paper and wood waste are summarized in Table 1.4. It is interesting to note the relatively large difference in the cellulose content of the two office papers that were tested. The office paper sampled in Wu et al. (2001) had an ash content of 11.6% compared to 1.4% in the sample obtained by Eleazer et al. (1997). This difference is likely due to the presence of inorganic coatings in the sample obtained by Wu et al.

Reference	Barlaz, Schaefer, & Ham 1989 ^a	Eleazer et al. 1997	Rhew & Barlaz 1995	Ress et al. 1998	Barlaz Unpub- lished	Price Unpub- lished	Price Unpub- lished	Barlaz Unpub- lished ^b
Cellulose	51.2	28.8	38.5	48.2	36.7	43.9	43.5	22.4
Hemicellulose	11.9	9.0	8.7	10.6	6.7	10.0	8.4	5.8
Lignin	15.2	23.1	28.0	14.5	13.6	25.1	33.5	11.0
(Cellulose + hemicellulose)/ lignin	4.15	1.64	1.68	4.06	3.19	2.15	1.55	2.57
Volatile Solids	78.6	75.2	NM	71.4	NM			NM

 Table 1.3 Organic Composition of Residential Refuse (% of Dry Weight)

^a The following additional analyses were performed on this sample: protein -4.2%, soluble sugars -0.35%, starch -0.6% and pectin -3%.

^b These values represent the composition of waste entering a Canadian landfill as described in the text. NM not measured

					Corrugated	Coated	
	Ne	wsprint	Offic	e Paper	Cardboard	Paper	Branches
	Wu	Eleazer	Wu	Eleazer	Eleazer	Eleazer	Eleazer
	et al.	et al.	et al.	et al.	et al	et al.	et al.
	2001	1997	2001	1997	1997	1997	1997
Cellulose	48.3	48.5	64.7	87.4	57.3	42.3	35.4
Hemicellulose	18.1	9	13.0	8.4	9.9	9.4	18.4
Lignin	22.1	23.9	0.93	2.3	20.8	15.0	32.6
Volatile solids	98.0	98.5	88.4	98.6	92.2	74.3	96.6

 Table 1.4 Chemical Composition of Paper Products Present in Municipal Waste

In summary, although the exact composition of waste that is buried in landfills varies, it is well established that the waste contains large concentrations of cellulosic materials. Paper is the major contributor to the cellulose and hemicellulose concentrations in landfills, followed by contributions from wood, yard debris, and food waste. With this basis for the type and composition of material that enters landfills, the decomposition of waste in landfills is reviewed in the following section.

2.0 LANDFILL MICROBIOLOGY

The decomposition of refuse to methane in landfills is a microbially mediated process which requires the coordinated activity of several trophic groups of bacteria. As described in the previous section, the principal substrates which decompose to methane in landfills are cellulose and hemicellulose. In the first part of this section, the general pathway for anaerobic decomposition is reviewed. Following this general pathway review, a four phase description of refuse decomposition is presented.

2.1 The Microbiology of Anaerobic Decomposition

Three trophic groups of anaerobic bacteria are required for the production of methane from biological polymers (cellulose, hemicellulose, and protein) as illustrated in Figure 2.1 (Zehnder, Ingvorsen, and Marti 1982). The first group of microorganisms is responsible for the hydrolysis of biological polymers. The initial products of polymer hydrolysis are soluble sugars, amino acids, long chain carboxylic acids, and glycerol. Hydrolytic and fermentative microorganisms then ferment these initial products to short-chain carboxylic acids (primarily propionic and butyric acids), alcohols, carbon dioxide, and hydrogen. Acetate, a direct precursor of methane, is also formed.

The second group of bacteria active in the conversion of biological polymers to methane is the obligate proton-reducing/fatty acid oxidizing acetogens. They oxidize the fermentation products of the first group of microorganisms to acetate, carbon dioxide, and hydrogen. The conversion of fermentation intermediates like butyrate, propionate, and ethanol is only thermodynamically favorable at very low hydrogen concentrations. Thus, these substrates are only utilized when the obligate proton-reducing acetogenic bacteria can function in syntrophic association with a hydrogen scavenger such as a methane-producing or sulfate-reducing organism.

The methanogens are the third group of bacteria necessary for the production of methane. The methanogens can utilize only a limited number of substrates including acetate and hydrogen, which are the major precursors of methane in landfills. The methanogens are most active in the pH range 6.8 to 7.4 (Zehnder 1978). As a group, the methanogens control the pH of their ecosystem by the consumption of acetate and regulate the flow of electrons by the consumption of hydrogen, creating thermodynamically favorable conditions for the catabolism of alcohols and acids. Should the activity of the fermentative organisms exceed that of the acetogens and methanogens, there will be an imbalance in the ecosystem. Carboxylic acids and hydrogen will accumulate and the pH of the system will fall, thus inhibiting methanogenesis.

2.2 The Microbiology of Refuse Decomposition

A complex series of chemical and biological reactions is initiated with the burial of refuse in a landfill. During the initial aerobic phase, oxygen present in the void spaces of the freshly buried refuse is consumed, resulting in the production of CO2 and an increase in waste temperature due to the waste heat of aerobic metabolism. The aerobic phase in a landfill should last only a few days because oxygen is not replenished once the waste is covered. During the aerobic phase, the waste is typically not saturated (Barlaz and Ham 1993). Most leachate produced during this phase results from short-circuiting of precipitation through the buried refuse.

As oxygen is depleted, the waste becomes anaerobic, which supports fermentation reactions. In the second phase of decomposition, the hydrolytic, fermentative, and acetogenic bacteria dominate, resulting in an accumulation of carboxylic acids, and a pH decrease. The highest BOD and COD concentrations in the leachate will be measured during this phase (Barlaz and Ham 1993; Reinhart and Grosh 1998; Kjeldsen et al. 2003). The BOD:COD ratio in the acid phase has been reported to be above 0.4 (Ehrig 1988) or 0.7 (Robinson 1995). As the pH is acidic, acid phase leachate is chemically aggressive and will increase the solubility of many metals.

The onset of the initial methanogenic phase (3) occurs when measurable quantities of methane are produced. The onset of this phase is associated with the pH of the refuse becoming sufficiently neutralized for at least limited growth of methanogenic bacteria. During this phase the acids that accumulated in the acid phase are converted to methane and carbon dioxide by methanogenic bacteria and the methane production rate will increase to a maximum (Barlaz,

Shaefer, and Ham 1989). Cellulose and hemicellulose decomposition also begins. COD and BOD concentrations begin to decrease and the pH increases as acids are consumed.

In the stable (or decelerated) methanogenic phase, the methane production rate will decrease from its maximum as the pool of soluble substrate (carboxylic acids) decreases. In this phase, the rate of CH_4 production is dependent on the rate of cellulose and hemicellulose hydrolysis. The pH stabilizes between 7 and 8 as steady-state carboxylic acid concentrations are relatively low. Some COD is present in the leachate but it is mostly recalcitrant compounds such as humic and fulvic acids (Barlaz and Ham 1993; Christensen et al. 1994). The BOD:COD ratio will generally fall below 0.1 in this phase as carboxylic acids are consumed as rapidly as they are produced.

The four phases of refuse decomposition described above have been defined on the basis of both field and laboratory-scale data that have been summarized in earlier reviews (Barlaz, Ham, and Schaefer 1990). However, environmental conditions in the landfill will have a significant impact on the rate of refuse decomposition and subsequently, the time required for decomposition to proceed to the point where methane production decreases to zero. Studies on the effect of a number of factors on refuse decomposition have been summarized (Barlaz, Ham, and Schaefer 1990). The factors that have most consistently been shown to affect the rate of refuse decomposition are the moisture content and pH. It is generally accepted that refuse buried in arid climates decomposes more slowly than refuse buried in regions that receive greater than 20 to 40 inches of annual infiltration into the waste.

The stoichiometric relationships for the conversion of cellulose and hemicellulose to methane are presented in equations 1 and 2 respectively.

$$(C_6H_{10}O_5)n + nH_2O \rightarrow 3nCO_2 + 3nCH_4$$
(1)

$$(C_5H_8O_4)n + nH_2O \rightarrow 2.5nCO_2 + 2.5nCH_4$$
 (2)

In each case, equal molar quantities of CO_2 and CH_4 are produced. Landfill gas typically contains more CH_4 because CO_2 is partially soluble in water and some dissolves in the leachate. As this leachate is released from the landfill and exposed to atmospheric conditions, the dissolved CO_2 would establish an equilibrium with the atmosphere and some CO_2 is likely released to the atmosphere.

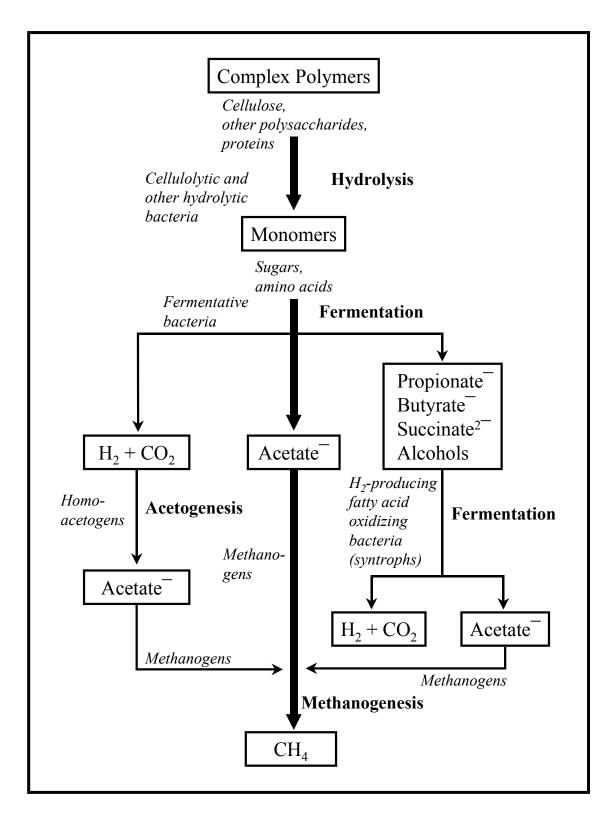


Figure 2.1 Overall Process of Anaerobic Decomposition Showing the Manner in Which Various Groups of Fermentative Anaerobes Act Together in the Conversion of Complex Organic Materials Ultimately to Methane and Carbon Dioxide (reprinted from Brock et al. 1994)

3.0 LITERATURE REVIEW

The objective of this section is to summarize relevant studies on waste decomposition in landfills. Because only a few studies focus specifically on paper and wood, all studies that report information on the decomposition of waste buried in landfills are reviewed. The first part of this section presents some background on the analysis of cellulose, hemicellulose, and lignin in refuse that will be useful for evaluation of the results of the published studies.

3.1 Analysis of Cellulose, Hemicellulose, and Lignin Concentrations in Samples of Excavated Refuse

The techniques used to measure the cellulose concentration have changed over the years. Initially, cellulose was measured based on a gravitimetric method originally described by Lossin (1971). In this method, the non-cellulosic material is dissolved in a combination of boiling nitric and glacial acetic acids, after which the cellulose content is quantified by loss on ignition at 550°C. This technique does not include hemicellulose. In this technique, any organic material that does not dissolve would be counted as cellulose. Thus, the reported cellulose content is likely to be artificially high. Later, a technique that utilized high pressure liquid chromatography (HPLC) was adopted (Petterson and Schwandt 1991). The basic procedure is hydrolysis of a solid sample in 72% (w/v) H_2SO_4 , followed by a secondary hydrolysis in 3% (w/v) H_2SO_4 . The hydrolyses convert cellulose and hemicellulose to their respective monomeric sugars. Typically, HPLC is used to quantify the concentrations of glucose, xylose, mannose, arabinose, and galactose in the acid hydrolyzate. The glucose originates from cellulose and the other sugars from hemicellulose.

The gravitimetric technique is likely to include some non-cellulosic material as cellulose and thus may give artificially high results. This problem should become worse in the samples that are most decomposed. This is because the concentration of non-cellulosic recalcitrant material will increase relative to the cellulose concentration in well decomposed samples. Ham, Norman, and Fritschel (1993) performed both analyses on 31 samples and the gravitimetric technique resulted in higher concentrations in all but three samples although the magnitude of the error was not reported. Similarly, Barlaz (1988) performed both analytical procedures on 17 samples and the gravitimetric technique resulted in higher concentrations in 16 of the samples. As predicted, the discrepancy was largest in the samples that were most decomposed.

Historically, daily cover has been applied to landfills. Thus, there is the potential to include soil in refuse samples that are excavated from landfills; this soil will dilute the cellulose, hemicellulose, and lignin concentrations. The most effective way to eliminate the effect of soil dilution is to analyze data based on the cellulose to lignin (C:L) or cellulose plus hemicellulose to lignin (CH:L) ratio. While advantageous from the perspective of eliminating soil, this method has the disadvantage that the initial ratio of the buried refuse is often unknown or only known within a range. As reported below, ratios of about 4 have been reported by Bookter and Ham (1982), while initial ratios of 1.64 to 4.15 were presented in Table 1.3. Both of these references apply to residential refuse which is somewhat different from what is actually buried in landfills.

In the following discussion, sampling and analysis methods are highlighted in several studies where they are suspected to have affected the results of the research.

3.2 Studies on the Decomposition of Refuse and MSW Components

Bookter and Ham 1982

Bookter and Ham (1982) reported on the analysis of samples from test lysimeters that contained approximately 91 mT (metric tons) of shredded MSW; their results are summarized in Table 3.1. Though not explicitly stated by the authors, it appears that a different lysimeter was sampled after 1, 6, and 9 years. The cellulose to lignin ratio decreased significantly at each time point, suggesting an overall trend of decomposition. The increased moisture content documents that moisture was likely not limiting decomposition. However, the acidic pH in the 6 and 9 year-old samples suggests that conditions for decomposition were not optimal in these samples. The C:L of the 9 year-old sample, 0.57, was higher than that of samples of non-shredded refuse excavated from landfills that are described below. Thus, the data presented in Table 3.1 suggest that decomposition was not complete.

One potential explanation for the failure of the refuse to completely decompose after 9 years is temperature, as the cells were only 4 feet deep. Cold temperatures during Wisconsin winters may have decreased the rate of decomposition. In describing the cells, the authors report temperatures of -1 to 10°C during the winter and 15.6 to 32.2°C during the summer. An alternate explanation for the absence of complete decomposition is that the measured cellulose content was artificially high based on the analytical protocol. However, the acidic pH suggests that pH limited decomposition and the presence of non-degraded cellulose would appear reasonable. The absence of complete decomposition after 9 years is interesting given the elevated moisture content and the fact that the refuse was shredded initially. It has implications for bioreactors where decomposition is expected to occur more rapidly.

Age (years)	pН	Moisture Content (%)	Cellulose %	Lignin %	C/L	Comments
Fresh	6.9	22.1	42.4	10.9	3.89	Samples taken from landfill on three successive days
1	6.9	52.7	28.8	13.9	2.07	Methane production had just begun at time of sampling
6	4.4	49.1	27.0	18.9	1.43	Erratic history of methane production and moderate leachate strength
9	5.3	38.5	8.2	14.5	0.57	Refuse had a humus- like appearance

Table 3.1 Decomposition of Refuse in Test Lysimeters (Bookter and Ham 1982)

Bookter and Ham also reported the results of samples excavated from four landfills across the U.S. Refuse that was estimated to be 2 to 10 years old was sampled from a landfill in Los Angeles that was also known to accept some non-hazardous liquid waste. Samples were taken at discrete depths from eight borings. Since refuse age increases with depth, more decomposed refuse would be expected in samples extracted from deeper in the landfill. However, only 2 to 3 samples were analyzed from the same boring, so definitive trends with respect to depth were not always apparent. In addition, the moisture content of the excavated refuse varied from 20 to 50% and the pH varied from 5.4 to 7.1. The lowest cellulose concentration and C:L measured were 7.2 % and 0.6, respectively. Cellulose concentrations can be misleading due to dilution with cover soil which likely varies from sample to sample. Interestingly, the sample with the lowest C:L had a moisture content of only 17.8%, which would likely have limited the extent of decomposition.

Samples were analyzed from a Chicago area landfill that also accepted some liquids and sludges. The moisture content of the samples ranged from 19.5 to 55.2%, while the pH of all samples except one was in a range that is favorable for methane production. No trends with depth were apparent and the refuse age was not reported. The lowest cellulose concentration and C:L measured were 6.6% and 0.55, respectively.

A New York area landfill was sampled. However, since the refuse samples were only 2 to 4 years old, these samples cannot be expected to represent well decomposed refuse. Interestingly, samples of fresh refuse obtained from the landfill working face contained a cellulose concentration and C:L of 63.4% and 4.04, respectively. The lowest C:L of the excavated samples was 0.87. This landfill was somewhat unique in that the refuse was not well compacted and no cover was applied. As such, moisture infiltration was higher than is otherwise typical and some aerobic biodegradation may have occurred. Moisture contents ranged from 28.2 to 49.1%.

Finally, relatively old samples were collected from a Wisconsin landfill. The results are summarized in Table 3.2. Note that the C:L ratios of these samples are lower than the values associated with other samples.

1 abit 5.2	a Summary Of K	ciuse sumples i			
	Estimated	Maintana	Celledere	T ::-	
Date of Refuse Placement	Refuse Age ^a (yr)	Moisture Content (%)	Cellulose (%)	Lignin (%)	C:L
1948	32	39.0	10.0	48.7	0.21
1954	26	33.4	6.1	38.0	0.16
1957	23	24.4	3.1	12.8	0.24

 Table 3.2
 Summary of Refuse Samples from a Wisconsin Landfill

^aThis assumes that samples were excavated in 1980.

In conclusion, the authors noted that the C:L was highest for the Los Angeles area landfill which is consistent with that area receiving the lowest amount of precipitation. Based on the entire study, the C:L of fresh refuse was about 4, the C:L of partially decomposed refuse was 0.9 to 1.2 and that of well decomposed refuse was about 0.2. In comparing these values to other studies, it should be noted that all analyses were conducted using the gravitimetric technique. Thus, the reported C:L may be artificially high.

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Fawcett and Ham 1986

The Mountain View Controlled Landfill Project was a field-scale demonstration designed to evaluate the effects of leachate recirculation and the addition of sludge and buffer on methane production. Test cells were approximately 100 ft² and 50 ft deep and designed to simulate actual landfill conditions. Solids were excavated from each of the six test cells 4.25 years after refuse burial; the results are summarized in Table 3.3. The measured C:L ratios are below typical values for fresh refuse summarized above. However, they are well above values for well decomposed refuse and suggest that significant additional biodegradation could be expected in each test cell. Interestingly, after 4.25 years, the control cell had a C:L that was lower than that of the cell that received water but not buffer addition. The cell that contained buffer and leachate recirculation had the lowest C:L. A full comparison of the effects of the different treatments is beyond the scope of this report and requires analysis of the chemical composition of the solids as well as moisture content and gas production data as there was evidence of water infiltration and gas production that were not measured.

Treatment	C:L
Sludge, buffer and water addition, leachate recirculation	1.2
Sludge and buffer addition	1.8
Water and buffer addition	2.4
No additions	1.9
Refuse from adjacent landfill	1.7

Table 3.3 Cellulose to Lignin Ratios for Samples Excavated from the Mountain View Controlled Landfill Project (Fawcett and Ham 1986)

Ham, Norman, and Fritschel 1993

Ham, Norman, and Fritschel (1993) characterized samples from the Fresh Kills Landfill in New York. This landfill had been receiving waste since 1948 and is the largest landfill in the U.S. if not the world, covering 1200 ha (3000 acres). Cellulose analyses were conducted using the HPLC technique described above. The refuse age ranged from 1 to 24 years, though it was not possible to date 5 of the 31 samples. The cellulose concentration ranged from 1.8 to 39.3% and the C:L ranged from 0.18 to 3.0. A regression analysis of the C:L with refuse age had a coefficient of determination (r^2) of 0.41; the data used to derive this value are reproduced in Figure 2.1¹. There is significant variability in the C:L for refuse that was less than 10 years old, while the C:L is less than 1.0 for 9 of the 10 samples that were more than 15 years old. The lowest C:Ls were 0.18 to 0.3, which is consistent with values reported in Table 3.2 for well

¹ The initial article by Ham, Norman, and Fritschel reported a coefficient of determination of 0.64 while an r^2 of 0.49 was reported in a summary article on the Fresh Kills Landfill study by Suflita et al. (1992). When the original data were analyzed by this author, an r^2 of 0.41 was determined as illustrated in Figure 1.1. One explanation for the discrepancy is that Ham, Norman, and Fritschel reported r and not r^2 as $(0.64)^2$ is 0.41. In Suflita et al. (1992), the year of refuse burial rather than refuse age was used and this may have influenced the correlation.

decomposed refuse, though slightly higher than values reported by Wang, Byrd, and Barlaz (1994) as summarized below.

The pH of 28 of the 31 samples was in a range that was favorable for refuse methanogenesis and the refuse moisture content was at least 30% for all but one sample. Thus, it appears that environmental conditions in the samples were generally suitable for methanogenesis. This contrasts somewhat with the results of tests to measure gas production in the refuse. The total gas production rate was about 1.5 L CH_4 /kg-yr, which is relatively low and suggests that either samples were already decomposed or that environmental conditions in the refuse were not favorable for methanogenesis. The author's analysis of the gas production data suggested that gas production rates could be related to refuse pH, temperature, and moisture content. Thus, low gas production can be attributed to unfavorable environmental conditions in the selected refuse samples and cannot be used to address the ultimate level of cellulose decomposition that can be expected.

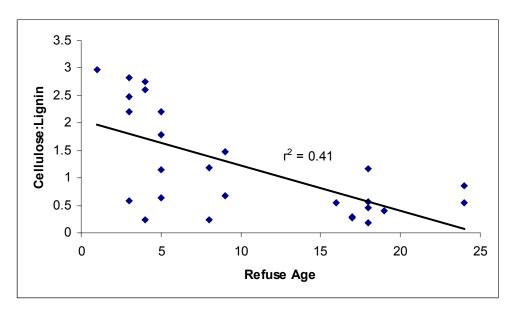


Figure 3.1 Cellulose to Lignin Ratio for Samples Excavated from the Fresh Kills Landfill (Ham, Norman, and Frtischel 1993)

Wang, Byrd, and Barlaz 1994

Wang, Byrd, and Barlaz (1994) reported on samples excavated from the North Waterfront Park Landfill in Berkeley, California; the results are summarized in Table 3.4. The sample age ranged from 7 to 21 years though samples were not dated during excavation. In addition to cellulose, the hemicellulose concentration was measured and the ratio of cellulose + hemicellulose to lignin (CH:L) was reported. This ratio ranged from 0.016 to 0.21 which is slightly lower than the range reported by Bookter and Ham (1982) for the Wisconsin samples (Table 3.2) and by Ham, Norman, and Fritschel (1993) for samples from the Fresh Kills Landfill. Note that the values reported by Wang, Byrd, and Barlaz (1994) include hemicellulose which increases the CH:L relative to the C:L values reported by Ham. The C:L ratio for the data from Wang, Byrd, and Barlaz (1994) are also reported in Table 3.4 and are substantially lower, ranging from 0.011 to 0.165. As presented in Table 3.4, cellulose concentrations ranged from 0.9 to 11.7% and lignin concentrations ranged from 70.9 to 85.6%. The high lignin concentrations serve to illustrate the degree to which most of the cellulose and hemicellulose has been consumed, resulting in the enrichment of the remaining solids in lignin. From Table 1.3, the lignin concentration in fresh refuse ranged from 11 to 33.5%. The high lignin concentrations suggest that there was not significant dilution with cover soil though no specific information on this issue was available.

Biochemical methane potential (BMP)² analyses were also conducted on these samples and the results showed that for cellulose concentrations below about 10%, there was not a good relationship between cellulose plus hemicellulose and the BMP. While the BMP is a measure of the anaerobically degradable carbon that is bioavailable, the results of this study show that at low levels of cellulose and hemicellulose, this relationship is less precise. Thus, the measured cellulose and hemicellulose concentrations in well decomposed samples did not represent bioavailable carbohydrates. In addition, the ratio of the measured BMP to the theoretical BMP is also presented in and showed considerable variability (0 to 0.53). The theoretical BMP is the BMP calculated based on the stoichiometric conversion of cellulose and hemicellulose to methane. The variability in this ratio and the absence of a good correlation between cellulose plus hemicellulose and BMP show the difficulty in trying to identify with any certainty an ultimate limit of cellulose decomposition for mixed refuse.

In summary, the samples characterized in Table 3.4 represent the most decomposed samples reported to date based on their C:L and BMP data. With the exception of sample 5, which contained the highest BMP and cellulose plus hemicellulose concentrations, the other samples likely represent close to the maximum extent of biodegradation that will occur in a landfill.

			· •	•			
Sample	Cellulose (%)	Hemicellulose (%)	Lignin (%)	CH:L ^a	C:L ^a	BMP (mL CH ₄ / dry gm)	$\frac{BMP_{m}^{b}}{BMP_{th}}$
1	0.9	0.5	85.6	0.016	0.011	1.0	0.17
2	4.5	1.5	73.2	0.082	0.061	9.1	0.37
3	3.8	1.9	76.9	0.074	0.049	3.2	0.14
4	1.9	0.8	77.2	0.035	0.025	2.5	0.23
5	11.7	2.5	70.9	0.2	0.165	30.7	0.53
6	5.4	1.8	75.3	0.096	0.072	9.1	0.3
7	1.5	0.6	84.5	0.025	0.018	1.0	0.12
8	1.0	0.4	85.2	0.016	0.012	0	0

Table 3.4 Chemical Composition and BMP of Samples Excavated from North Waterfront
Park Landfill (Wang, Byrd, and Barlaz 1994)

(Continued on next page. See notes at end of table.)

² The biochemical methane potential (BMP) assay is designed to measure the quantity of anaerobically degradable carbon present in a sample. A sample of refuse that has been dried and ground to pass a 1-mm screen is added to a batch reactor, typically a 160 mL serum bottle, along with growth medium and an inoculum of anaerobic bacteria. The volume of methane that can be attributed to the refuse is measured after a 60-day incubation.

Sample	Cellulose (%)	Hemicellulose (%)	Lignin (%)	CH:L ^ª	C:L ^a	BMP (mL CH ₄ / dry gm)	$\frac{BMP_m}{BMP_{th}}^b$
9	5.7	2.5	72.1	0.113	0.079	0	0
10	4.6	1.7	68.2	0.092	0.067	1.6	0.06
11	1.0	0.5	72.0	0.21	0.014	not measured	

 Table 3.4
 Continued

^a The ratio of cellulose plus hemicellulose to lignin (CH:L) or cellulose to lignin (C:L).

^b Measured BMP divided by theoretical BMP as calculated from the stoichiometric conversion of the measured cellulose and hemicellulose concentrations.

Stinson and Ham 1995; Cummings and Stewart 1994

The objective of these studies was to evaluate the significance of lignin in the decomposition of newsprint. A series of samples was prepared in which newsprint was partially to nearly completely delignified by an acid-chlorite treatment. Biodegradability was then assessed by the use of BMP assays. Results showed that delignification of newsprint significantly enhanced its anaerobic biodegradability in BMP tests and that after delignification, the cellulose in newsprint degrades at the same rate as the cellulose in filter paper which contains no lignin. Additional work showed that the printing ink used on the newsprint did not inhibit newsprint conversion to methane.

Cummings and Stewart (1994) evaluated newsprint biodegradation by five cellulolytic bacteria, all isolated from a landfill in the UK. The authors stated that solubilization of the newspaper was poor and that the poor degradation was attributed to the high lignin concentration in the newsprint. The composition of the newsprint tested was 52.8% cellulose, 19.3% hemicellulose, and 24% lignin. Although the actual fraction of the newsprint degraded was not stated, the value would not be relevant to a landfill since the work was done with pure cultures in liquid media. The study also showed that while the presence of ink was not toxic, the ink covered cellulose fiber and reduced its bioavailability.

Eleazer et al. 1997

Eleazer et al. (1997) measured the biodegradability of the major components of MSW in laboratory-scale (2-L) reactors that were operated to maximize anaerobic decomposition. Methane yields and the extent of cellulose and hemicellulose decomposition are summarized in Table 3.5. Two parameters were developed to evaluate the decomposition of each waste component. The methane generation efficiency is the ratio of the measured methane to the methane that would be produced if all of the cellulose and hemicellulose were converted to methane. MC, MH, and ML represent the mass of cellulose, hemicellulose and lignin, respectively, remaining after decomposition, divided by the mass present initially.

Considering all of the waste components, the trend of more extensive cellulose biodegradation (MC decreasing) in the less lignified substrates (CH:L increasing) is weak (Figure 3.1; $r^2 = 0.28$). However, this trend is more definite among the four paper components ($r^2 = 0.76$).

There is not a linear relationship between CH:L and the methane generation efficiency (plot not shown; $r^2 = 0.02$). The results for grass suggest that the lignin concentration does not always reflect the degree to which lignin inhibits cellulose bioavailability. Apparently, the lignin in grass is not as restrictive to microorganisms as the lignin in other components such as branches. This result is consistent with a report by Akin et al. (1995) who stated that "The chemistry of grass lignocellulose varies considerably from that of wood."

Finally, the results for residential MSW measured in this study are compared to earlier data reported by Barlaz, Ham, and Schaefer (1989). Despite wide variation in the initial chemical composition of the two samples, and wide variation in CH:L, the extent of cellulose and hemicellulose decomposition between the two samples was similar.

		Initia	l Composit	ion					
Reactor	Yield mL CH4/	Cellulose	Hemi- cellulose	Lignin					Methane Generation
Series	dry gm ^b	(%)	(%)	(%)	CH:L	MC ^c	MH^{c}	ML ^c	Efficiency ^d
Seed	25.5	23.4	4.7	22.5	1.25	0.18	0.36	0.83	21.8
sd	5.7					0.02	0.03	0.07	
Seed-2	5.8	18.3	3.7	22.1	1.00	0.34	0.69	0.85	6.3
sd	0.6					0.01	0.11	0.01	
Grass	144.4	26.5	10.2	28.4	1.29	0.19	0.42	0.78	94.3
sd	15.5					0.01	0.06	0.07	
Grass-2	127.6	25.6	14.8	21.6	1.87	nm	nm	nm	75.5
sd	21.8								
Leaves	30.6 ^e	15.3	10.5	43.8	0.59	0.43	0.68	0.90	28.3 ^e
sd	8.6					0.05	0.10	0.10	
Branches	62.6 ^e	35.4	18.4	32.6	1.65	0.52	0.59	0.93	27.8 ^e
sd	13.3					0.05	0.02	0.08	
Food	300.7	55.4	7.2	11.4	5.49	0.24	0.58	0.80	84.1
sd	10.6					0.02	0.04	0.07	

Table 3.5 Methane Yield and Initial and Final Solids Composition Data (Eleazer et al. 1997)^a

(Continued on next page. See notes at end of table.)

		Initial	Composit	ion	_				
Reactor		Cellulose		•					Methane Generation
Series	dry gm ^b	(%)	(%)	(%)	CH:L	MC ^c	MH ^c	ML ^c	Efficiency
Coated Paper	84.4	42.3	9.4	15	3.45	0.54	0.58	1.03	39.2
sd	8.1					0.01	0.06	0.03	
Old Newsprint	74.3	48.5	9	23.9	2.41	0.73	0.46	0.99	31.1
sd	6.8					0.05	0.06	0.06	
Old Corrugated Containers	152.3	57.3	9.9	20.8	3.23	0.36	0.38	0.93	54.4
sd	6.7					0.01	0.01	0.00	
Office Paper	217.3	87.4	8.4	2.3	41.65	0.02	0.09	0.95	54.6
sd	15.0					0	0.01	0.18	
MSW	92.0	28.8	9.0	23.1	1.64	0.25	0.22	0.95	58.4
sd	4.1					0.03	0.05	0.02	
$\mathrm{MSW}^{\mathrm{f}}$	86.9	51.2	11.9	15.2	4.15	0.29	0.23	0.92	36.7

Table 3.5 Continued

^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. Data originally published in Eleazer et al. (1997) except as noted.

^b A seed of decomposed refuse excavated from a landfill was used to initiate methane production form each component. All data are corrected for the methane yield attributable to the seed.

^c The ratio of the mass of cellulose (MC), hemicellulose (MH) or lignin (ML) remaining in a reactor divided by the initial mass.

^d The methane generation efficiency is the measured methane yield divided by the yield calculated assuming conversion of 100% of the cellulose and hemicellulose (and protein in the case of food waste) to methane and carbon dioxide. ^e Data exclude L2 and B4.

^f Data for residential MSW as reported by Barlaz, Ham, and Schaefer (1989) and Barlaz, Schaefer, and Ham (1989). NM not measured

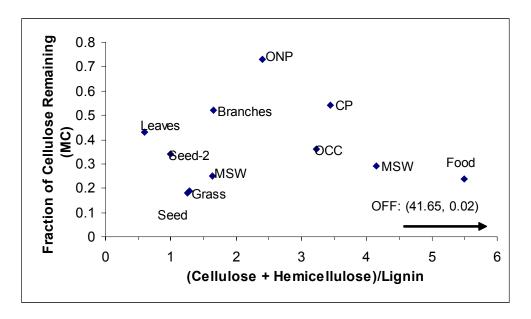


Figure 3.2 Relationship between Extent of Cellulose Decomposition and Cellulose Plus Hemicellulose to Lignin Ratio for Individual Components of MSW

Micales and Skog 1997

Micales and Skog (1997) reviewed work done on refuse decomposition and methane production as they apply to wood and paper waste. This paper is thorough in its review of pertinent literature through its date of publication, and includes some difficult to obtain conference proceedings and final reports. The paper contains several statements on the rate of refuse decomposition, methane emissions, and carbon storage that warrant discussion. Data on refuse decomposition and carbon storage are discussed here and comments on methane production decay rates are discussed in the following section.

Several statements allude to the fact that some readable newsprint can be identified in excavated refuse that is as old as 20 to 30 years or more. Statements such as these are not inconsistent with reports on the chemical composition of excavated refuse. The presence of readable newsprint or food waste that can be recognized does not represent a quantitative measure of decomposition as does the C:L or CH:L. The presence of such materials in old refuse may represent outliers or pockets of undecomposed material but does not represent average conditions in the landfill.

Micales and Skog calculate the total fraction of the carbon released from refuse based on methane yield data reported by Doorn and Barlaz (1995). The data reported by Doorn and Barlaz (1995) are a preliminary version of the data published by Eleazer et al. (1997). While their calculation is straightforward, their estimates of carbon storage were developed before a report by Barlaz (1998) in which C sequestration values were measured directly. The reported values are presented for comparison in Table 3.6. The difference is most significant for office paper, where the measured values suggest significantly less carbon storage than the values calculated from methane yield data. The reason for the large difference is that the mass balances for office paper gave only a 55% methane recovery (Eleazer et al. 1997). Thus, the methane yield is likely higher than what was measured. As such, the carbon storage factor presented in Barlaz (1998) is

a better value. The value for coated paper is almost certainly artificially high as it suggests essentially no methane production when in fact some methane production was measured (Table 3.5). Sources of error include a) errors in the methane mass balance (in 1997, Eleazer et al. reported a methane potential recovery of 84%0, and b) the possibility that some inorganic carbon in the coated paper was measured as organic carbon.

Finally, Micales and Skog state that Doorn and Barlaz (1995) considered wood as having a methane potential of zero. In all subsequent work, Barlaz has assumed the methane potential of wood to be the same as the methane potential of branches. The validity of this assumption has yet to be tested.

		· · · · · · · · · · · · · · · · · · ·	- · ·	· · ·
	Total fraction C Released	Total Fraction C Stored	C Sequestered	Total Fraction C Stored
	(kg C released/ kg C)	(kg C stored/ kg C) ^a	(kg C stored/ dry kg material)	(kg C stored/ kg C) ^b
	Micales and	Skog 1997	Barla	z 1998
Wood (Micales and Skog 1997) or Branches (Barlaz 1998)	0-0.032	0.968 – 1	0.38	0.77
Newspaper	0.157	0.843	0.42	0.85
Corrugated Containers	0.315	0.685	0.26	0.55
Office Paper	0.382	0.618	0.05	0.125
Coated Paper	0.175	0.825	0.34	0.99

Table 3.6 (Carbon Storage	Values Presented by	y Micales and Sko	g (1997) and Barlaz ((1998)

^a These data are calculated as one minus the fraction of carbon released.

^b These data were converted by using the carbon content of each material as follows: wood (branches in Barlaz 1988) – 49.4% of dry wt, newspaper – 49.2%, corrugated containers – 46.9%, office paper – 40.3%, coated paper – 34.3%.

Baldwin, Stinson, and Ham 1998

The objective of this study was to follow the decomposition of specific waste components in landfills over six years. The study was designed to eliminate some of the disadvantages of previous research on refuse decomposition as the starting materials and the sample age were known precisely. Materials were selected for testing based on their expected degradability. Filter paper, pasta, and lima beans were expected to be very degradable because of their high cellulose and low lignin content. Broccoli and peanuts were expected to degrade at intermediate rates as they contain both cellulose and lignin, while newspaper and sunflower seeds were chosen to represent highly lignified materials that were be expected to degrade the slowest. In addition, four types of diapers that contained various amounts of cellulose, lignin, and an absorbent gel material were tested. Materials were buried about 3 m below the surface in bags

designed to facilitate moisture infiltration and sample retrieval. Unfortunately, it was not possible to locate all samples at each of the three landfills after two or six years.

In the case of filter paper, decomposition was most extensive in the Florida landfill that received the most moisture. The authors noted that more filter paper was recovered in the Pennsylvania and Wisconsin landfills but the extent of filter paper decomposition could not be quantified. The authors concluded that filter paper is decomposable. Data for the loss of both total mass and holocellulose (cellulose plus hemicellulose) for newsprint are summarized in Table 3.7. The authors noted some problem with debris accumulation on the newspaper which explains the mass increases in some samples. Nonetheless, the holocellulose loss accounted for essentially all of the mass loss in the newspaper.

	% Change in Dry Mass Relative to Initial Weight Buried					
		Newspaper				
	1 yr ^a	2 yr ^a	6 yr ^a			
Wisconsin-sand cover	2.5 ^b	11.6 ^b	-8.5			
Wisconsin – clay cover	8.5 ^b	11.0 ^b	-5.6			
Florida	-7.2 ^b	-17.4				
Pennsylvania	14.0 ^b					

Table 3.7 Decomposition of Buried Samples of Newsprint(Baldwin, Stinson, and Ham 1998)

% Change in Cellulose Plus Hemicellulose Relative to Initial Weight Buried

	Newspaper				
	1 yr ^a	2 yr ^a	6 yr ^a		
Wisconsin-sand cover	-0.5 ^b	13.8 ^b	-13.1 ^b		
Wisconsin – clay cover	12.0	17.1	-30.8		
Florida	-17.1 ^b	-13.8			
Pennsylvania	35.9				

^a A negative number represents a loss relative to the starting material.

^b This change is not significantly different, at the 95% confidence level, from the initial mass.

In general, the greatest mass losses were reported for samples with the highest moisture contents, and samples from the Florida landfill showed the largest and most rapid increase in moisture content. Samples from the Florida landfill were also the warmest, which would contribute to more rapid decomposition rates. The results from the Florida landfill also demonstrated the inhibitory effects of lignin on holocellulose decomposition over the six-year study period.

This study further emphasizes the importance of moisture content as a critical parameter for the enhancement of refuse decomposition and shows that MSW components that are more heavily lignified degrade more slowly. This study also documents the degradability of cellulose and hemicellulose in landfills, the importance of temperature and moisture content, and the limiting effect of lignin.

Chugh et al. 1999

Chugh et al. (1999) performed research to demonstrate the biodegradation of shredded MSW in a two-stage anaerobic digester operated with leachate recirculation. Volatile solids losses of 55 to 69% were reported for MSW subjected to two months of anaerobic digestion. The chemical composition of the volatile solids was not reported though it would likely have been dominated by cellulose, hemicellulose, and lignin as illustrated by the chemical composition data presented in Table 1.3. On average, measured methane yields in the reactor system were 75% of the ultimate methane yields as measured by BMP tests. This is consistent with the observation that the BMP of the residual MSW after digestion was an average of 27.5% of the BMP of the feedstock. Thus, BMP data suggest that about 70 to 75% of the degradable solids can be degraded in a reactor system. This is consistent with work by Barlaz, Ham, and Schaefer (1989) and Barlaz, Schaefer, and Ham (1989) in which 71 and 77% of the cellulose and hemicellulose, respectively, added to a reactor system in the form of shredded MSW were degraded.

Rodriguez et al. 2001

Rodriguez et al. (2001) used an enzymatic technique to evaluate the bioavailability of cellulose in refuse excavated from landfills. Sugar release from refuse samples was measured in the presence of a mixture of three hydrolytic complexes isolated from 3 well characterized cellulose degrading organisms (*Trichoderma reesei, Humicola insolens,* and *Aspergillus aculeatus*); cellulose bioavailability was expressed as a fraction of the cellulose in a sample that was hydrolyzed. The results showed a decreasing trend in the fraction of the refuse that was hydrolyzed in samples of increasing depth though no information was provided on the sample age. There was no correspondence between C:L and the fraction hydrolyzed and there was only a weak relationship between cellulose content and the fraction hydrolyzed. No cellulose was hydrolyzed in samples containing less than 5% cellulose. These results are not inconsistent with reports from other research groups on relationships between decomposition and both depth and degree of lignification.

The approach presented here represents an alternate way to assess cellulose bioavailability which corresponds to methane potential. One advantage is that it is much faster than the BMP test that requires 30 to 60 days to complete. However, it is not clear that the hydrolytic enzyme complex used for this test is relevant to landfills dominated by anaerobic conditions. Nonetheless, this technique is useful for a relative assessment of biodegradability within a specific set of samples.

Harries, Cross, and Smith (2001)

A test lysimeter was filled with MSW and the cellulose content was monitored over time. The initial cellulose concentration was only 20% and after 1500 days, by which time methane production was nearly complete; the cellulose concentration had decreased to about 10%.

However, the total mass of cellulose converted was not reported. In addition, the older gravitimetric cellulose analytical protocol was used.

Zhao et al. (2001)

The researchers attempted to measure waste decomposition in a 0.3 ha landfill test cell that was constructed in China. However, there was not a consistent decrease in the cellulose content of the waste over the monitoring period, which ranged from 4 to 7 years, and methane production data were not reported. The technique used for cellulose analysis did not appear to insure the separation of all non-cellulosic organics from the refuse, which may explain the absence of a significant repeatable decrease in cellulose content.

Mehta et al. (2002)

A side by side comparison of refuse decomposition in the presence and absence of leachate recirculation was conducted in 0.25 acre test cells, each containing about 7730 mT of waste. Solids were excavated 3.4 years after burial for chemical analyses and BMP measurements. Leachate was recirculated for about 2 years but was not recirculated for about a year prior to solids sampling.

The average CH:L ratios were 1.09 and 1.44 in the enhanced and control cells, respectively, and these ratios were significantly different. While no measure of the CH:L was available for the refuse initially buried, the measured ratios are well below the values reported in Table 1.3 for fresh refuse. However, these ratios are also well above values reported by Wang, Byrd, and Barlaz (1994), Bookter and Ham (1982), and Ham, Norman, and Fritschel (1993) for decomposed refuse.

These results are most useful as a comparison between a control and leachate recirculation cell and clearly demonstrated enhanced decomposition in the cell operated with leachate recirculation. However, the results should not be used as an indicator of the ultimate biodegrad-ability of refuse because the CH:L and BMPs are not representative of well decomposed refuse. The absence of complete solids decomposition can be explained by the fact that leachate was only recirculated for two years and the method of leachate infiltration, small shallow infiltration trenches, was sub-optimal. Moisture content data showed that the leachate recirculation cell was wetter than the control cell. However, moisture contents ranged from 28 to 55%, indicating that some of the refuse in the leachate recirculation cell was likely below its optimal moisture content.

Gardner et al. (2003)

Samples were excavated from two landfills in Australia to study the decomposition of wood products (Gardner et al. 2003). The first landfill (Site A) was operated in a conventional manner and samples were excavated 19 years after burial. The second landfill (Site B) was operated with leachate recirculation and samples were excavated 29 years after burial. Samples were excavated with a bucket auger and wood products were removed from the samples at 0.5 m depth intervals. The types of materials analyzed included rough sawn and dressed boards, moldings, furniture components, and broom and tool handles. Where possible, samples were identified as hardwoods or softwoods. Cellulose and hemicellulose were analyzed by using detergent fiber techniques. While these techniques have limitations for bulk refuse, they are suitable for wood products.

The average moisture content of the samples recovered from Sites A and B were 44.2 and 58.8%, respectively, which is consistent with the manner in which these sites were operated. Selected

solids analysis data from the referenced paper are summarized in Table 3.8. The authors indicate that the only significant change was a decrease in the hemicellulose content at Site A. According to the authors, there was a smaller and insignificant decrease in hemicellulose content at Site B. The CH:L ratios suggest a decrease in Site A hardwoods only.

The greater decrease in CH:L from Site A is surprising in that samples excavated from Site A were younger and drier while Site B was operated to enhance decomposition. pH data were not reported, so it is not possible to assess whether conditions at Site B were suitable for methanogenesis. Since methane was recovered in commercial quantities at Site A, it is likely that conditions were suitable for methanogenesis in at least parts if not all of the landfill.

These data show that wood products decay very slowly in landfills and the decomposition reported is significantly less than that of branches reported in Eleazer et al. (1997). However, there are some limitations to this study that must be recognized. First, the results could have been validated by analyzing the solids content of refuse at the same depths as the wood samples. If the authors could demonstrate a decrease in CH:L for the bulk refuse, this would validate the absence of a decrease in CH:L for the wood products. Second, information to indicate that environmental conditions in Site B were suitable for decomposition would have been useful to advance the argument that wood products do not decay to an appreciable extent in landfills. Finally, it is not possible to prove whether some wood was not identified because it was highly decomposed, though this appears unlikely from the study results. This will always be the case with a field study.

The authors reported a total carbon loss of 2.5 and 4.1% for sites B and A, respectively, based on the hemicellulose data. However, it is unclear how these figures were derived. An attempt was made to calculate the carbon loss based on the reported data. Starting with the chemical composition for fresh hardwood, the lignin concentration is 23.4%. Assuming that lignin is recalcitrant in landfills, the lignin can be treated as a conservative compound. Assuming an initial control sample mass of 100 gm, the equivalent sample mass of this 100 gm after decomposition would be 90.7 gms based on the measured hardwood lignin concentration of 25.8% and an equivalent mass of lignin in the initial and decomposed sample. Assuming that 100 gm were decomposed to 90.7 gms, the calculated hemicellulose loss would be 5.81 gm or 2.61 gm C. A loss of 2.85 gm C corresponds to a loss of 5.1% based on the carbon content of fresh hardwood. This procedure also results in a cellulose-carbon loss of 1.92 gm C, an additional 3.75%. This approach fails for softwood because the lignin concentration decreased where it would be expected to increase if other material were decomposed. Thus, while the data suggest little decomposition of wood products, it is not possible to be quantitative. If it is assumed that the hardwood carbon loss was 9.32% and the softwood loss was 0, then the average loss is 4.66%. If the carbon loss is calculated from the carbon data, again assuming that lignin is conservative, then the carbon loss is 11.15% and 10.06%, respectively. Whether this represents ultimate decomposition cannot be determined from the available data. If 10% of the carbon in wood is decomposed, and it is all hemicellulose, then the equivalent methane yield would be 0.094 L CH_4 /gm dry wood. Finally, it would be more appropriate to apply the procedure described here to individual samples rather than to the average data as the range of lignin concentrations in the Site A hardwoods was 13.2 to 40%.

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	Carbon	Cellulose	Hemicellulose	Lignin	CH:L	
		CONTR	OLS			
Hardwood	51.1	53.7	13.7	23.4	2.88	
Softwood	51.13	47.6	12.3	31.0	1.93	
		SITE	A			
Hardwood	50.1	54.4	8.7	25.8	2.44	
Softwood	50.7	51.1	8.7	29.6	2.02	
SITE B						
Hardwood	51.1	59.9	9.4	24.1	2.88	
Softwood	50.9	52.6	11.3	31.0	2.06	

Table 3.8 Summary of Analyses Presented by Gardner et al. (2003)^a

^a Data are expressed as a percentage of dry weight.

3.3 Discussion of Literature Review

There has been a wide range of studies on the degradability of refuse buried in landfills beginning with the work of Ham and Bookter in 1982 and concluding with the work of Gardner et al. in 2003. These studies are generally consistent in demonstrating that MSW decomposes in landfills and that CH:L ratios can go as low as at least 0.2, and in the case of Wang, Byrd, and Barlaz (1994), as low as 0.02. Based on the composition of refuse presented in Tables 1.2 to 1.4, it is apparent that cellulose and hemicellulose decompose in landfills and that the major input of these constituents to landfills is paper.

Cellulose and hemicellulose decomposition are limited by both environmental conditions in landfills and by the presence of lignin. With respect to environmental conditions, the two most critical variables are moisture content and pH (Barlaz, Ham, and Schaefer 1990), and the operation of landfills as bioreactors will provide more suitable environmental conditions for biodegradation.

A number of studies have shown that the presence of lignin inhibits cellulose decomposition. The presence of lignin is important because it is both a physical and chemical barrier to microbial attack (Colberg 1988; Dehority and Johnson 1961; Tong, Smith, and McCarty 1990; Pfeffer and Khan 1976). In the work reviewed in this report, the presence of lignin has been shown to limit the bioavailability of cellulose and hemicellulose in a number of studies (Wang, Byrd, and Barlaz 1994; Stinson and Ham 1995; Eleazer et al. 1997; Baldwin, Stinson, and Ham 1998). Several additional studies have shown the presence of lignin to be the major impediment to the digestion of forage (Allison and Osbourn 1970; Barton and Akin 1977; Dehority and Johnson 1961; Jung and Vogel 1986) and other types of biomass (Tong, Smith, and McCarty 1990). The lignin concentration has been related to both the rate and extent of cellulose plus

hemicellulose degradation. However, the degree to which this relationship can be modeled varies between studies. Dehority and Johnston (1961) reported that cellulose digestibility was not directly correlated to the lignin concentration. In their study, legume cellulose was digested to a lesser extent than the cellulose in four types of grass, although the grasses and legumes had similar lignin concentrations. Similarly, the methanogenic conversion of white fir and wood grass, which have similar lignin concentrations, was 9 and 66%, respectively (Tong, Smith, and McCarty 1990). However, a curvilinear relationship was shown to relate increasing lignin concentrations to a decrease in the digestibility of cellulose in eight species of grass (Jung and Vogel 1986). Similarly, Eleazer et al. (1997) showed that lignin concentration alone is not a good predictor of cellulose bioavailability as evidenced by different effects of lignin associated with grass and paper.

Ideally, it would be possible to calculate the amount of degradable cellulose based on the lowest residual concentration in refuse. To evaluate this possibility, a theoretical final cellulose concentration was calculated from the BMPs of excavated refuse as described by Wang, Byrd, and Barlaz (1994). The range of final calculated cellulose concentrations was 0.6 to 5.7% (avg = 2.7, sd = 1.7). Thus, no definitive statements regarding a lower level of cellulose degradation were possible.

While most of the research summarized in this section focused on mixed refuse, the studies presented by Stinson and Ham (1995), Cummings and Stewart (1994), Eleazer et al. (1997), Baldwin, Stinson, and Ham (1998) and Gardner et al. (2003) presented data specific to paper and paperboard, and wood products. Micales and Skog (1997) also mention wood. However, their numbers were largely derived from preliminary reports on the work of Eleazer et al. (1997). Stinson and Ham (1995) demonstrated the significance of lignin as an inhibitor of newsprint decomposition and, along with Cummings and Stewart (1994), verified that the inks used on paper are not toxic to cellulolytic bacteria. Eleazer et al. (1997) were the first to publish methane yields and to document the extent of cellulose and hemicellulose decomposition for specific types of paper in a reactor system. Their results also suggested that lignin limited the bioavailability of cellulose and hemicellulose (Figure 3.2). This importance of lignin was also verified by Baldwin, Stinson, and Ham (1998) who measured the decomposition of newsprint samples that were deliberately buried in landfills in Wisconsin, Florida, and Pennsylvania. The data from Gardner et al. (2003) are unique in that they look at changes in the chemical composition of specific materials after long-term burial, though additional validation data are needed to support quantitative results.

4.0 METHANE PRODUCTION DECAY RATES

The objective of this section is to review reports that present information on the rate of methane production which is, in essence, the rate of refuse decomposition. Methane production from landfills is typically modeled using some form of equation 3:

$$G = W_a L_o k e^{-kt}$$
(3)

where G = methane production rate (ft³/yr), W_a = annual waste acceptance rate (ton/yr), L_o = ultimate methane yield (ft³ CH₄/ton), k = decay rate (yr⁻¹), and t = years since the landfill started receiving waste. This model is the same as that used by USEPA in its LandGem model (USEPA 1998b; IPCC 1997). Alternate forms of this equation are used when only the total waste in place is known or when the deposition rate is constant. In other cases, the waste acceptance rate is

replaced with terms to represent the degradable organic carbon (DOC). However, all forms of equation 3 require the use of a decay rate. Reports of alternate decay rates are reviewed in this section.

Prior to the published decay rate information review, some further clarification of equation 3 is appropriate. In AP-42 (USEPA 1998a), EPA reports the integrated form of equation 3 which is given in equation 4:

$$G = W L_o \left(e^{-kc} - e^{-kt} \right) \tag{4}$$

In equation 4, G represents the methane production rate from all years' refuse (ft^3/yr) and will be referred to as G*, W = the average annual waste acceptance rate (ton/yr), and c is the time since waste burial ceased. L_o, t, and k are as defined for equation 3. Equation 3 is used in this report because it is more flexible and readily accommodates annual variation in the waste disposal rate. The results of a comparison of methane production rates for equations 3 and 4 are presented in Table 13. For use of equation 3, all refuse was assumed to be disposed of in the middle of the year, whereas all refuse was disposed of at the end of each year in equation 4. Thus, the comparison in Table 4.1 is based on the methane production rate calculated after 9.5 years in equation 3 and 10 years in equation 4. As illustrated in Table 4.1, the difference between equations 3 and 4 is not significant.

k	Equation 3	Equation 4	% Error ^b
0.05	127306794	127320056	0.010
0.1	164127351	164195746	0.042
0.15	195225810	195408886	0.094
0.2	188082581	188396209	0.167
0.25	171030924	171476666	0.261
0.3	150280101	150844286	0.375
0.35	15818846	15899712	0.511
0.4	109417616	110148527	0.668
0.45	91737935	92513935	0.846
0.5	76360491	77158402	1.045

 Table 4.1 Difference in Predicted Methane Production Rate Between Equations 3 and 4^a

^a The methane production rates are reported in ft^3/yr for a landfill that receives 300,000 tons/yr for 6 years with an L₀ of 1 ft^3/lb . The reported rates are for year 10.

^b Calculated as 100* (rate from eqn. 4 – rate from eqn. 3)/rate from eqn. 3. The error does not vary as a function of time.

The application of equation 3 for a series of decay rates is illustrated in Figures 4.1 and 4.2. In Figure 4.1, methane production rates are illustrated for the decomposition of 286,000 tons of refuse buried at the beginning of year 1. In Figure 4.2, methane production rates are illustrated for the decomposition of 286,000 tons of refuse buried at the beginning of years 1 through 20. Each figure is based on refuse with an ultimate yield (L_0) of 1.5 ft³ CH₄/lb. Data are reported as landfill gas flow rates which include CH₄ plus CO₂. Landfill gas was assumed to consist of a 50/50 mix of CH₄ and CO₂. As the decay rate increases, the rate of methane production increases and the period of time over which significant methane production can be expected decreases. As a point of reference, the Clean Air Act Amendments (CAA) require a default decay rate of 0.05 yr^{-1} for mixed refuse while AP-42 suggests decay rates of 0.04 and 0.02 for areas receiving greater than and less than 25 inches of rain per year, respectively. The AP-42 documentation states that "[allthough the recommended k and L_0 are based upon the best fit to 21 different landfills, the predicted methane emissions ranged from 38 to 492% of actual, and had a relative standard deviation of 0.85 inches. The peak methane production rate occurs at the time of refuse burial because the model presented in equation 3 (or equation 4) does not include a lag time. It is a simple matter to add a lag time to these equations which would in effect shift the curves presented to the right. The EPA defaults were based on their own internal analysis of landfill gas collection data which are available in the public docket for the rule (Susan Thorneloe, personal communication, 6/7/03).

In USEPA's model, equation 3 is typically used to represent all refuse as one material. However, equation 3 can be used to represent multiple fractions of the refuse and the results summed. For example, the 286,000 tons per year buried could be divided into separate masses that decompose at different rates. This is typically done by Emcon, in their methane generation model, as well as in a model developed in Holland and discussed below (Coops et al. 1995).

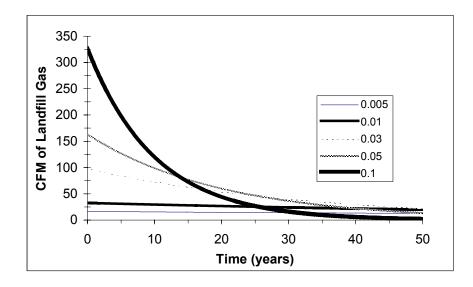


Figure 4.1 The Effect of Decay Rate on the Methane Production Rate Based on the Burial of 286,000 Tons of Refuse at Time Zero (The curve is based on equation 3 and an L_o of 1.5 ft³ CH₄/ton.)

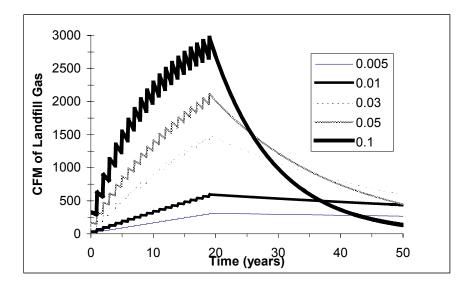


Figure 4.2 The Effect of Decay Rate on the Methane Production Rate Based on the Burial of 286,000 Tons of Refuse Annually for 20 Years (The curve is based on equation 3 and an L_o of 1.5 ft³ CH₄/ton. The jagged nature of the curve is because refuse is added to the landfill annually.)

Doorn and Barlaz (1995)

Doorn and Barlaz (1995) published an estimate of global methane emissions based on a series of surveys conducted by USEPA to obtain methane generation data from U.S. landfills coupled with country-specific estimates of waste generation and waste management practices. The report contains several statements that have implications for methane decay rates. "Gas flow rates correlated most strongly with refuse age for 10 to 20 year old refuse. Although these rates were not conclusive, they suggest that the generation time for gas production is 20 to 30 years with an average of 25 years ..." The meaning of the term "generation time" is not clear given the asymptotic nature of methane production rate curves (Figures 4.1 and 4.2). Given that the data were collected from full-scale landfills with gas collection systems, it is likely that generation time refers to a period of time when there is sufficient gas available for an energy recovery project. However, the volume of gas available will be a function of the mass of refuse buried so this statement cannot be used to infer a decay rate.

Coops et al. (1995)

The objective of this study was to evaluate alternate models for prediction of landfill gas generation. Four models were evaluated, including a zero order model, a first order model, a second order model; and a multi-phase model. The multi-phase model is similar to the first order model except that the waste is divided into three fractions that degrade at different rates (slow, moderate, and rapid). Each model was tested on landfill gas production data from nine landfills; 18 separate annual gas production rates were used.

Data on the mass of waste buried annually were available for each of the 18 data sets as were estimates of the types of wastes received. Waste composition was assumed based on a previous study of waste composition in The Netherlands. The authors recognized the distinction between landfill gas production and recovery and they estimated the landfill gas collection efficiency for each landfill for each data set based on the geometry of the landfill, the type of cover, and the distribution of landfill gas collection wells. The estimated landfill gas recovery efficiencies at the nine landfills that were applied to the 18 data sets were 70, 60, 90, 45, 75, 65 (20% for part of the landfill), 95, 60, 90 (55% for part of the landfill). Thus, the published model is intended to represent "methane formation" as opposed to "methane recovered."

The study showed that the multi-phase model performed slightly better than either the first order or the second order model. The mean relative error for the zero order, first order, multi-phase, and second order models were 44, 22, 18 and 22%, respectively. However, the authors explained that because the multi-phase model had more parameters than the other models, a better fit was to be expected and that the multi-phase model would not necessarily provide better predictions of landfill gas formation. Ultimately, the recommended model is given as equation 5 along with the best fit parameters.

$$\alpha = \xi \sum_{j=1}^{3} 1.87 A C_{0,1} k_{1,i} e^{-k_{1,i}t}$$
(5)

 α = gas production rate (vol./yr)

 ξ = a generation factor which represents the fraction of the organic waste that is degradable (0.58)

1.87 L CH_4/kg = organic carbon degraded (This value can easily be misinterpreted. Based on the stoichiometry of cellulose decomposition, there are 184 L CH_4/kg C degraded. However, this value is appropriate when multiplied by the fraction of organic carbon expressed in kg/wet tonne.)

A = mass of waste in place

C = the fraction of the waste that is organic carbon (kg/tonne)

 $k = decay rate (k_1 = 0.185, k_2 = 0.1, k_3 = 0.03 yr^{-1})$

The authors reported that the best fit was obtained with the values given parenthetically above. Interestingly, their search procedure for the multi-phase model began by constraining k_3 to 0.03 and ξ to 0.58. ξ was found to be 0.58 in the search routine based on the first order model. Equation 5 differs slightly from the conceptual basis for the model presented in equation 3 where methane generation is represented by a simple yield (L_o) without consideration of the fraction of the waste that is degradable. The decay rates for the rapid and moderate fractions are faster than the decay rates given in both AP-42 and the CAA. Average rainfall in Holland between 1989 and 2002 has been 87.4 cm (34.4"), slightly lower than values typical in the eastern and midwestern U.S. For purposes of comparison, the best fit parameters for the first order model, which is most comparable to the model used in AP-42 and the CAA, were $\xi = 0.58$ and k = 0.094 yr⁻¹.

Pingoud, Savolainen, and Seppala (1996)

The objective of this study was to evaluate the greenhouse gas impact of the forest products industry in Finland. The study included waste management in the analysis and also includes some work on paper and wood decomposition in landfills. The papers used the protocols set forth by the International Panel on Climate Change (IPCC) for estimation of gas emissions from landfills and did not contain any primary data. Pingoud, Savolainen, and Seppala recognize that there is considerable uncertainty in estimates of methane emissions from landfills and that one highly uncertain factor is the fraction of the DOC that actually decomposes in landfills.

SCS and Augenstein (1997)

The objective of this study was to evaluate several models for methane production from landfills. This study is analogous to the work on Dutch landfills by Coops et al. (1995). Gas recovery and waste deposition data were obtained from 18 landfills and four models were evaluated including a zero order model, a simple first order model (equation 3), a modified first order model (equation 6) and a multi-phase model. The modified first order model includes term(s) to account for the rate of increase in methane production. The multi-phase model described by SCS and Augenstein is similar to equation 3 except that the waste is divided into two fractions with different decay rates but the same L_0 .

$$G = WL_{o}[(k+s)/s][(1-e^{-s(t-t_{j})})(ke^{-k(t-t_{j})})]$$
(6)

- G = methane generation rate
- W = waste in place
- $L_o =$ ultimate methane yield
- T = time after waste placement
- $T_1 = lag time$
- k = decay constant
- s = first order rise constant

This study focused on the prediction of recoverable methane and did not attempt to correct for varying collection efficiencies across the landfills studied. However, landfills were only selected for study if they were operated to maximize gas recovery efficiency. As such, the L_o values estimated in this work represent recoverable methane. No specific recommendations were given as to which of the four models evaluated was most appropriate. However, all four models resulted in reasonably good fits of the available data, with ultimate yields of 0.85 to 1.25 ft³ CH₄/lb and decay rates of 0.03 to 0.07 yr⁻¹ for the simple and modified first order models. For the first order model, the following combinations of L_0 in ft³ CH₄/lb and k in yr⁻¹ gave the best fits: 1.05, 0.07; 1.1, 0.04; and 1.25, 0.03. In the multi-phase model, decay rates as high as 0.08 were used for the rapidly degradable fraction.

Micales and Skog, 1997

Micales and Skog (1997) reviewed work done on refuse decomposition and methane production as it applies to wood and paper waste. The paper contains several statements on the rate of refuse decomposition, methane emissions, and carbon storage that warrant discussion. With respect to methane production rates and half-lives, it is stated that "[i]f methanogenesis does become established, methane production can last for 8 to 40 years (Augenstein and Pacey 1991), although quantities of methane high enough for commercial production are generally formed for only 5 to 20 years (Suflita et al. 1992)." In the case of Augenstein and Pacey, the comments are based on their experience in the landfill gas industry and on proprietary data. The data undoubtedly originate from landfills of various ages, size, and design operating at varying temperatures and moisture contents. For this reason, although the Augenstein and Pacey, and Suflita studies are important, it is difficult to use the findings in a quantitative analysis.

Micales and Skog refer to half-lives estimated by Augenstein and Pacey that range from 2 to 5 years for wet regions, 5 to 10 years for regions with moderate precipitation, and 10 to 25 years for dry regions. Half lives of 5, 10, 15, 20, and 25 years are equivalent to decay rates of 0.139, 0.069, 0.046, 0.035, and 0.028 yr⁻¹, respectively³. The half-lives presented by Augenstein and Pacey (1991) are based on analysis of landfill gas production data. The precise reference is a memorandum that has been reproduced as Appendix B. The estimates reflect collected methane as opposed to produced methane.

Skog and Nicholson 1998

The objective of this study was to develop estimates of the amount of carbon sequestered in wood and paper products in the U.S. As part of their analysis, the authors evaluated the quantity of carbon sequestered in wood and paper products buried in landfills and commented on the rate and extent of paper and wood decomposition. The paper does not contain primary data on waste decomposition in landfills.

³ The decay rate is calculated as $\ln 2/t$.

IPCC Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories, Chapter 5 – Waste (IPCC 2001)

In general, the IPCC method uses equation 3 to estimate methane production from waste disposal and then adds various correction factors for collection and methane oxidation. The subject document indicates that published data support decay rates for mixed MSW of 0.03 to 0.2 yr^{-1} and in the absence of better information, they recommend a decay rate of 0.05. This recommendation is based on judgment and appears reasonable based on the reports reviewed for this report.

The IPCC method requires the use of degradable organic carbon (DOC) and the fraction of DOC in various waste components. DOC is an operational definition since the fraction of the total organic carbon that is degradable will vary with the ecosystem (e.g., aerobic versus anaerobic). The IPCC provides guidance on the DOC for several different types of waste.

4.1 Discussion of Landfill Gas Model Parameters

Summaries of reported values for L_o and k are presented in Tables 4.2 and 4.3, respectively. These tables include data measured under both laboratory and field conditions and incorporate values for mixed MSW, specific components of MSW (including paper and paperboard), and some data specific for pulp and paper mill wastes. As specified in the table, in many cases the L_o was calculated based on DOC data and the IPCC equation (7) which requires assumed values for a number of parameters that are not of concern when L_o is measured directly.

$$L_o = DOC^*DOC_f^*16/12^*F^*MCF$$

(7)

DOC = fraction of waste that is degradable organic carbon

 DOC_f = fraction of DOC that actually degrades

F = fraction of methane in landfill gas

MCF = factor to reduce methane production based on landfill operation:

- 1.0 for a modern landfill
- 0.4 for a shallow unmanaged landfill (< 5m deep)
- 0.8 for a deeper landfill (>5 m deep)

The L_o values for MSW, both measured and calculated, vary from about 80 to 160 L CH₄/wet kg but many of the values are in a tighter range around 100 L CH₄/wet kg. The value used by USEPA in the NSPS, 272 L CH₄/wet kg, is an outlier and likely reflects a very conservative stance by the agency for a regulatory purpose.

The Swedish authorities use one value for pulp and paper mill waste but no supporting information for this value could be identified. Given the wide variety of pulp and paper mill wastes, the use of one value is suspect. As presented in Table 4.3, there is a reference to a Finnish study in which DOC data were presented. These DOC data were used to calculate L_o values based on the defaults given in Table 4.3 and equation 7. The original references for the Finnish data were published in Finnish and could not be evaluated. However, it is important to differentiate between the DOC data provided in the Finnish report and the L_o data that were calculated. The calculation does not take into account that some waste may not degrade due to elevated pH, for example.

The published decay rates summarized in Table 4.4 are all based on MSW with the exception of a value for wood presented in a Canadian report. Most of the variation in decay rates can be explained by the use of different models. When a multi-phase model is used, two or three decay rates may be specified, while only a single decay rate can be specified in a model that does not differentiate specific fractions of the waste undergoing decomposition.

The refuse decomposition decay rate is a critical model parameter as it influences the rate and duration of methane production. As illustrated in Figures 4.1 and 4.2, variation in the decay rate will lead to changes in the shape of a methane production rate curve. While the literature contains a number of estimates of the decay rate, there is no formal consensus on an appropriate rate that can be supported by rigorous science. The best available work is that of Coops et al. (1995) and SCS and Augenstein (1997), where data from full-scale landfills were used to derive decay rates. As presented above, Coops et al. (1995) reported that a multi-phase model with three decay rates gave the best fit to existing data. Decay rates of 0.185, 0.1, and 0.03 were applied to the rapidly, moderately, and slowly degradable fractions of refuse. However, Coops et al. also recognized that the multi-phase model may not provide better predictions of landfill gas production than a first order model. The values reported by Coops et al. encompass the range of values that were reported by SCS and Augenstein and also the values used by USEPA and the IPCC. The use of a multi-phase model does allow more flexibility to represent different types of waste and actual waste composition on either a site-specific or country-specific basis.

The use of a multi-phase model is justified and appropriate based on current knowledge of the degradability of specific waste components, but data limitations and the lack of familiarity with such models suggest that the simple first order model (equation 3) will remain the most frequently used for the foreseeable future. For this reason, where possible, Table 4.4 also includes the first order decay rates that best fit the data from the studies included in this review.

Table 4.2Summary of Values for Ultimate Yield Reported in the Literature

		animal of anima	DUILING OF THINGS IN COMPANY AND THE WAY WAY IN THE AND THE AND		
Study (Reference)	Method or Equation	Model Defaults	Calculated or measured L _o (L-CH4/dry kg)	Comments	
IPCC (1997)	eqn. 7	DOC ^a : 0.18 – 0.21 DOC _f ^b : 0.5-0.6 F ^c – 0.5 MCF ^d – 1.0	MSW: 105 to 122.5 for DOCs of 0.18 - 0.21	Reported values assume 20% moisture on wet weight basis. Using the average values in Table 3 and assuming that lignin is 40% C, the DOC would be 0.31 kg C/kg dry waste or 0.25 kg C/kg wet waste at 20% moisture. Thus, the default DOC is perhaps low. An alternate approach to the MCF factor would be to assess what fraction of buried waste decomposes under anaerobic conditions.	
UK (AEA Technology 2001)	eqn. 7	DOC – varies with waste type DOC _f – 0.6 F – 0.5 MCF – 1		If DOC varies with waste type, then DOC _f should also vary with waste type because the fraction of the cellulose and hemicellulose that actually degrade vary markedly among waste components (Table 9).	n
Sweden (Swedish Env. Prot. Agency 2001) Finland (Tech. Res. Cntr. for Finland 2001) 2001)	value specified eqn. 7	Not applicable DOC: see note e DOC _f : 0.5 F - 0.5 MCF - 0.7	Volue Pulp and paper mill waste: - 126 (90 kg/ton) 126 (90 kg/ton) DCC: see note e MSW: 114.9 (20%) DOCré: 0.5 Paper and cardboard: 196.5 - 0.5 (5%) - 0.5 Wood and bark: 200 (30%) ACF - 0.7 Wood and bark: 200 (30%) Deinking waste: 93.4 (5%) Forest industry sludge: 210 Paper stindustry fiber Sludge: 140 Forest industry fiber Sludge: 140	 This value given was assumed to be based on a wet ton and was converted to dry mass assuming 50% moisture There are many types of pulp and paper mill waste with varying methane potentials. Where needed, the assumed moisture content is given in parentheses. The L_o values were calculated with MCF = 1.0 so that values represented an intrinsic property of the waste rather than Finland's waste management practices. The value for mixed MSW is based on research of the mean composition of MSW conducted in Finland in the 1990s 	
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Study (Reference)	Method or Equation	Model Defaults	Calculated or measured L _o (L-CH ₄ /dry kg)	Comments
Canada (Environment Canada 1999)	value specified	DOC: 0.18-0.21 ^f	Wood waste: 206.5 (20%) MSW: 204.8 (20%)	Assumed moisture content is given in parentheses. The value for wood waste is considerably higher than the value of 62.6 for branches in Table 9. There is no reference to original data in Canadian document. The ratio of the values for wood waste and MSW (~ 1) is quite different from the measurements in Table 9 (~ 0.68) .
USEPA (AP- 42)	value specified	not applicable	MSW – 125 (20%)	Assumed moisture content is given in parentheses.
USEPA (NSPS)	value specified	not applicable	MSW – 340 (20%)	Assumed moisture content is given in parentheses.
Coops et al. (1995)	analysis of field data	eqn. 5	MSW – 184.4 (20)	Assumed moisture content is given in parentheses. Based on an organic carbon content of 136 kg C/wet tonne for household waste in Holland
SCS and Augenstein (1997)	analysis of field data		MSW - 81.8 - 97.4 (20)	Assumed moisture content is given in parentheses. The range is based on alternate techniques for data analysis. Values are based on collected gas only and do not account for collection efficiencies below 100%.
Eleazer et al. (1997)	laboratory measured values	not applicable	Grass: 136.0 Leaves: 30.6 Branches: 62.6 Food: 300.7 Coated paper: 84.4 ONP: 74.3 OCC: 152.3 Office: 217.3 (346) MSW: 92.0	Data are based on laboratory measurements and expressed per dry kg. The MSW is residential waste. The higher value for office paper is the result of a recalculation done by EPA (USEPA 2002b).
		(Continued on ney	(Continued on next page. See notes at end of table.)	ole.)

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4.2
Table

Study	Method or Equation	Model Defaults	Calculated or measured L _o	Comments
(Reference)			(L-CH4/dry kg)	
Barlaz,	laboratory measured values	not applicable	MSW: 86.9	Residential waste
Schaefer, and				
Ham (1989)				
Fairweather	laboratory measured values	not applicable	MSW: 123.3 (average of 3	Residential waste
and Barlaz			reactors)	
(1998)				
Ress et al.	laboratory measured values	not applicable	MSW: 80.6 (average of 12	Residential waste
(1998)			reactors)	
Thomas and	laboratory measured values	not applicable	MSW: 122.9 (average of 2	Residential waste
Barlaz (1999)			reactors)	
Rhew and	laboratory measured values	not applicable	MSW: 120.1 (avg. of 9	Residential waste, methane production was
Barlza (1995)			reactors)	stimulated by the addition of lime to the cover
				material
^a DOC - fraction	^a DOC - fraction of waste that is degradable organic carbon (this is assumed to be on a wet weight basis)	nic carbon (this is ass	umed to be on a wet weight basi	s)
^b DOC _f – fractio	^b DOC _f – fraction of DOC that actually degrades			
^c $F -$ fraction of	F – fraction of methane in landfill gas			
	-	., 11.21 1 1		

^d MCF – factor to reduce methane production based on landfill operation:

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1.0 for a modern landfill
0.4 for a shallow unmanaged landfill (< 5m deep)
0.8 for a deeper (>5 m deep)
(Table notes continued on next page)

Waste Type	DOC (wet wt. unless
	otherwise specified)
Mixed MSW	0.197
Paper and cardboard, wet weight basis	0.4
Wood and bark, wet weight basis	0.3
Deinking waste (definition unclear as some deinking wastes are listed separately), wet weight basis	0.1
Forest industry sludge - unspecified, dry weight basis, 30% solids	0.45
Deinking sludge, dry weight basis, 30% solids	0.3
Forest industry fiber sludge, dry weight basis, 30% solids	0.3
* The source data for the values given in this table are from a reference that was written in Finnish.	hat was written in Finnish.

^e DOC for Specific Waste Types Assumed in Finland*

^g DOC for Specific Waste Types Assumed in Canada*

Waste Type	DOC (wet weight basis)
Paper and textiles	0.4
Garden and park waste, and other (non-food) organic	0.17
purcescibles	
Food waste	0.15
Wood and straw waste	0.3*
* Data adapted from Bingemer and Crutzen (1987)	
** Excluding carbon from lignin	

Rates ^a	
Decay	•
of Published	
Summary o	,
Table 4.3	

	I and T.	and to building of the potential of the
Study	$K (yr^{-1})$	Comments
Coops et al. (1995)	in multi-phase model rapidly degradable – 0.185	Decay rate is based on fitting field data to equation 5.
	moderately degradable – 0.1 slowly degradable – 0.03	Although the multi-phase model gave a slightly better data fit, the decay rate for a simple first order model is presented since this model (eqn. 3) is in
	in simple first order model 0.094	common use
SCS and Augenstein (1997)	rapidly degradable – 0.06, 0.08	- These values are based on the application of equation 3 to two separate fractions of the waste with a constant 1, as evaluated in the text. The
	slowly degradable – 0.06,	first value is based on minimization of the arithmetic error, the second
	0.04	value is based on minimization of the absolute value of the logarithmic
	in first order model (eqn. 3)	error. The corresponding fractions of rapidly and slowly degradable material are 40 and 60% for the arithmetic error technique and 20 and
	k (yr ⁻¹), L_0 (LCH ₄ /wet kg):	80% for the logarithmic error technique
	0.07, 65.4	- When the data in this study are fit to the first order model (eqn. 3), three
		combinations of k and L _o were reported to give the best fit as described
	0.03, 77.9	in the text.
AP-42	0.04 (wet); 0.02 (dry)	Wet is defined as greater than 25 inches of rain a year
CAA-NSPS	0.05	
Micales and Skog 1997	Paper: 0.035-0.07	Decay rates are based on estimated half-lives, but there is no indication that
	Wood: 0.017-0.035	these are primary data.
UK (AEA Technology	in multi-phase model	The decay rate for the rapidly degrading fraction is the same as that
2001)	0.185 (rapid)	suggested by Coops et al. (1995). See Coops et al. 1995 (above) for first
1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	0.00 (SIOW)	oldel decay late.
Sweden (Swedish Env. Prot. Agency 2001)	0.092	
Canada	0.01 (wood) 0.003-0.028 (MSW)	The values for MSW are lower than what has been suggested in other reports and no justification for these values is presented.
IPCC Good Practice Guidance (IPCC 2001)	0.05	
^a Data are for MSW unless otherwise noted	erwise noted.	

5.0 **RECOMMENDATIONS FOR ADDITIONAL WORK**

5.1 Experimental Work

As indicated throughout this report and as recognized by Pingoud, Savolainen, and Seppala (1996), the fraction of the DOC that actually degrades in landfills is not known with precision. This is due to some uncertainty in the actual extent of decomposition for each waste component as well as variability in environmental conditions in landfills. The available research that addresses the decomposition of paper and wood products is limited to two laboratory-scale studies (Stinson and Ham 1995; Eleazer et al. 1997) and two field studies (Baldwin, Stinson, and Ham 1998; Gardner et al. 2003). In the experimental design used by Baldwin, Stinson, and Ham (1998), samples of known composition were buried in bags that made it possible to expose samples to environmental conditions in the landfill. Unfortunately, this work highlights the difficulties involved in studying decomposition at field-scale. Problems included the inability to locate all samples, the contamination of samples which made weight measurements difficult, variability in landfill conditions, and the relatively short time over which samples were monitored relative to the times required for complete decomposition (6 years). Presumably, a more intensive project could address these problems though the time required to obtain sufficient data could extend 10 to 20 years in non-bioreactor landfills. Even after 20 years, some degradable material may well be present, particularly for the materials slowest to decompose. Thus, modeling will be required to extrapolate the acquired data to an ultimate extent of decomposition.

An alternate to the experimental design used by Baldwin, Stinson, and Ham (1998) would be to bury fully contained reactors filled with known samples in a landfill and to measure methane production. This design would enable a sample to be exposed to the temperature regime of the landfill and the sample decay rate could be calculated from the rate of methane production. However, it would be necessary to seed paper and wood samples with the requisite bacteria initially to insure the initiation of decomposition. In addition, these samples would not be exposed to leachate from the mixed refuse which would likely be a significant source of nutrients for nitrogen limited samples such as paper and wood. Thus, it would be desirable to add leachate from a mixed waste landfill to provide nutrients. Unfortunately, the addition of leachate would stimulate decomposition rates so that the measured rate no longer reflected decomposition in a non-bioreactor landfill.

Finally, in the short-term, it would be useful to repeat the work of Eleazer et al. (1997) with wood products.

5.2 Additional Assessment of Available Data

The work by SCS and Augenstein (1997) should be reviewed to identify specific recommendations for an appropriate decay rate. While the report contains valuable data, additional work with these data might result in a more specific recommendation for a useful decay rate

There are consulting firms in the U.S. that have databases with information on the volume of landfill gas recovered from specific landfills. Firms have in some cases used these data to derive decay rates. Thus, there may be additional information that could be used to obtain further estimates of the decay rate for waste buried in landfills. However, care should be taken with this approach for several reasons:

a. At least some of these data are likely the same as that published by SCS and Augenstein (1997) and can be evaluated independently.

- b. The amount of field data used to derive a decay rate may be limited to a period of a few years. In addition, the best fit model may be derived by varying both k and Lo which results in fitted parameters that may not be appropriate for use in a predictive mode.
- c. The collection efficiency may not be considered in the data analyzed and the collection efficiency likely varies with time for landfills that have not yet received their final cover. In addition, while techniques are available to measure the collection efficiency, this parameter is most often estimated using engineering judgment.

5.3 Quantification of the Impact of Uncertainty on Model Outputs

An emphasis of this report has been to identify appropriate decay rates and L_o values for use in models to estimate methane production that can be attributed to paper and forest industry products on an annual basis. The major inputs to such an estimate will include the mass of waste buried, L_o , k, the manner in which the landfill is operated, annual rainfall, the gas collection efficiency, the extent of methane oxidation in landfill cover soils and the manner in which gas produced from the landfill is managed. While there is some uncertainty in all of these values, typical values are known within reasonable limits. As such, a model could be constructed to calculate both point estimates and a cumulative probability distribution in which a mean estimate and the probability of exceeding the mean estimate are presented. This would serve to quantify the extent of uncertainty in model outputs and likely provide a degree of comfort that the level of uncertainty is not so high as to limit the use of the currently available information.

5.4 Workshop to Review This Report

Finally, while this report provides an analysis of the available data, it has been developed by one person. This report could be used as the basis for a one-day workshop in which participants are asked to review the report in advance and to make recommendations for improved data analysis and experimental protocols. Such a workshop would have to be arranged in a way to insure that participants receive proper credit for their ideas in subsequent publications.

6.0 SUMMARY AND CONCLUSIONS

There has been a wide range of studies on the degradability of refuse buried in landfills. These studies are generally consistent in demonstrating that MSW decomposes in landfills and that the ratios of cellulose plus hemicellulose to lignin (CH:L ratios) can go as low as at least 0.2, and in the case of Wang, Byrd, and Barlaz (1994), as low as 0.02. Based on the composition of refuse presented in Tables 1.2 to 1.4, it is apparent that cellulose and hemicellulose decompose in landfills and that the major input of these constituents to landfills is paper. While most of the research conducted has focused on mixed refuse, a few studies presented data specific to paper and paperboard, and wood products. Laboratory-scale work has established the ultimate biodegradability for individual paper types and branches. However, it is not clear that the branches data can be extended to wood and it would be useful to repeat the work of Eleazer et al. (1997) with selected wood products. Limited field-scale work has been conducted to assess the biodegradability of paper and wood products in landfills. While this work has not established the ultimate biodegradability of these materials, the available field data indicate that some paper and wood products may degrade quite slowly.

The use of a multi-phase model for landfill gas production is justified and appropriate based on current knowledge of the degradability of specific waste components. The use of such a model would facilitate studies in which researchers are interested in the behavior or impact of specific waste components on a process or policy alternative. Although practitioners focused narrowly on

recoverable gas may continue using a single-phase model, this should not discourage the use of a multi-phase model where it can be helpful.

While the relationship between methane production and solids decomposition is understood, there is not currently an attempt to link these two measures. This is surprising given the interest in carbon sequestration which, by definition, represents the initial carbon buried minus that converted to methane and carbon dioxide. To relate methane production and carbon sequestration, consider 1 dry kg of waste with the following composition: 45% cellulose which is 44.4% C; 10% hemicellulose which is 45.4% C; 15% lignin which is assumed to be 50% C. Thus, this 1 kg of dry refuse would contain 320.2 gm C from cellulose, hemicellulose, and lignin, in addition to carbon from plastics and other organics. To evaluate how much of this carbon is assumed to be sequestered by using a landfill gas model, assume that this 1 kg of dry refuse has an ultimate methane yield of 1.875 ft³ CH₄/dry lb (1.5 ft³ CH₄/wet lb at 20% moisture on a wet weight basis). This ultimate yield is equivalent to the production of 106.2 L of gas $(CH_4 + CO_2)$ or 113.8 gm C per kg. Thus, 206.4 gm C would be stored per kg of dry waste. This same calculation can be performed for any waste component based on its methane yield. Thus, in the absence of more specific information, the methane yield of any component can be used to calculate the amount of carbon that is sequestered. This approach can be used to calculate carbon storage factors from the ultimate methane yield data presented in Table 4.4. In Table 6.1, the resulting carbon storage factors of primary interest to the forest products industry are summarized.

Material	Reference	L CH ₄ /kg dry matl.	Fraction carbon ^a	Carbon Storage Factor ^b
MSW	Eleazer et al. 1997	92	0.23	0.62
Coated Paper	Eleazer et al. 1997	84.4	0.43	0.79
Newspapers	Eleazer et al. 1997	74.3	0.5	0.84
Corrugated Boxes	Eleazer et al. 1997	152.3	0.42	0.61
Office Papers	USEPA 2002b (recalculated from Eleazer et al. 1997)	346	0.42	0.12
Paper and Cardboard	Technical Research Center of Finland 2001	196.5	0.45	0.53
Wood and Bark	Technical Research Center of Finland 2001	200	0.5	0.57
Deinking Waste	Technical Research Center of Finland 2001	93.4 to 140	0.25	0.60 to 0.70
Forest Industry Sludge	Technical Research Center of Finland 2001	140 to 210	0.45	0.67 to 0.75
Wood Waste	Environment Canada 1999	206.5	0.5	0.56
Pulp and Paper Mill Waste	Swedish EPA 2001	126	0.45	0.70

Table 6.1 Summary of Carbon Storage Factors of Interest to the Forest Products Industry

^a The first 5 values in this column reflect the values used by EPA in USEPA 2002b. The rest are assumed.

^b defined as the fraction of the carbon in the original product that is sequestered in the landfill

There is little quantitative information on the rates at which forest products decay in landfills. Landfill gas generation data generated at municipal solid waste landfills suggest that decay rates for combined waste vary by more than an order of magnitude. The data in Table 4.4 reveal first order decay rates ranging from 0.003 to 0.094 yr⁻¹. Most of the values for first order decay rates are in the range of 0.02 to 0.07 yr⁻¹.

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APPENDIX A COMPOSITION OF WASTE IN OTHER COUNTRIES

A limited amount of information on waste composition in other countries was readily available. Waste composition data for Spain, the UK, and Singapore are presented below. In addition, less detailed information on the biodegradable fractions of waste throughout the world were developed for USEPA's estimate of global methane emissions (Doorn and Barlaz 1995). These data are also presented.

Material	Percentage of MSW
Paper	21%
Textiles	5%
Plastics	11%
Glass	7%
Metals	4%
Organics	44%
Other	8%
Total	100%

Table A1 Composition of MSW in Spain

SOURCE: Spanish Ministry of the Environment 2000 Annual Report: www.mma.es/info_amb/estado_ma/memor/memoria00/index.htm (accessed summer, 2002)

		1		
Material	DEMOS	INCPEN	Enviros Aspinwall	Average
	(1999)	(2000)	(2000)	
Paper and paper packaging	22.6	23.0	19.5	21.7
Plastics	4.5	11.0	8.1	7.9
Textiles	2.4	3.0	6.8	4.1
Glass	8.3	6.0	8.4	7.6
Kitchen and garden waste	44.3	44.0	38.8	42.4
Metal	3.0	5.0	3.6	3.9
Miscellaneous	14.9	8.0	14.9	12.6

Table A2 Results of MSW Composition Studies for the UK

SOURCE: *Waste Watch information sheet: History of waste*; http://www.wastewatch.org.uk/informtn/ history.htm#analysis (accessed November 28, 2002)

			Waste	Waste	
	Waste	Waste	Disposed	Recycled	Recycle
	Generated	Compositio	(metric	(metric	Rate
Waste Type	(metric tons)	n (%)	tons)	tons)	(%)
Food Waste	1,092,500	23.5	1,081,700	10,800	1.0
Paper/Cardboard	1,000,800	21.6	573,900	426,900	42.7
Plastics	205,700	4.4	161,500	44,200	21.5
Construction Debris	319,000	6.9	125,300	193,700	60.7
Wood/Timber	273,200	5.9	247,900	25,300	9.3
Horticultural Waste	130,600	2.8	75,200	55,400	42.4
Earth Spoils	75,200	1.6	75,200		
Ferrous Metals	989,000	21.3	75,200	913,800	92.4
Non-Ferrous Metals	89,500	1.9	13,900	75,600	84.5
Used Slag	218,700	4.7	119,700	99,000	45.3
Sludge (Industry/PUB)	50,100	1.1	50,100		
Glass	35,500	0.8	30,600	4,900	13.8
Textile/Leather	25,000	0.5	25,000		
Scrap Tires	11,900	0.3	5,500	6,400	53.8
Others	126,600	2.7	125,300	1,300	1.0
Total	4,643,300	100	2,786,000	1,857,300	40.0

 Table A3
 Waste Generation and Recycling Rates in Singapore in 2000

SOURCE: Ministry of the Environment: The Annual Report 2000, http://www.nea.gov.sg/cms/ccird/pg_34_41.pdf (accessed June 8, 2003)

Country	Metric Tons	Food	Paper) Yard	Fraction to
	Generated	(%)	(%)	Waste (%)	Landfill
United States (from USEPA 2002a)	2.11E+06	11.2	37.4	12.0	55.3
Africa					
Algeria	5.20E+06	47	16	2	0.4
Angola	3.00E+05	37	14	5	0.4
Benin	7.00E+05	52	16	8	0.4
Botswana	2.00E+05	37	14	5	0.4
Burkina	1.10E+06	52	16	8	0.4
Burundi	7.00E+05	63	6	2	0.4
Cameroon	2.20E+06	52	16	8	0.4
Central African Republic	4.00E+05	52	16	8	0.4
Chad	8.00E+05	17	2	0	0.4
Congo	4.00E+05	52	16	8	0.4
Egypt	3.90E+06	47	16	2	0.7
Ethiopia	1.70E+06	17	2	0	0.4
Gabon	2.00E+05	52	16	8	0.7
Ghana	2.30E+06	76	7	0	0.4
Guinea	7.00E+05	52	16	8	0.4
Ivory Coast	2.20E+06	52	16	8	0.4
Kenya	1.30E+06	63	6	2	0.4
Lesotho	2.00E+05	23	25	15	0.4
Liberia	4.00E+05	17	2	0	0.4
Libya	8.00E+05	47	16	2	0.4
Madagascar	1.60E+06	63	6	2	0.4

Table A4 Composition of the Biodegradable Fraction of Waste Throughout the World
(adopted from Doorn and Barlaz, 1995)

	Ta	ble A4 (Cont	inued)		
Country	Metric Tons Generated	Food (%)	Paper (%)	Yard Waste (%)	Fraction to Landfill
Malawi	1.10E+06	63	6	2	0.4
Mali	1.30E+06	17	2	0	0.4
Mauritania	3.00E+05	17	2	0	0.4
Mauritius	2.00E+05	63	6	2	0.4
Morocco	3.90E+06	47	16	2	0.4
Mozambique	2.20E+06	63	6	2	0.6
Namibia	4.00E+05	23	25	15	0.7
Niger	9.00E+05	17	2	0	0.4
Nigeria	1.62E+07	52	16	8	0.4
Rwanda	9.00E+05	63	6	2	0.4
Senegal	1.10E+06	52	16	8	0.4
Sierra Leone	6.00E+05	52	16	8	0.4
Somalia	3.00E+05	47	16	2	0.4
South Africa	1.12E+07	23	25	15	0.9
Tanzania	2.30E+06	63	6	2	0.7
Togo	5.00E+05	52	16	8	0.4
Tunisia	1.40E+06	47	16	2	0.6
Uganda	9.00E+05	70	5	10	0.2
Zaire	3.20E+06	63	6	2	0.4
Zambia	1.30E+06	37	14	5	0.4
Zimbabwe	1.40E+06	37	14	5	0.9
Sudan	3.10E+06	47	16	2	0.4

 Table A4 (Continued)

	Ta	ble A4 (Cont	inued)		
Country	Metric Tons Generated	Food (%)	Paper (%)	Yard Waste (%)	Fraction to Landfill
Asia					
Afghanistan	1.30E+06	29	8	3	0.4
Armenia	7.00E+05	33	20	10	0.7
Azerbaijan	1.50E+06	33	20	10	0.7
Bangladesh	7.20E+06	35	3	4	0.8
Bhutan	1.00E+05	35	3	4	0.4
China	3.16E+08	29	8	3	0.6
Hong Kong	2.10E+06	9	32	0	0.7
India	5.83E+07	35	3	4	0.6
Indonesia	2.17E+07	72	6	3	0.6
Iran	1.21E+07	36	16	15	0.6
Iraq	4.30E+06	36	16	15	0.6
Israel	1.10E+06	36	16	15	0.7
Japan	4.10E+07	31	36	5	0.3
Jordan	9.00E+05	36	16	15	0.4
Kampuchea	1.10E+06	37	14	15	0.4
Kazakhstan	2.40E+06	33	20	10	0.7
Kirghizia	9.00E+05	33	20	10	0.7
Kuwait	1.30E+06	8	32	18	0.7
Laos	8.00E+05	37	14	15	0.4
Lebanon	8.00E+05	36	16	15	0.4
Malaysia	3.50E+06	64	12	6	0.6
Mongolia	2.00E+05	29	8	3	0.5
Myanmar (Burma)	3.90E+06	37	14	15	0.6
Nepal	1.30E+06	35	3	4	0.4
North Korea	4.70E+06	29	8	3	0.6

Table A4 (Continued)

Country	Metric Tons Generated	Food (%)	Paper (%)	Yard Waste (%)	Fraction to Landfill
Omen	5 00E±05	47	16	2	0.7
Oman	5.00E+05				
Pakistan	8.70E+06	46	2	2	0.4
Philippines	5.10E+06	31	17	12	0.4
Saudi Arabia	7.50E+06	8	32	18	0.7
Singapore	1.00E+06	41	28	3	0.7
South Korea	3.32E+07	26	14	2	0.6
Sri Lanka	1.70E+06	68	8	7	0.6
Syria	2.70E+06	36	16	15	0.4
Tadzhikistan	7.00E+05	33	20	10	0.7
Taiwan	2.70E+06	27	20	3	0.7
Thailand	5.20E+06	37	14	15	0.5
Turkmenistan	5.00E+05	33	20	10	0.6
United Arab Emirates	9.00E+05	8	32	18	0.7
Uzbekistan	4.20E+06	33	20	10	0.6
Vietnam	1.38E+07	72	6	3	0.6
Yemen	1.70E+06	50	16	7	0.4
Europe					
Albania	7.00E+05	36	16	15	0.4
Austria	5.50E+06	17	33	8	0.6
Belgium	3.20E+06	36	24	16	0.8
Bulgaria	2.20E+06	36	16	15	1.0
Byelorussia	2.20E+06	33	20	10	0.9
Czech R. & Slovakia	3.90E+06	33	20	10	0.7
Denmark	4.00E+06	23	34	10	0.3

Table A4 (Continued)

Table A4 (Continued)					
Country	Metric Tons Generated	Food (%)	Paper (%)	Yard Waste (%)	Fraction to Landfill
Estonia	3.00E+05	33	20	10	0.9
Finland	3.30E+06	18	35	11	0.9
France	4.98E+07	17	30	8	0.5
Georgia	1.20E+06	33	20	10	0.7
Germany	6.00E+07	19	21	9	0.7
Greece	2.10E+06	60	20	4	1.0
Hungary	3.20E+06	17	33	8	1.0
Ireland	1.10E+06	16	34	7	1.0
Italy	1.73E+07	29	22	13	0.8
Latvia	1.40E+06	33	20	10	0.4
Lithuania	8.00E+05	33	20	10	0.9
Moldavia	8.00E+05	33	20	10	0.7
Netherlands	8.50E+06	36	24	16	0.5
Norway	2.00E+06	19	40	11	0.7
Poland	7.90E+06	33	20	10	1.0
Portugal	2.90E+06	29	22	13	0.8
Romania	4.50E+06	33	20	10	1.0
Russia	3.18E+07	33	20	10	1.0
Spain	9.40E+06	35	3	4	0.8
Sweden	2.30E+06	19	40	11	0.4
Switzerland	2.80E+06	20	33	9	0.2
Turkey	1.18E+07	36	16	15	0.9
Ukraine	1.12E+07	33	20	10	0.9
United Kingdom	3.56E+07	16	34	7	0.9
Yugoslavia (former)	4.20E+06	33	20	10	0.7

	Ta	able A4 (Cont	inued)		
Country	Metric Tons Generated	Food (%)	Paper (%)	Yard Waste (%)	Fraction to Landfill
Oceania					
Australia	1.20E+07	36	30	24	1.0
New Zealand	2.10E+06	36	30	24	0.7
Papua New Guinea	3.00E+05	72	6	3	0.4
Solomon Islands	0.00E+00	63	6	2	0.4
Other non-USA					
Argentina	8.10E+06	32	26	2	0.4
Bolivia	1.20E+06	56	22	2	0.4
Brazil	3.10E+07	32	26	2	0.7
Canada	2.13E+07	21	39	13	0.9
Chile	2.90E+06	32	26	2	0.4
Columbia	5.30E+06	56	22	2	0.4
Costa Rica	5.00E+05	40	17	16	0.6
Cuba	2.30E+06	40	17	16	0.4
Dominican Republic	1.00E+06	40	17	16	0.4
Ecuador	1.70E+06	56	22	2	0.4
El Salvador	7.00E+05	40	17	16	0.4
Guatemala	1.10E+06	40	17	16	0.4
Haiti	6.00E+05	40	17	16	0.4
Honduras	6.00E+05	40	17	16	0.4
Jamaica	4.00E+05	40	17	16	0.4
Mexico	1.98E+07	11	22	1	0.7
Nicaragua	8.00E+05	40	17	16	0.7
Panama	3.00E+05	56	22	2	0.4

Table A4 (Continued)

	10		macaj		
Country	Metric Tons Generated	Food (%)	Paper (%)	Yard Waste (%)	Fraction to Landfill
D		<i></i>	10	<i>(</i>	0.4
Paraguay	5.00E+05	55	12	6	0.4
Peru	3.60E+06	30	21	2	0.4
Puerto Rico	1.20E+06	8	32	18	0.7
Trinidad and Tobago	2.00E+05	40	17	16	0.7
Uruguay	6.00E+05	32	26	2	0.4
Venezuela	4.20E+06	40	35	5	0.5

Table A4 (Continued)

APPENDIX B

EXPLANATION OF HALF-LIVES ATTRIBUTED TO AUGENSTEIN AND PACEY (1991)

The following references were retyped from Augenstein and Pacey (1991) to provide detail on the source of the information.

Reference 19: McGuinn, Young (Radian Corporation). "Use of a Landfill Gas Generation Model to estimate Volatile Organic Carbon Emissions from Landfills." Memorandum to Susan Thorneloe, U.S. Environmental Protection Agency. June 1988. This work used methane recovery data from a large number of existing landfills, in combination with assumptions on yield, to back calculate first order rate constants for use in a Scholl Canyon model. Recovery efficiencies were unknown but assumed up to 100%; methane yield figures up to approximately 3.7 cubic feet per pound were assumed. Rate constant values (k) ranging from 0.001 to 0.21 year⁻¹ were obtained corresponding to $t_{1/2}$ values of 4 to 700 years. Despite the likelihood, recognized by this survey, or large uncertainties (deriving from data quality, and gas yield and recovery assumptions, among other causes) this survey's results are suggestive of a very wide variation in generation time for U.S. landfills.

Reference 20: Soriano, P. (personal communication with Y. McGuinn, 1988, cited from reference 19 above). Scholl Canyon Model first order constants were estimated for "dry", "medium", and "wet" precipitation areas of the U.S. at 0.03 - 0.07, 0.07 - 0.14, and 0.14 - 0.3 years⁻¹. These values would result in values of $t_{1/2}$ as stated in the text.