

The Decomposition of Forest Products in Landfills

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Large quantities of forest products are disposed of in landfills annually. The fate of this vast pool of carbon is important since carbon sequestration and the generation of landfill gases have important implications for global warming. Published estimates of methane yields were used to estimate the amount of carbon released into the atmosphere from landfilled forest products. These calculations suggest that maximally only 30% of the carbon from paper and 0-3% of the carbon from wood are ever emitted as landfill gas. The remaining carbon, approximately 28 Tg in 1993, remains in the landfill indefinitely. Some of this carbon may be removed during leachate treatment, but a large portion is permanently sequestered where its impact on global warming is negligible. The placement of forest products in landfills serves as a significant carbon sink, and its importance in the global carbon balance should not be overlooked. Published by Elsevier Science Limited

INTRODUCTION

In 1993, the US Environmental Protection Agency (EPA) (1994) estimated that 50 Tg¹ of paper and 18.6 Tg of wood were discarded in the United States in municipal solid waste (MSW). Approximately 16% of all discarded MSW is incinerated (EPA, 1994); the remainder is disposed of in landfills. It has been estimated that 30-40% of US landfill volume is taken up by paper; 13% by newspaper alone (Barlaz *et al.*, 1990; EPA, 1994; Rathje & Murphy, 1992a; Barlaz *et al.*, 1990; EPA, 1994). This represents a tremendous amount of carbon that is being buried every year. What happens to this carbon? Are landfills our most effective carbon sink, removing large amounts of carbon from the environment? Is this carbon metabolized by landfill bacteria and released back into the environment as carbon dioxide and methane? If so, how much of these greenhouse gases is generated from landfilled wood products? Methane is an extremely potent greenhouse gas, twenty-five times more effective than carbon dioxide at blocking radiative heat loss from the earth (Rodhe, 1990). Are methane and carbon dioxide emissions from the decomposition of forest products in landfills high enough to affect global

warming? President Clinton's Climatic Change Action Plan (Clinton & Gore, 1993) sets a US objective of restraining carbon emission growth and increasing sequestration by a total of 100 Tg/year by the year 2000. Can the emission of carbon dioxide and methane be reduced by increased recycling of forest products within the United States? This paper will review the processes involved in anaerobic decomposition of forest products in landfills and will estimate the amount of forest product carbon that will be emitted and the amount of carbon that is permanently sequestered by recent additions to US landfills.

QUANTITIES OF FOREST PRODUCTS DISCARDED IN LANDFILLS

We must first estimate how much wood-based material is being discarded annually in MSW and other waste streams. A recent EPA report (EPA, 1994) estimates the quantity of wood-based materials in MSW based on material flows methodology using production data for the materials in the waste stream with adjustments for imports, exports, and product lifetimes. The report estimates how much of each material is generated, how much is discarded, and how much is reprocessed via recycling. The amount of

¹Tg = teragram = 1 million metric tons (tonnes). 1 tonne = 1.102 short tons.

discarded wood and paper products is shown in Table 1.

The EPA study concentrates only on material found in the MSW stream. It does not include construction and demolition debris, which can also contain significant quantities of woody material. Waste from the construction and remodeling of residential and nonresidential buildings has been estimated at 6.1 Tg of wood and 0.2 Tg of paper and paperboard. Demolition waste contributes an additional 22.7 Tg of wood and 0.3 Tg of paper and paperboard to the waste stream (Younquist *et al.*, in press). Not all construction and demolition waste reaches the landfill; increasing amounts are being recycled. Field studies have shown consistently that construction and demolition debris make up 28% of landfill discards (by both volume and weight) (Rathje, 1992) or 57 Tg in 1993 (based on EPA estimates of 147 Tg total MSW). Construction and demolition debris is composed of 50% rubble (concrete, bricks, asphalt, etc.), 25% wood products (pallets, landclearing debris, construction and demolition wood waste, and treated wood),

and 25% miscellaneous waste, which includes some lumber, plywood, particleboard, and veneer (O'Leary & Walsh, 1992). If we assume that 30% of all construction and demolition waste is wood, this would result in an additional 17 Tg (wet weight) of forest products being added to US landfills in 1993 from sources other than MSW.

STAGES OF DECOMPOSITION AND GAS GENERATION IN LANDFILLS

Decomposition in landfills occurs in a series of stages, each of which is characterized by the increase or decrease of specific bacterial populations and the formation and utilization of certain metabolic products (Aragno, 1988; Barlaz *et al.*, 1989a, 1990). Landfills are extremely heterogeneous microenvironment. Different areas of a landfill, even in close proximity, can be at different 'stages' in the decomposition process concurrently (Barlaz *et al.*, 1990).

The first stage of decomposition, which usually lasts less than a week (Augenstein & Pacey, 1991),

Table 1. Dry Weights of Materials Derived From Forest Products Discarded in the Municipal Waste Stream (EPA, 1994)^a

Products	Thousands of tonnes						
	1960	1970	1980	1990	1991	1992	1993
Newspapers	3838	5210	5834	6349	5130	5297	5094
Books and Magazines ^b	1328	1604	2206				
Books ^c				610	515	566	602
Magazines ^c				1756	1292	1386	1487
Office papers	922	1429	2177	3185	2910	2997	3280
Telephone books ^c				414	450	464	493
Third class mail ^c				2576	2358	2213	2518
Other commercial printing	820	1299	1937	2823	2823	3178	3178
Tissue paper and towels	791	1495	1669	2155	1959	1995	2184
Paper plates and cups	196	305	435	472	493	544	602
Trash bags ^c				566	559	610	646
Disposable diapers		268	1676	1916	1974	1995	1959
Other nonpackaging paper	1887	2482	3040	2801	2888	3018	3461
Corrugated boxes	3490	7241	7786	9055	8700	8773	8511
Milk cartons ^c				363	363	348	341
Folding cartons ^c				2808	2902	2837	3077
Other paperboard packaging	2569	3120	232	210	196	203	218
Bags and sacks ^c				1611	1429	1437	1342
Wrapping papers ^c				80	58	58	51
Other paper packaging	1974	2460	617	740	762	813	798
Wood packaging	1542	1596	3038	5774	5620	6060	6276
Other wood in MSW (furniture, miscellaneous durable goods, pallets, etc)	2321	3068	5212	9190	9097	9090	9537
Total	21 676	31 577	35 858	55 452	52 477	53 882	55 655

^aDry weights calculated based on average moisture contents of 20% for paper products and 15% wood (Augenstein, 1992).

^bCategory subdivided after 1990.

^cData not collected before 1990.

is characterized by the removal of oxygen from the waste by aerobic bacteria. Fresh oxygen can continue to diffuse into the top layers of refuse through the cover soil to a depth at which it is consumed by microbial and chemical processes. The environment then becomes oxygen-limiting, and facultative bacteria shift to a fermentative metabolism (Augenstein & Pacey, 1991; Barlaz *et al.*, 1989a).

In the second stage, which has been termed the "anaerobic acid stage," a diverse population of hydrolytic and fermentative bacteria hydrolyze polymers, such as cellulose, hemicellulose, proteins, and lipids, into soluble sugars, amino acids, long-chain carboxylic acids, and glycerol. These hydrolytic products are then fermented further into short-chain carboxylic acids, carbon dioxide, hydrogen, acetate, and alcohols (Mackie *et al.*, 1991; Wolin, 1982; Zehnder, 1978). As acids are produced, the pH of the refuse decreases but then begins to stabilize as acetate becomes a bacterial carbon source and is removed from the environment (Barlaz *et al.*, 1990). Buffering also occurs by the release of metallic cations from the reaction of the acids on the metallic and mineral fractions of the waste, thus diluting the hydrogen ions. Sodium and potassium cations are very soluble in a wide pH range and are unlikely to be reprecipitated once they are released. The volatility of calcium varies widely within the pH range of 4.8, with increased volatility at low pH. Large quantities of calcium can thus be dissolved to counteract high acid concentrations. As the pH reaches approximately 6.0, the calcium reprecipitates. This variable calcium buffering at low pH, combined with the permanent sodium and potassium buffering at neutral pH, permits the bacteria to be homeostatic and to establish a neutral leachate in which acidic compounds can still exist as substrates (Young, 1995).

The development of a neutral pH allows the landfill to enter the final stage of decomposition which involves the most complex interaction of microorganisms. Obligate hydrogen-producing (proton-reducing) acetogens further oxidize the remaining short-chain carboxylic acids into acetate, carbon dioxide, and hydrogen. These reactions are thermodynamically unfavorable in the presence of hydrogen, and they can only proceed if the hydrogen is removed as it is produced. The acetogens therefore must grow in association with hydrogen-consuming bacteria, such as those that reduce sulphate or produce

methane. This bacterial association is termed "syntrophy" and occurs when two or more microorganisms combine their metabolic resources to catabolize a substance that cannot be catabolized by either species alone (Aragno, 1984). Bacteria associated with syntrophy often cannot be grown in pure culture. In natural environments, they form flocs, pellets, and other aggregates that put them in close proximity with their syntrophic partners (Mackie *et al.*, 1991; Schick, 1992).

The type of syntrophic relationship that develops in the landfill is dependent on the terminal electron acceptors available. Nitrates and sulphates can both serve as terminal electron acceptors and have a higher affinity for hydrogen than does carbon dioxide due to their higher redox activities. If nitrates or sulphates are present in the landfill, methanogenic bacteria will not grow and methane will not be produced (Barlaz *et al.*, 1990; Suflita *et al.*, 1992; Wolin, 1982; Zehnder, 1978). In the absence of nitrates and sulphates, methane is formed from the small molecular weight products of the proton-reducing acetogens by CO₂ reduction, acetate cleavage, or methyl group reduction of C-1 compounds such as trimethylamine or methanol (Whitman *et al.*, 1992). These separate reactions are often performed by different species of methanogens. The removal of these small molecular weight substrates increases the rate of acid degradation, thus increasing substrate availability for all the different methanogenic species. Some methanogens are able to derive energy from several different compounds (Whitman *et al.*, 1992). They can often grow rapidly on the most favored substrate and then switch over to alternate energy sources after the favored one is depleted (Young, 1995).

The methanogens are the least robust of the landfill bacteria (Young, 1995). Only a few species have been identified, including *Methanobacterium formicicum*, *Methanobacterium bryanti*, *Methanosarcina barkeri*, an unidentified coccus (Fielding *et al.*, 1988), and an unidentified species of *Methanogenium* (Barlaz *et al.*, 1990). The initial populations of methanogens in the refuse are usually fairly low and unequally distributed. They grow more slowly than the other species and are more susceptible to adverse conditions (Suflita *et al.*, 1992; Young, 1995). Methane production may be prevented or delayed if the methanogenic population cannot become established. The onset

of methanogenesis indicates that the landfill ecosystem is in balance. If the activity of the fermentative microorganisms greatly exceeds that of the acetogens and methanogens, organic acids and hydrogen accumulate, the pH drops excessively, and the growth of methanogens is inhibited. In the absence of hydrogen scavengers, lactate, ethanol, and other reduced end-products are formed but are not further oxidized (Barlaz *et al.*, 1987). When fermentative and methanogenic activities are not balanced, the reduced end-products accumulate in the leachate (Barlaz *et al.*, 1990).

If methanogenesis does become established, methane production can last for 8–40 years (Augenstein & Pacey, 1991), although quantities of methane high enough for commercial production are generally formed for only 5–20 years (Sufлита *et al.*, 1992). Eventually methane production decreases and the landfill becomes an inert soil-like mass (Aragno, 1988; Bogner, 1990a; Bogner & Spokas, 1993, 1995a, b). Although methane and carbon dioxide are both formed during anaerobic decomposition in landfills, most of the modeling literature is centered on methane production. Methane can be burned as an energy source, so there is a commercial interest in this area. Methane is also extremely important due to its pronounced effect on global warming (Augenstein, 1992). In controlled laboratory experiments, landfill gas generated during the methanogenic stages of refuse decomposition was composed of approximately 55–60% methane and 40–45% carbon dioxide with trace amounts of other gases (Barlaz *et al.*, 1989a). Landfill gas generated from the degradation of cellulose, the principle metabolizable component of forest products, yielded 51% methane and 49% carbon dioxide (O'Leary & Walsh, 1991). Carbon dioxide is more highly soluble in water than methane, and a significant portion of the carbon dioxide may be transferred to the leachate, thus increasing the methane: carbon dioxide ratio of the gas that is actually emitted from the landfill (Christensen & Kjeldsen, 1995).

FACTORS THAT INFLUENCE THE RATE OF DECOMPOSITION IN LANDFILLS

Many different factors influence the rate of decomposition in landfills. These include (1) waste management and processing variables, such as the

size of the waste particles (i.e. whether the material is shredded, crushed, or bailed), (2) the composition of the waste, (3) factors that influence bacterial growth, such as moisture, available nutrients, pH, and temperature, (4) the design of the landfill and whether landfill gases are contained or extracted (for flaring or for energy production), and (5) the operation of the landfill, including the amount of compaction, the type and thickness of covering materials, the amount of natural moisture from precipitation or groundwater, and whether the leachate is recycled or removed for treatment (Augenstein & Pacey, 1991; Bogner & Spokas, 1993; Laquidara *et al.*, 1986).

Most studies have concluded that the most critical Factor in landfill decomposition is the amount of moisture in the refuse (Bogner & Spokas, 1993; Halvadakis *et al.*, 1988; Rees, 1980; Sufлита *et al.*, 1992). Landfills are strictly managed to prevent the infiltration of rainwater, thus preventing the formation of large quantities of leachate. Leachate develops when free water in the landfill percolates through the waste, accumulating many metabolic byproducts and toxic compounds. Leachate can contaminate groundwater when leaks develop in landfill liners, so regulations are in place to minimize leachate formation. Much of the moisture present in landfills originates from the waste itself. Augenstein (1992) estimated that the average moisture content of landfill waste is 22%, with paper having 20% moisture and wood averaging 15%. Landfills with higher moisture contents, such as the Fresh Kills Landfill outside of New York City, have much higher rates of biodeterioration in comparison to normal landfills. In the Fresh Kills landfill, only 14.4% of exhumed waste was recognizable as paper. This value is usually 40% in normal 'dry' landfills (Sufлита *et al.*, 1992). Moisture levels can vary greatly within a single landfill, thus causing highly variable rates of decomposition within close proximity (Blakey *et al.*, 1995; Straka *et al.*, 1993; Sufлита *et al.*, 1992). It is very common to find intact food, paper, and other degradable materials that are 20–30 years old in one part of a landfill and high levels of decomposition in other areas (Sufлита *et al.*, 1992).

Peer *et al.* (1993) determined that climatic moisture is not an important variable in estimating methane emission levels. Methane recovery data for 25 different landfills were

surveyed and related to climate, age of refuse, and physical characteristics at the site. Methane recovery was correlated with the mass of refuse in the landfill, landfill depth, and age of the refuse. There were no statistically significant relationships between methane recovery and climatic factors, including precipitation, temperature, and dew point. These conclusions may reflect the impermeability of modern landfills to external sources of water.

DECOMPOSITION OF CELLULOSE, HEMICELLULOSE, AND LIGNIN IN LANDFILLS

Cellulose and hemicellulose can both be degraded under the anaerobic conditions found in landfills. These two compounds make up 91% of the methane potential of most refuse (Barlaz *et al.*, 1989b). It has been estimated (Barlaz *et al.*, 1990) that 41% of municipal refuse is composed of paper and paperboard, 7.9% is food waste, and 17.9% is yard waste. The majority of cellulose and hemicellulose in landfills therefore originates from forest products.

Cellulose can be quite variable in its biodegradability due to variations in its degree of crystallinity and its association with lignin. The cellulose found in paper is less resistant to biodegradation than 'fresh' cellulose since its primary structure has already been destroyed during the pulping process (Rees, 1980). Cellulose is hydrolyzed into glucose and cellobiose in the landfill. These compounds are then fermented into other compounds, including carbon dioxide, hydrogen, ethanol, and acetic, propionic, butyric, valeric, and caproic acids (Rees, 1980).

Hemicellulose, which coats the cellulose fibrils in wood and paper, is also metabolized by landfill bacteria (Ghosh *et al.*, 1985). Bogner & Spokas (1995b) showed that xylan and mannan were preferentially metabolized over other hemicellulosic sugars, including arabinan, galactan, and rhamnan.

Lignin is not metabolized by anaerobic bacteria and does not significantly decompose in landfills (Aragno, 1988; Barlaz *et al.*, 1989a, b, 1990; Cummings & Stewart, 1994; Ham *et al.*, 1993; Khan, 1977; Pfeffer & Khan, 1976; Sufliita *et al.*, 1992; Wang *et al.*, 1994; Young & Frazer, 1987). In optimized laboratory studies of anaerobic degradation, 71% of the cellulose and 77% of the

hemicellulose from typical landfill refuse was degraded, Lignin degradation was negligible, even under the most ideal conditions (Barlaz *et al.*, 1989 b).

The presence of lignin can interfere greatly with cellulose and hemicellulose degradation. Wood is very resistant to decay in landfills because its cellulose and hemicellulose is embedded in a matrix of lignin (Ham *et al.*, 1993a, b; Wang *et al.*, 1994). Newsprint, which represents a major portion of the paper in landfills, contains approximately 20–27% lignin, limiting the degradation of newsprint and other low quality/high lignin containing types of paper (Cummings & Stewart, 1994; Owens & Chynoweth, 1993; Rees, 1980). Under optimal laboratory conditions, the cellulose in newsprint was degraded about 40% , while the cellulose in computer paper and Whatman filter paper was degraded 90 and 97% , respectively (Khan, 1977). It has been estimated that at least 18% of the cellulose and hemicellulose in refuse is recalcitrant to degradation because of its close association with lignin (Bingemer & Crutzen, 1987; Bogner, 1992; Bogner & Spokas, 1995a).

METHANE GENERATION MODELS

Numerous models have been proposed to predict the amount of methane produced throughout the lifetime of a landfill. These models generally fall into four different categories: zero-order, first-order, multi-phase, and second-order (Coops *et al.*, 1995).

In zero-order models, landfill gas formation from a certain amount of waste is assumed to be constant with time. This type of model is used for estimating national and global emissions with the assumption that there is no major change in waste composition or the amount of material landfilled. An example of a zero-order model is the EPA Regression Model (Peer *et al.*, 1993). This model is based solely on a linear correlation of methane recovery and refuse mass. Moisture levels, decomposability of the refuse, and other factors are not considered. The assumed methane generation potential of the refuse is estimated as 0.023–0.061 grams of methane per gram of wet refuse. This model predicts US methane emissions from landfills to be 2–6 Tg assuming an annual placement of 100 Tg refuse.

Another zero-order model that is frequently

cited in the literature was proposed by Bingemer & Crutzen (1987). This model has been used to estimate global methane emissions using mass balance analysis. The assumed methane generation potential was 0.070–0.155 grams of methane per gram of wet refuse, yielding a total methane accumulation of 7–16 Tg from US landfills based on an annual placement of 100 Tg refuse (Peer *et al.*, 1993). Most researchers (Augenstein, 1992; Bogner & Spokas, 1993; Peer *et al.*, 1993; Subak *et al.*, 1993) consider these values too high.

First-order models include the effect of age in methane generation. Landfill gas formation in a certain amount of waste is assumed to decay exponentially with time. Modifications of simple first-order models have also been made to include the build-up of the methanogenic phase and temperature dependency (Coops *et al.*, 1995).

More commonly, numerous first-order models are combined to express methane generation from different fractions of the refuse. These are called “multi-phase” models (Coops *et al.*, 1995). An example of a multi-phase model is the EMCON MGM kinetics model (Augenstein & Pacey, 1991; Augenstein, 1992). In this model, there is a lag time in which no methane is generated. This is followed by a phase of constant rate increase, followed by an exponential decrease in methane production. A useful term in this model is “ $t_{1/2}$ ” which refers to the time from placement of the waste to the time when gas generation equals half the estimated yield (similar to a ‘half-life’ for radioactive materials). Estimates by Soriano (as described in Augenstein & Pacey, 1991) imply values of $t_{1/2}$ of 10–25 years for ‘dry’ regions of the US, 5–10 years for regions with a medium level of precipitation, and 2–5 years for wet regions of the country. The actual time that a landfill generates methane may be several multiples of $t_{1/2}$ depending on the actual shape of the generation curve.

The EMCON model also separates out the decomposition rates of different types of waste: ‘rapidly decomposable’ waste (i.e. food and garden waste), ‘moderately decomposable’ waste (i.e. paper and paper products), and ‘slowly decomposable’ waste (i.e. wood and other organic materials that are resistant to anaerobic decomposition) under wet and temperate conditions. Paper products have a lag time of 1.5–2.0 years before methane production, a ‘half-life’ of 10–20 years, and a potential methane yield of 0.086–0.159 grams methane per gram of dry

refuse. Wood has a lag time of 5 years, an estimated ‘half-life’ of 20–40 years, and a potential methane yield of 0.013–0.022 grams methane per gram of dry refuse (Augenstein, 1992) or 0.033–0.061 grams of methane per gram of wet refuse assuming an average moisture content of 22%. Annual US landfill methane emissions are predicted to be 3–8 Tg assuming an annual placement of 100 Tg refuse (Peer *et al.*, 1993). These values are very similar to those calculated by Peer *et al.* (1993) using zero-order kinetics.

Second-order models have also been proposed to predict methane emissions based on the complicated chemistry and microbiology of methane synthesis. Since a large number of reactions are involved, all with different reaction rates, second-order kinetics are used to predict total methane generation. An example of this is the model by Swarbrick *et al.* (1995). This model uses both physical and biochemical parameters, but with only two rate-limiting steps. A more complex model by Young (1995) models the populations of methanogenic bacteria, treating the earlier processes as being dependent solely on substrate and nutrient availability.

Which model is the most accurate? Peer *et al.* (1993) concluded that the most important factor in the calculation was the assumed methane potential of the refuse and not the actual mathematical model used to obtain the estimates of methane emissions. Coops *et al.* (1995) validated all four models by comparing predicted values to field data from nine landfills in the Netherlands. They concluded that the first-order, second-order, and multi-phase models were all similar in describing landfill gas formation although the multi-phase model was slightly more accurate than the others in predicting actual methane emissions. The zero-order model was the most unreliable. Discrepancies between values predicted by the models and actual field data were due to uncertainties in waste quantity and composition, the heterogeneity of the landfill microenvironment, and uncertainties in the actual recovery efficiency of the methane collection system rather than from differences in the models themselves.

ESTIMATIONS OF CARBON CONVERSION

What are the maximum amounts of methane and carbon dioxide that are generated from wood-

based material in landfills? Methane yield estimates in the literature are highly varied and have been derived from theoretical stoichiometric calculations, laboratory measurements of small-scale simulated landfills, or actual field measurements from landfills (Peer *et al.*, 1993). Data from sampling full-scale landfills are extremely variable and are only 1–50% of the yields calculated by stoichiometry (Barlaz *et al.*, 1990). It is also difficult to extrapolate methane production data from laboratory tests to field-scale landfills (Barlaz *et al.*, 1990). Peer *et al.* (1993) compared published methane potentials determined by different types of studies. Values ranged from 0.003–0.193 grams of methane per gram of wet refuse. Bogner (1990a) and Bogner & Spokas (1995a) reported even broader ranges of published values, as much as six orders of magnitude (from 3×10^{-3} – 3×10^3 g m⁻² day⁻¹) in the latter report.

Despite the high degree of variability among estimates of methane potentials, some of these values have been used successfully on a commercial basis to estimate the amount of methane obtained at a given landfill. Estimations of methane generation from the EMCON model (Augenstein, 1992) are very similar to those used in the EPA regression model (Peer *et al.*, 1993). Further, Doorn & Barlaz (1995) presented methane potentials for different types of materials, including different types of paper. The methane potential given by Doorn & Barlaz (1995) for the

assumed average paper waste was practically identical to that used by the EM CON model for moderately degradable material (i.e. paper) in a temperate environment (Augenstein, 1992). These values are used in Table 2 to calculate the percent of carbon potentially released from landfilled wood and paper products and to estimate the methane potential and carbon sequestration of all forest products landfilled in 1993 (Tables 3 and 4).

These calculations lead to the conclusion that only 0–3% of the carbon from wood and an average of 26% of the carbon from paper is potentially released into the atmosphere as carbon dioxide and methane once the material has been landfilled. Different types of paper vary in the amount of methane and carbon dioxide that is released, primarily due to their lignin content. Newsprint and paper coated by insoluble materials are quite resistant to anaerobic decomposition (carbon conversion only 16–18%). Office paper and boxes are more likely to be degraded (carbon conversion 32–38%). Paper and paper products that were deposited in landfills in 1993 have a total methane potential of about 3.7 Tg (Table 3). The methane would be released gradually over a period of 5–40 years following the patterns shown in the various methane generation models (Augenstein & Pacey, 1990; Coops *et al.*, 1995; Hoeks, 1983). The landfilling of wooden materials in 1993, including construction and demolition debris, would have far less impact on methane emissions with a

Table 2. Estimate of the Maximum Proportion of Carbon in Paper and Wood that is Converted to Methane and Carbon Dioxide in Landfills

Carbon source	Methane potential (g/g) ^a	Fraction carbon released as methane ^b	Fraction carbon released as CO ₂ ^c	Total fraction carbon released as landfill gas ^d
Wood	0.000–0.013	0.000–0.019	0.000–0.013	0.000–0.032
Paper (average)	0.090	0.157	0.105	0.262
Newspaper	0.054	0.094	0.063	0.157
Boxes	0.108	0.189	0.126	0.315
Office paper	0.131	0.229	0.152	0.382
Coated paper	0.060	0.105	0.070	0.175

^aMethane potential = $\frac{\text{grams methane released}}{\text{grams dry paper/wood}}$

From Augenstein (1990) and Doorn & Barlaz (1995). Values converted from ft³/lb to g/g (at STP with a methane density = 0.7168 g/l).

^bCalculated as:

methane potential $\times \frac{\text{carbon atomic weight}}{\text{methane atomic weight}} \times \frac{1}{\text{carbon content of paper/wood}}$

Carbon content of wood = 0.51 for softwoods (Forest Products Laboratory, 1987) and 0.43 for paper (Farquhar & Rovers, 1973).

^cAssuming landfill gas is emitted as 60% methane and 40% carbon dioxide (O'Leary & Walsh, 1991), the carbon dioxide values are 2/3 of the methane values.

^dSum of fraction released as methane and fraction released as carbon dioxide.

Table 3. Estimated Maximum Potential Methane Generated From Forest Products Deposited in Landfills in 1993

	Dry weight discarded in 1993 (Tg)	Methane potential (g methane/g refuse) ^a	Total potential methane (Tg)
Newspapers	5.10	0.054	0.275
Books	0.60	0.090	0.054
Magazines	1.49	0.090	0.134
Office papers	3.28	0.131	0.430
Telephone books	0.49	0.090	0.044
Third class mail	2.52	0.090	0.227
Other commercial printing	3.18	0.090	0.286
Tissue paper and towels	2.19	0.131	0.286
Paper plates and cups	0.60	0.090	0.054
Trash bags	0.65	0.108	0.070
Other nonpackaging paper	3.46	0.090	0.311
Corrugated boxes	8.52	0.108	0.920
Milk cartons	0.34	0.060	0.020
Folding cartons	3.10	0.108	0.335
Other paperboard packaging	0.22	0.108	0.024
Bags and sacks	1.34	0.108	0.145
Wrapping papers	0.05	0.090	0.004
Other paper packaging	0.80	0.090	0.072
Total paper			3.693
Wood packaging	6.9	0-0.013	0 - 0.090
Other wood in MSW (furniture, miscellaneous durable goods, pallets, etc.)	10.5	0-0.013	0 - 0.137
Wood (construction and demolition debris)	16.1	0-0.013	0 - 0.209
Total wood			0 - 0.436

Methane potentials from Doorn & Barlaz (1995) and Augenstein (1992). The average methane yield for paper was used for paper categories with unspecified methane potentials. Dry weights calculated based on average moisture contents of 20% for paper products and 15% wood (Augenstein, 1992).

maximum methane potential of 0.4 Tg. Approximately 32 Tg of carbon would remain in the landfill from all forest products deposited that year (Table 4). This stored carbon would be partitioned into aqueous intermediates, microbial biomass, and long-term storage material (Bogner, 1992; Bogner & Spokas, 1993, 1995a, b).

The resistance of most forest products to anaerobic decomposition in landfills is significant. These estimations have important implications for carbon cycling and waste disposal. It appears that US landfills serve as a tremendous carbon sink, effectively preventing major quantities of carbon from being released back into the atmosphere.

At the heart of these calculations are the estimates of methane potential (Augenstein, 1992; Doorn & Barlaz, 1995). Augenstein (1992) admitted that EMCON's yield potentials are low compared to what could be obtained by the complete stoichiometric conversion of organic waste to methane, assumed by Bingemer & Crutzen (1987). This low conversion rate has been

validated by EMCON's own field measurements and is used commercially by this company to predict the potential methane recovery from a given landfill (Augenstein & Pacey, 1991). Similar values have been obtained in laboratory studies (Augenstein *et al.*, 1976a, b; Barlaz *et al.*, 1990) and are used in other methane emission models (Peer *et al.*, 1993). The methane yield estimates published by Doorn & Barlaz (1995) were practically identical to Augenstein's (1992). One slight difference occurred with estimating methane production from wood. Augenstein (1992) considered wood as being slowly decomposable and releasing very low levels of methane and carbon dioxide. Doorn & Barlaz (1995) considered wood as having a methane potential of zero.

Our calculations correlate well with other published estimates of carbon conversion in landfills. Aragno (1988) stated that 35-40% of organic matter in a landfill can be converted into methane and carbon dioxide under ideal conditions, but that decomposition within an

Table 4. Estimates of the Potential Amount of Carbon Sequestered and Released From Landfills From Forest Products Materials Deposited in 1993

Carbon Source	Total quantity landfilled (dry wt) (Tg)	Total carbon (Tg)	Total carbon emitted as landfill gas (Tg)	Total carbon remaining in landfills (Tg)
Paper	39.9 ^a	17.2 ^b	4.5 ^c	12.7 ^d
Wood	30.4 ^e	15.5 ^b	0.5 ^c	15.0 ^d
Total	70.3	32.7	5.0	27.7

^aEPA, 1994. Dry weight calculated based on an average moisture content of 20% for paper products (Augenstein, 1992).

^bTotal carbon = total quantity of landfilled material X average percent of carbon in paper (0.43) (Farquhar & Rovers, 1973) or wood (0.51) (Forest Products Laboratory, 1987).

^cTotal carbon emitted as landfill gas = total carbon X average percent carbon emitted from paper (0.262) or wood (0.032) as landfill gas (Table 2).

^dTotal carbon remaining in landfills = total carbon – total carbon emitted as landfill gas.

^eFrom 7.3 Tg of wood (wet weight) from packaging + 11.2 Tg of wood (wet weight) from miscellaneous durable goods in MSW (EPA, 1994) + 17.1 Tg of wood (wet weight) from construction and demolition debris (EPA, 1994; Rathje, 1992; O'Leary & Walsh, 1992; see text for explanation of calculation). Dry weight calculated based on an average moisture content of 15% for wood (Augenstein, 1992).

actual landfill would occur much more slowly. Bogner & Spokas (1993) reported carbon conversion values of 25–40% for all landfilled material, including that which is readily decomposable (and theoretically reaches a greater level of decomposition than wood or paper). They concluded that more than 75% of the carbon deposited in landfills remains as sedimentary storage. Bramryd (1982) and Richards (1989) estimated the stored carbon component at 50% or more. Again, since these authors were considering all refuse components, the portion remaining from the degradation of wood and paper would be considerably higher. Cummings & Stewart (1994, 1995) determined that newspaper is highly resistant to degradation because of its high lignin content (approximately 25%) and heavy inking (which prevents bacterial adhesion). There are also many reports of undegraded paper and wood in excavated landfills. Walsh & LaFleur (1995) reported that wood was recovered from 'landfills' in New York City that were constructed as early as 1844. Paper was found only in those landfills constructed after 1954. The older landfills were constructed in marsh and tidal land and contained much higher moisture levels than are found in today's sanitary landfills. Kinman *et al.* (1990) reported a landfill in Chicago still contained 12.3% wood (by weight) and 32.7% paper after 20 years of decomposition. Fresh refuse generally contains 30–40% paper and 6–8% wood (Barlaz *et al.*, 1990; Rathje & Murphy, 1992a,b; EPA, 1994). The rate of degradation was highly variable, however, since the amount of paper in individual samples ranged from 2 to 62%. These

disparate values may reflect the heterogeneity of the original refuse as well as variation in the rate of degradation. In a different landfill, Rathje & Murphy (1992b) compared the amount of paper deposited with the amount of paper recovered from the landfill 8 years later and found no significant decomposition during this time. Numerous reports exist of recovering newspapers that are still legible after being in a landfill for more than 20 years (Bogner, 1990a; Kinman *et al.*, 1990; Rathje & Murphy, 1992a, b).

There are a few contradictory reports in the literature suggesting higher levels of degradation. Owens & Chynoweth (1993) reported ultimate methane potentials of different types of paper that were two to three times higher than those determined by Doorn & Barlaz (1995). The high values were probably due to the extremely fine particle size of the ground paper used in laboratory assays. Bingemer & Crutzen (1987), in a widely cited manuscript, assumed from stoichiometric calculations that paper degrades entirely within 5–20 years and that the nonlignified portion of wood decomposes within 20–100 years. Stoichiometric calculations generally overestimate methane yields by as much as 50% (Barlaz *et al.*, 1990). A multi-phase model developed by Findikakis *et al.* (1988) separated the readily, moderately, and slowly degradable landfill components. The estimated time for 90% carbon conversion was approximately 5 years for moderately decomposable material (paper) and 12.5 years for slowly decomposable material (wood). This is rapid compared to other estimates (Augenstein, 1991; Hoeks, 1983). Excavations of

landfills (Kinman *et al.*, 1990; Rathje & Murphy, 1992a, b; Suflita *et al.*, 1992), in which large quantities of degradable materials are still recognizable after 20–30 years, do not support these conclusions.

The amount of cellulose and hemicellulose in landfilled refuse is known to decrease significantly with time (Bookter *et al.*, 1982; Ham *et al.*, 1993a, b; Suflita *et al.*, 1992; Wang *et al.*, 1994). The ratio of cellulose:lignin has been used to demonstrate the extent of biodegradation since the amount of lignin remains constant with time (Aragno, 1988; Barlaz *et al.*, 1989a, b, 1990; Cummings & Stewart, 1994; Ham *et al.*, 1993a, b; Khan, 1977; Pfeiffer & Khan, 1976; Suflita *et al.*, 1992; Wang *et al.*, 1994; Young & Frazer, 1987). Bookter *et al.* (1982) determined that fresh refuse has a cellulose:lignin ratio of 3.8–4.0:1.0. This ratio generally declines as the refuse depth increases (Bookter *et al.*, 1982; Ham *et al.*, 1993a, b; Jones *et al.*, 1983; Suflita *et al.*, 1992), although exceptions have been reported demonstrating the large amount of variability of decomposition within most landfills (Ham *et al.*, 1993a; Wang *et al.*, 1994). Suflita *et al.* (1992) calculated that the cellulose:lignin ratio in the Fresh Kills Landfill in New York decreased at a rate of 0.71 +/- 0.016 per year. Based on this degradation rate, it would take more than 10 years for a 50% change to occur in the cellulose:lignin ratio. Since the lifespan of most landfills is usually only 5–20 years (Suflita *et al.*, 1992), complete degradation probably would not occur.

It appears that cellulose and hemicellulose can be degraded at greater rates in landfills than indicated by methane and carbon dioxide emissions from landfills. The remaining carbon accumulates in the landfill as aqueous intermediates, microbial biomass, and recalcitrant solids (Bogner, 1992; Bogner & Spokas, 1993, 1995a, b). If free water is present in the landfill, the aqueous intermediates accumulate in the leachate, which may be removed from the landfill for treatment at local sewage plants or at the landfill site. In the future, leachate may be recirculated through the landfill to increased levels of biodeterioration in order to obtain higher methane yields and lower treatment costs (Barlaz *et al.*, 1989a, 1990; Bogner & Spokas, 1995a, b; Pohland, 1980). Leachate recirculation is permitted under RCRA (Resource Conservation and Recovery Act, subtitle D) regulations for landfills that are lined and have

leachate collection systems (Bogner & Spokas, 1995a, b). Currently less than 1% of the 6000 existing landfills in the US recycle leachate (Uehling, 1993), but the number is expected to increase (Bogner & Spokas, 1995a, b). By keeping leachate in the landfill, this process would permanently sequester additional carbon. It may also result in higher levels of methane generation and emission unless saturated conditions lead to the build-up of acidic compounds which would inhibit the onset of methanogenesis (Barlaz *et al.*, 1987). The current trend in landfill construction is to build extremely large landfills in sparsely populated areas with minimal precipitation. This practice will minimize the amount of methane released into the environment and slow biodegradation and carbon cycling to an absolute minimum (Ham, 1991).

The carbon that is not converted into landfill gas or aqueous intermediates eventually turns into a nonreactive solid mass, similar to humic material. Most of this carbon is derived from lignin although about one third of it originates from the cellulose and hemicellulose that is resistant to degradation due to its close association with lignin (Bogner & Spokas, 1995a). It has been estimated that 30 million metric tons of this material accumulates per year (Bogner & Spokas, 1995). A large portion of this inert fraction originates from forest products.

FACTORS THAT REDUCE METHANE EMISSION

When global warming is considered, the amount of methane emitted into the atmosphere is the most important by-product of the anaerobic degradation of forest products in landfills. There are several factors that reduce the total amount of methane released into the environment below that estimated in our calculations. The most important factor is lack of water. Most landfills, especially those constructed after the mid-1980s, are extremely dry, and biodeterioration slows to an absolute minimum. Refuse is mummified instead of degraded, and little or no methane is released (Rathje & Murphy, 1992a,b). The carbon conversion estimates given in Tables 2 and 3 are maximum yields only. Under field conditions, the amount of carbon converted into landfill gas is generally much lower.

The extreme variability of landfills also needs to

be stressed. The methane yields quoted in this paper are maximum values. The landfill ecosystem is a very delicate environment, and methanogenesis frequently does not reach its potential, even if moisture is available. If the refuse contains a high proportion of easily digestible materials, such as large quantities of food waste, organic acids and hydrogen may build up too rapidly and drop the pH of the refuse to a point where methanogens cannot grow (Barlaz *et al.*, 1987, 1989a). A similar situation develops if there is too much water in the landfill, as may occur when leachate is recycled. The increased level of biodeterioration associated with increased moisture can result in the build-up of too many acidic intermediates. Methanogenesis may be delayed, prevented, or inhibited (Barlaz *et al.*, 1987; Bogner & Spokas, 1995b).

High concentrations of sulfate and other alternate electron acceptors, including nitrate, iron, and manganese, may also inhibit methanogenesis since these electron transfers are thermodynamically favored over methane production as discussed earlier (Adrian *et al.*, 1994; Gurijala & Suflita, 1993; Suflita *et al.*, 1992; Zehner, 1978). Gurijala & Suflita (1993) showed that most of the paper samples collected from a landfill were covered with a fine layer of sulfate, and that the cellulose fibrils in this paper were resistant to biodeterioration. The paper and textile fragments in the landfill probably absorbed the sulfate after it had been solubilized from gypsum-based construction and demolition debris. Sulfate was not present in significant levels in fresh paper. Samples that contained construction debris did not form methane when incubated in the laboratory. The addition of molybdenum, which inhibits sulfate reduction, increased methanogenesis. Many landfills contain toxic chemicals that also prevent bacterial growth.

Another factor that mitigates the amount of methane that reaches the atmosphere is the oxidation of methane by soil-inhibiting bacteria that reside in the landfill's cover soil. These organisms metabolize a portion of the methane back into carbon dioxide. The amount of methane oxidation can be highly variable in different portions of the landfill (Novzhevnikova *et al.*, 1993). Methane that escapes rapidly from landfills through cracks or macropores in the cover soil is not exposed to methane-oxidizing bacteria and will not be oxidized (Christensen & Kjeldsen, 1995; Doorn & Barlaz, 1995). Methane oxidation

can also vary seasonally due to changes in soil moisture (Bogner *et al.*, 1995). Many references cite Mancinelli & McKay's (1985) estimate that 10% of landfill methane is oxidized in the cover soil (Bogner & Spokas, 1993; Doorn *et al.*, 1994; Doorn & Barlaz, 1995; Peer *et al.*, 1993). Higher estimates of methane oxidation have also been made (Knightly *et al.*, 1995; Whalen *et al.*, 1990). Published rates of oxidation have varied by four orders of magnitude (Bogner & Spokas, 1995c; Bogner *et al.*, 1995). Bogner *et al.* (1995) recently reported that a landfill in Illinois served as a methane sink due to extremely high levels of methane oxidation in the cover soil and a methane recovery system that lowered the overall pressure of methane in the landfill.

Some landfills collect and flare methane so that it is not released into the environment. Larger US landfills partially purify the methane and sell it as an energy source. In each of these cases, the methane is converted into carbon dioxide upon combustion. It has been estimated that approximately 1.4 Tg of methane is recovered or flared per year in the US of the 8–17 Tg of methane emitted from municipal and industrial landfills (Doorn & Barlaz, 1995). This percentage will probably increase in the near future due to the proposed Clean Air Act Amendments of 1990, which are expected to have a major impact on the amount of methane generated from both new and existing landfills (Bogner, 1990b). Methane emissions are expected to decrease by 5–7 Tg/year because of these regulations. Although recycling and incineration are both projected to increase, the amount of waste deposited in US landfills is also expected to increase to levels that will offset benefits from recycling and incineration (Doorn & Barlaz, 1995; Ham, 1991). It can be assumed that the current rate of landfill gas generation will continue for several more decades and that reductions in emissions may be expected only from increased recovery of methane (Doorn & Barlaz, 1995). The amount of carbon dioxide released into the environment will increase proportionately.

CONCLUSION

Recent global carbon balance calculations have failed to account for over a billion tonnes of carbon per year (Tans *et al.*, 1990). The amount of carbon emitted from and stored in landfills,

much of it originating from forest products, can account for some of this missing carbon. The amount of paper and wood stored in US landfills in 1993 alone has the potential of ultimately releasing 5 Tg of carbon into the atmosphere as methane and carbon dioxide. This leaves approximately 28 Tg of carbon still in the landfill. Some of this remaining carbon may be removed from the landfill during leachate treatment and may reenter the carbon cycle in another form, but a large portion of this carbon is permanently sequestered in the soil where its impact on global warming is negligible. The placement of forest products in landfills serves as an important carbon sink, and its importance in the global carbon balance should not be overlooked.

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