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Relationships between analytical methods utilized as tools in the evaluation of landfill waste stability

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Abstract

In this study, the refuse from 12 landfills of various ages ranging from fresh refuse to material 11 years old was collected, and changes in the bio-stability parameters were determined. The parameters measured included cellulose, lignin, biochemical methane potential (BMP) and volatile solids, along with plastics. These parameters, along with the cellulose to lignin ratio were compared to determine which were most indicative of the bio-stability of the refuse. Lignin and volatile solids measurements were affected by plastics in refuse samples. Plastics increased both lignin and volatile solids measurements by approximately 10%. Cellulose and volatile solids measurements correlated well with age, each other, and with BMP measurements and were therefore considered the best parameters to determine stability. Data for the Riverbend landfill, a landfill with a moisture content of 48%, which is similar to that of bioreactor landfills, showed that degradation was nearly complete after 5 years as indicated by low values for cellulose and BMP.

1. Introduction

Many solid waste landfills are now being designed or modified to permit more rapid stabilization of refuse. These ‘‘bioreactor landfills’’ are constructed similar to most sanitary landfills, but have increased moisture contents, which allow for an increased rate of biological activity. This concept was an outgrowth of the practice of leachate recirculation, which was originally designed as a method to limit the discharge of leachate and improve its quality. The leachate recirculation studies showed that leachate recirculation improved leachate quality, but in addition, the time needed for landfill stabilization was substantially reduced due to the increased moisture content.

The advantages of leachate recirculating systems have been well documented at both the bench scale and pilot-scale (Reinhart and Townsend, 1998). The practice of leachate recirculation was studied during the 1970s and early 1980s as a method of leachate treatment (Pohland, 1975; Leckie et al., 1979; Ham and Bookter, 1982). More recently, full-scale bioreactor studies have been undertaken to determine the relevant design and operational parameters for these systems (Reinhart and Townsend, 1998).

Bioreactor landfills should provide substantial economic and environmental benefits. Total degradation of the waste mass can produce settlements approaching 30% (Green, 2000) and can result in additional capacity that can be captured during the active phase of the landfill. Moreover, the

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cost of leachate management may be substantially lowered and the need for off-site management is largely eliminated.

The landfill bioreactor operation enhances the organic biological degradation rate and, therefore, increases anaerobic gas production. Methane gas generation is enhanced and concentrated during the active life of the landfill. This makes it easier to manage the gas and should lower the total mass of climate change gases emitted to the environment. The closed landfill that has been operated as a bioreactor should become much less of a long-term environmental liability and can be integrated back into the conventional land use system much more quickly. There is a larger spectrum of positive economic end uses for a closed landfill with a stabilized waste mass and no leachate or gas concerns (Green, 2000).

While the advantages of bioreactor landfills are clearly understood, documentation of their performance has been limited. Landfill stabilization may require 50 years or longer for conventional landfills, while 5 years or less may be needed to achieve stabilization using landfill bioreactor technology (Kilmer and Tustin, 1999; Pohland, 1975; Watson, 1993). The point at which waste is completely degraded and the landfill is stable is not clearly defined, but important indicators of stability include waste composition, leachate quality, and gas quantity (Reinhart and Townsend, 1998).

The purpose of this study is to evaluate the analytical methods used to determine landfill waste composition. It is important to establish both the methods that accurately describe the biodegradable or organic fraction of waste and the point at which solid waste is biologically stable and represents little or no potential environmental impact. The biochemical and physical parameters evaluated to determine the rate of stabilization of the landfill waste mass have historically been cellulose, lignin, C/L, pH, volatile solids and biochemical methane potential. Certain parameters such as volatile solids are very simple and low cost to analyze, and take very little time while other analyses such as biochemical methane potential are complex and require extensive time to obtain the result.

The objective of examining the relationships between these analyses is to determine if sufficient information can be gleaned from fewer and simpler methods that will allow an accurate determination to be made as to the stability status of any given landfill regardless of the operational method. This will allow for the simplest and most cost effective analyses to be performed on landfill waste. The evaluation will also help provide a sound basis for determining the end point for waste stabilization for both conventional and modified landfills.

2. Materials and methods

2.1. Landfills

Twelve landfills were sampled, and refuse was collected and shipped to Virginia Tech for analysis. For the Lake Mills and Maplewood landfills, no estimated age data was available. The landfills were operated primarily to receive municipal refuse, but industrial and commercial wastes including wastewater sludges were accepted at some of the landfills. Some of the variability in the results could be attributed to the variation in materials deposited in the landfills. However, since the goal of this study was to assess measurable parameters for operating landfills, waste variation is one of the considerations in evaluating the parameters. No gas data or leachate data was collected from the landfills because the study was undertaken after most of these landfills had been in operation for varying periods of time.

2.2. Sample collection

Municipal solid waste (MSW) samples were collected from 12 landfills beginning in the fall of 1999 to Spring 2003 using the sampling procedures following those traditionally used in the industry. A drill rig equipped with a 0.91 m (3 ft) bucket auger was used. Each location was sampled with the bucket auger in 3.05 m (10 ft) vertical sections with one representative composite sample prepared for each 3.05 m section. In order to obtain the composite sample, each time the auger was removed for cleaning, a shovel full (about 4.54 kg (10 lb)) of refuse was randomly collected from material discarded by the auger. The shovel full of waste was placed on a plastic container and each time the auger was removed another shovel full was obtained until the auger had progressed 3.05 m. Typically this was 15–20 samples of material. The material was then thoroughly mixed with the shovel and material from this pile was used to fill an 18.9 L (5 gal) bucket for shipping to Virginia Tech.

The initial 1.52 m (5 ft) of material was generally discarded because it contained a large amount of soil cover. A 4.54 kg (10 lb) sample was extracted from each full auger and placed in a roll-off box to collect a representative composite sample. The appearance of the waste was observed and recorded as the boring advanced. Each composite sample was thoroughly mixed while in the roll-off and a subsample was removed and placed in an appropriate plastic container for express shipment to the laboratory.

Age information was only available for 9 landfills that ranged from 0 to 11 years of age and was gathered from records kept for each individual landfill. Records indicated the location and date solid waste was placed in the landfill. The number of samples analyzed in this study (over 250) should allow for an accurate evaluation of the data collected from a variety of landfills at different stages of stabilization and being operated under several treatment scenarios.

Some limited lab-scale studies were also performed as part of this study. Solid waste from the Metro Landfill, Wisconsin (USA) was placed in 2.44 m × 0.46 m (8 ft × 18 in.) high-density polyethylene columns and exposed to various moisture, temperature, and leachate recirculating conditions to simulate landfill and bioreactor environments. The columns were heated using a hot water heater that recirculated water through plastic tubes that were wrapped around the columns and covered with insulating material. Leachate was recirculated using a sump pump in a bucket set below the columns. When the bucket became full enough to trigger the sump pump, water would be recirculated until the bucket was empty. Recirculation periods were variable, some returned flow several times a week and some approximately every 2 weeks. The moisture content was checked every month and if the moisture content dropped to 3% less than the desired amount, distilled water was added. The columns were operated for 18 months. Each column contained four sampling ports at equal intervals along the 2.44 m (8 ft) column length. Approximately, 100 g was removed from each of the four ports and a composite sample prepared for each column by thoroughly mixing the samples. The purpose of the lab study was to simulate and closely control landfill bioreactor conditions that are not always possible in full-scale landfills.

2.3. Analytical parameters

2.3.1. pH

A slurry was prepared in the field by adding approximately 250 mL of deionized water to 100 g of waste. The pH was also determined when samples arrived at the laboratory.

2.3.2. Moisture

The moisture content was determined by modified Standard Method 2540-B (APHA, 1992). At least 1 kg of as-collected solid waste was dried in an aluminum pan at 105 °C to a constant weight after cooling under desiccation. The moisture content is determined by weight loss from the original sample and expressed as a percent.

2.4. Laboratory sample preparation

Oven dried (105 °C) MSW was reduced to a particle size less than 0.3 cm in a bench top glass and stainless steel blender. Further grinding in a Wiley Mill with a 10-mesh screen achieved a powder-like consistency. Non-grindables such as rocks nails, etc., were removed by hand before processing. These were not considered in the analysis. Ground MSW was used for the volatile solids, cellulose, lignin, and biochemical methane potential (BMP) methods.

2.5. Volatile solids (VS)

The volatile solids procedure followed a modified version of Standard Methods APHA Method 2440-E. Samples were dried once again at 105 °C to a constant weight and held in a desiccator. Approximately, 2 g of dried MSW were placed in pre-weighed porcelain crucibles and inserted into a muffle furnace at 550 °C for 2 h. Samples were removed and allowed to cool in a desiccator to a constant weight. The percent weight loss on ignition yields the total amount of volatile matter.

2.6. Cellulose and lignin

The cellulose and lignin analysis followed ASTM E 1758-95^{e1}. A sample size of 300 mg of dry, milled MSW was used for this measurement. The cellulose was hydrolyzed into glucose monomers in two stages using sulfuric acid. The samples were digested in 3 mL of 72% sulfuric acid in a water bath at 45 °C for 2 h. Samples were then transferred to 250 mL septa bottles using 84 mL of nanopure water and autoclaved for 1 h at 121 °C and 15 psi. The samples were subsequently filtered using standard TSS glass fiber filters. The volatile suspended solids (combusted at 550 °C) remaining after hydrolysis were considered lignin. The filtrate was then neutralized using $CaCO_3$ powder directly. The glucose was quantified using a HPLC with a refractive index detector and HPX-87C carbohydrate column.

2.7. Biochemical methane potential

Biochemical methane potential (BMP) was modified from a procedure described by Owens and Chynoweth (1993) and later by Stinson and Ham (1995). Dry, shredded MSW (2 g) were added to a 250 mL Boston round septa bottle. Then, 100 mL of revised anaerobic media was added to each bottle. The media was made following these methods except for two modifications. The vitamin solution was not included, and anaerobic biosolids from the Peppers Ferry anaerobic digester, Fairlawn, Virginia (USA) were added as the inoculum (10% by volume). The bottles were incubated inverted for 45 days at 35 °C. At the end of the incubation period, 1-L Teflon gas sampling bags were connected to each bottle for 20 min while agitating the bottle to relieve excess pressure. A 100-μL sample was taken from the gas-sampling bag and injected into a GC with a carbosieve packed column and a flame ionization detector. The volume of gas in the gas-sampling bag was measured using a 60 mL plastic syringe. This test was run in triplicate with one blank for every six bottles. The amount of methane measured in the blanks was deducted from that of the samples. The BMP was reported as milliliters of methane per gram of dry MSW at STP (mL/g).

2.8. Plastics

All plastic material from each laboratory column sample was identified by sight, removed by hand, and weighed. Both the plastic and non-plastic portions of the samples were dried to determine moisture and then split into two equal parts by weight. One-half total dried weight of plastic and one-half total dried weight non-plastic for each sample were thoroughly mixed to give a representative sample of original column solid waste and the percent plastic material on a dry weight basis calculated. The remaining portion of non-plastic sample was also retained and properly labeled as containing no plastic.

3. Results and discussion

Plots of volatile solids, cellulose, BMP and lignin with age of the landfill for the samples are presented in Figs. 1 and 2. It can be seen that little degradation occurs over the first 4 years except for the samples from the Riverbend landfill. After 4 years, a steady decline occurs as indicated by VS, cellulose and BMP. Lignin also appears to decline for the "9 landfills" but at a much lower rate than for the other parameters. The VS, cellulose and BMP data for the Riverbend landfill shows a much faster degradation rate, but lignin did not decrease. Riverbend contains much more moisture than the other landfills, as indicated in Table 1, and this could lead to more rapid degradation of the waste in this landfill. However, since the initial refuse for this landfill was not available for characterization, it may be that the lower cellulose and BMP could be the result of lower levels in the initial waste. The variability in the data is high because each landfill has unique biodegradation environments, the waste is very heterogeneous, and moisture distribution can vary temporally and spatially. In addition, age data were provided by the landfill operators and data for a specific year typically reflects landfilling over a time period of up to a year.

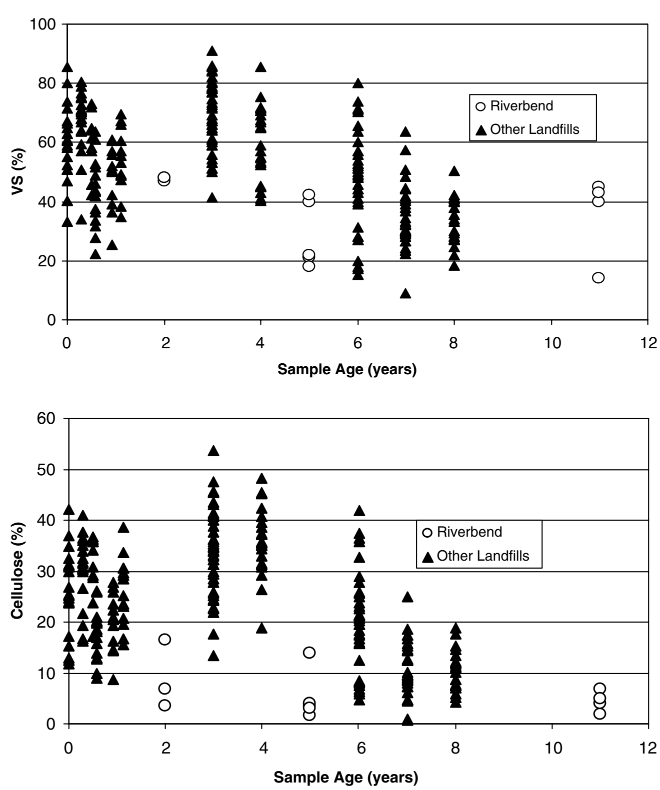

Fig. 1. Change in volatile solids (top) and cellulose (bottom) with landfill age for 10 landfills.

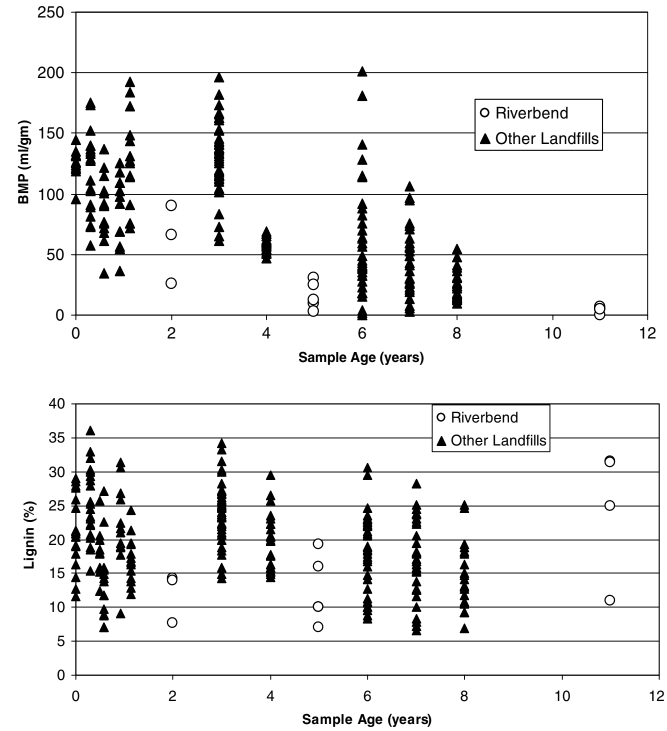

Fig. 2. Change in BMP (top) and lignin (bottom) with landfill age for 10 landfills.

Table 1
Moisture content for 12 landfills

Landfill	# Samples	% Moisture	Standard deviation
Atlantic	30	49.7	11.8
Central	21	33.5	7.4
Evergreen	29	29.6	8.7
Green Valley	7	33.9	7.0
King George	44	37.2	6.9
Lake Mills	30	41.7	11.9
Maplewood	29	38.3	7.8
Metro	74	31.0	6.4
Middle Peninsula	24	36.0	4.3
Plantation Oaks	25	31.7	9.2
Riverbend	12	48.3	13.6
Spruce Ridge	18	30.4	9.8

Samples with very low volatile solids are generally considered stable because they contain little organic material. Most of the VS in the low VS samples would be expected to consist of plastics and lignin. Due to the presence of plastics and lignin, the VS seldom decreased to less than 15%, even for aged, low cellulose and low BMP samples.

There is a substantial decrease in the percent cellulose with age for those landfills greater than 4 years old. Of interest are the samples from the Riverbend landfill in Oregon. As indicated in Table 1, most samples from this landfill had a moisture content around 50% so these data provide a reasonable idea of the benefits of a bioreactor landfill. For this landfill, most samples had a cellulose content less than 5% after 5 years in the landfill. Cellulose appears to be a more sensitive parameter than VS and this is due to the specificity of the measurement compared to VS, which includes biologically resistant organics. The Atlantic landfill also had a high moisture content and portions of this landfill were operated as a bioreactor landfill. All data for this landfill were approximately 3 years old. A comparison of the average values for bioreactor cells with the control or non-bioreactor cell is shown in Table 2. From these data, it can be seen that the samples from the bioreactor landfill are more degraded as evidenced by lower VS, cellulose, lignin and BMP in the bioreactor cells.

The biochemical methane potential analysis, although not an accepted standard method, measures the amount of material in MSW that can be converted to methane. The BMP is essentially the anaerobic equivalent to the BOD test used for wastewater characterization. The BMP should be an excellent indicator of refuse stability but is subject to variability based upon inoculum type (specific acclimated cultures versus anaerobic digester solids), container volume and variations in the sample amount used. The BMP test as performed in this study was designed to simulate the anaerobic landfill environment, yet be relatively simple to conduct. When BMP was plotted versus landfill age (Fig. 2), a similar trend to the response of cellulose with age was found. That is, after 3 years, the BMP began to decline and by year 8, was generally less than 50 mL/g. The samples from the Riverbend landfill were very low, less than 15 mL/g by year 11.

The values for lignin in the landfills ranged from 7% to 35%. The plot of lignin with age of the landfill (Fig. 2) suggests that lignin declines over time, except for the Riverbend samples. For the Riverbend samples, the lignin content increased with age and that is consistent with the expected recalcitrance of lignin and plastic. Historically, lignin has been thought to be either anaerobically nondegradable with no methane potential (Ham et al., 1993; Barlaz et al., 1990) or at best only slowly degradable (Eleazer et al., 1997). However, some literature indicates that lignin slowly degrades in anaerobic environments. Benner and Hodson (1985) found a significant, but low percentage (2–4%) of radiolabeled lignin was anaerobically degraded to methane and carbon dioxide during 60-day incubations at 55 °C. Approximately, the same percentage (2–4%) of lignin-labeled substrates was recovered as dissolved degradation products. In another study, Benner et al. (1984) found that 1.5–16.9% of radiolabeled lignin was anaerobically degraded to methane and carbon dioxide within 294 days.

These studies suggest that some portion of the lignin can be degraded in anaerobic environments similar to landfills, but variations in lignin degradation require further study.

Table 2
Comparison of Atlantic landfill data from bioreactor and control cells

	VS (%)	Cellulose (%)	Lignin (%)	BMP (mL/g)	Temp (°C)
Bioreactor cells	72.0	37.4	30.2	144	146
Control cell	66.5	30.0	25.3	128	95

One study indicated that lignin solubilization increases as the temperature increases (Örså and Holmbom, 1994). Therefore, solubilization of lignin could account for some of the losses observed in the data. The stability of lignin in the Riverbend landfill could be due to the high moisture content and frequent rainfall that could dissipate heat in this landfill. A comparison of data at a depth of 9.14 m (30 ft) for the Riverbend, Middle Peninsula, Spruce Ridge and Atlantic landfills showed average temperatures of 120 °F at Riverbend, 118 °F at Middle Peninsula and 125 °F at Spruce Ridge. For the Atlantic landfill, the temperature was 146 °F in the bioreactor cells and 95 °F in the control cell (Table 2).

Comparisons of cellulose, lignin, BMP and cellulose + lignin to volatile solids are shown in Fig. 3. The cellulose versus VS indicates a relatively strong correlation between these two parameters. The cellulose method is approved and accurate and the results should vary only with the sample source. The volatile solids measurement is a simple test but would be expected to vary more than cellulose due to the presence of non-biodegradable organics. The values of volatile solids in the samples ranged from 8% to 90% while cellulose values ranged from 2% to 52%. The slope of the linear fit to the data was approximately 2:1 and the intercept was near 10%. The intercept of 10% could indicate the presence of some non-cellulosic substances and could be assumed to be plastics and some lignin. The value of cellulose would appear to be reasonably predicted by VS so it could serve as a surrogate for cellulose as long as the limitations are recognized.

Using a *t*-test, data for individual landfills were compared with regard to the VS and cellulose relationship to determine if the individual landfills were also described by the relationship shown in Fig. 3(a). Of the 12 landfills, three did not have a significant correlation with the slope of the entire data set at the 90th percent level. The landfills that were not significantly correlated with the overall data were Maplewood, Riverbend and Spruce Ridge. Both Maplewood and Spruce Ridge were characterized by highly variable data, with low correlation coefficients. Riverbend, in addition to being the most degraded waste, is also a very small data set (12 data points).

The relationship between cellulose and volatile solids is also shown in Fig. 4 for the lab-scale columns, which show a strong correlation between cellulose and volatile solids. Data from the same samples are plotted in Fig. 4, but with the plastics removed. Plastics were determined by hand separation and weighing and comprise approximately 10% of the solid waste by weight as indicated by the difference between the two plots. These data indicate that the cellulose can be estimated from the amount of volatile solids minus plastic (approximately 10%) in a sample and suggest that much of the non-degradable material in the VS measurement is plastics.

The relationship between lignin and volatile solids is presented in Fig. 3(b). The relationship between these parameters is not as strong as for cellulose and volatile

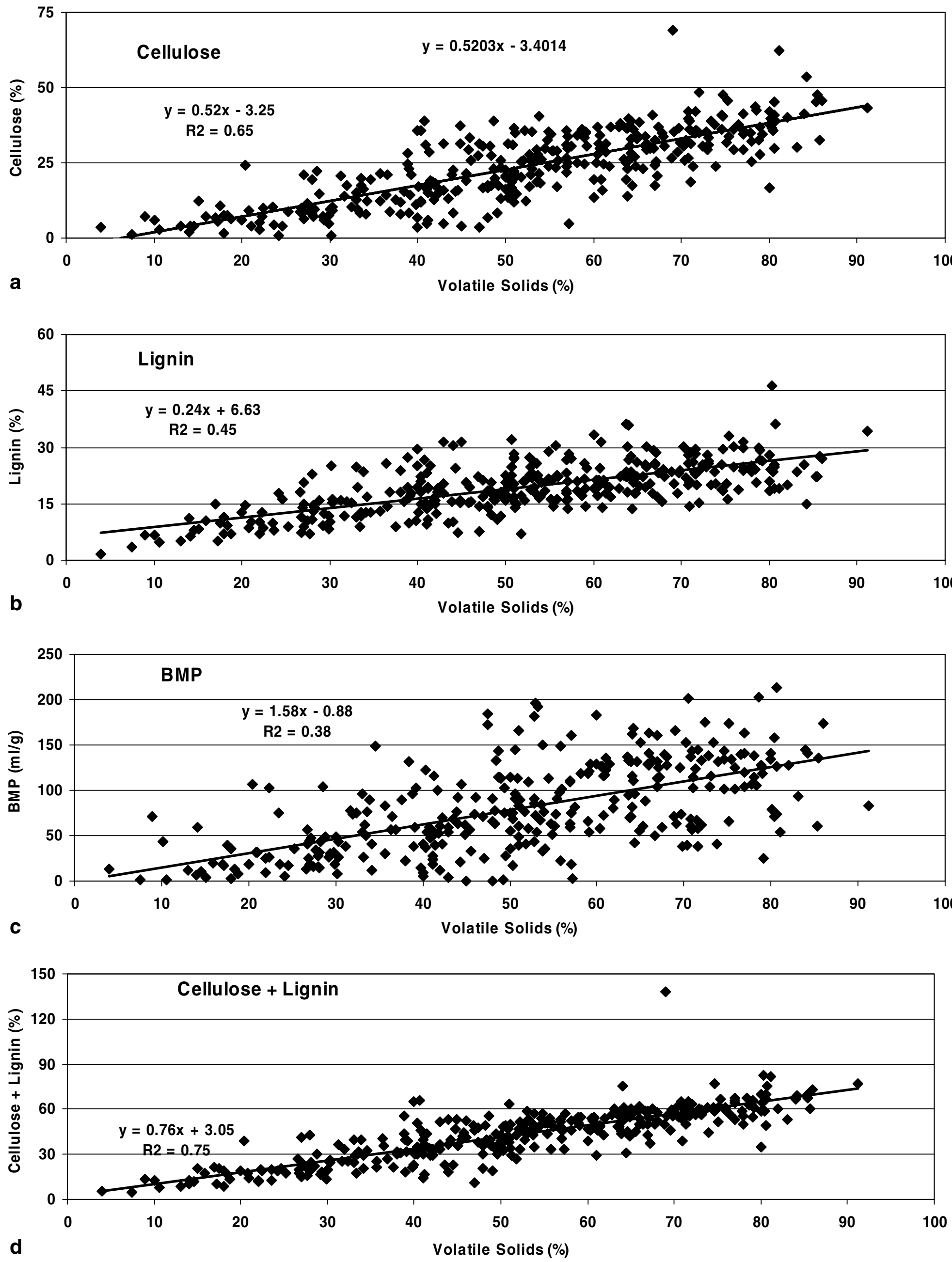

Fig. 3. Relationship between volatile solids and (a) cellulose, (b) lignin, (c) BMP and (d) cellulose + lignin using data from 12 landfills.

solids, but does suggest that lignin makes up a substantial portion of the volatile solids. A linear fit of the data passes through the y-axis at approximately 7%. In Fig. 3(c), it can be seen that the correlation of volatile solids with BMP is weak while the plot of cellulose + lignin versus VS (Fig. 3(d)) provides the best correlation for any of the data. The combined lignin + cellulose makes up about 85% of the VS. Although other organics are disposed in landfills, it is cellulose and lignin that comprise the bulk of the organic matter (especially since the lignin test includes plastics) and therefore, determines the stability of landfills.

The BMP values ranged from little or no methane production to nearly 200 mL/g dry sample. It would be expected that if the primary source of methane was from degradation of cellulose, the correlation between cellulose and BMP would be strong. However, the correlation between cellulose and BMP was weak with an r^2 of 0.32 (data not shown). In general, none of the parameters for characterization of refuse correlated well with BMP, suggesting that the variability in BMP limits its usefulness for characterizing the stability of refuse. Although the relationship between cellulose and BMP provided only a weak correlation, the patterns for both cellulose and BMP with age were very similar (Figs. 1 and 2), indicating that both cellulose and BMP will be low in well-degraded refuse.

The cellulose/lignin ratio has been used to infer the degree of biological degradation that has occurred in a landfill (Stinson and Ham, 1995). The variation in C/L

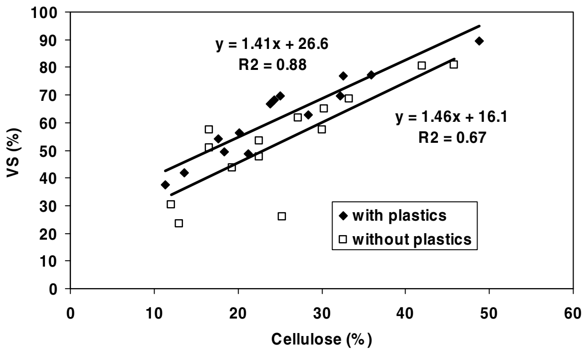

Fig. 4. Relationship between volatile solids and cellulose for test column samples with and without plastics.

with landfill age is shown in Fig. 5. The change in the C/L ration is similar to that for both cellulose and BMP, declining with age after 4 years. Cellulose and lignin are the principal components of wood and paper products and the disappearance of cellulose from MSW is considered to be a direct indicator of biological stabilization. The use of the cellulose/lignin ratio is based on the premise that cellulose is easily consumed in an anaerobic environment while lignin is not. The C/L ratio for fresh refuse has been shown to be variable because different types of paper and cardboard have highly varying C/L ratios. Cardboard has a high lignin content, while office paper is low in lignin. The C/L ratio would be high in fresh MSW and lower in older, more degraded waste. Reinhart and Townsend (1998) proposed a C/L of less than 0.2 as a definition of a stable waste. Using the Reinhart and Townsend criteria, the only wastes from this study that can be considered stable are the 11-year-old samples from Riverbend. However, the cellulose content indicates that Riverbend waste is relatively stable at year 5.

The volatile solids and cellulose measurements from this particular study appear to be the best parameters to determine the stability of municipal solid waste. These two parameters correlate very well with each other, with BMP, and with the age of the solid waste sample. The cellulose measurement is accepted and shows low variability. The VS measurement has higher variability than cellulose, but is so much easier to perform that this provides distinct advantages over other parameters. Since the VS measurement is affected by plastic, it may be advisable to remove all plastics before completing the test or subtract a standard value (perhaps 10%) from VS measures.

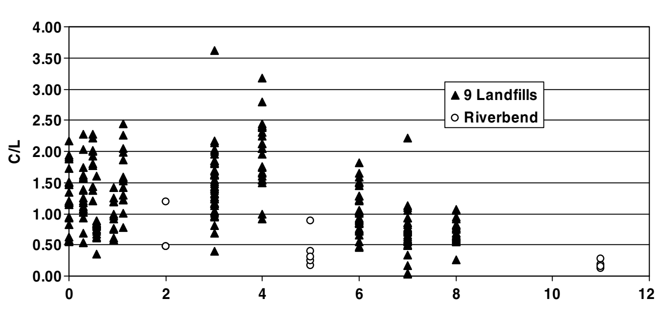

Fig. 5. Change in C/L with age for 10 landfills.

4. Conclusions

- Cellulose and volatile solids appear to be the best parameters for characterizing landfill waste bio-stability. Because the VS measurement is affected by plastic, it is advisable to account for plastics when using VS data for determining waste stability.
- Measured values for lignin also include plastics, unless these are specifically removed.
- Because the lignin measurement includes plastics and is highly variable, the use of the C/L ratio as an index of degradation has no advantage over cellulose or volatile solids.
- Biochemical methane potential (BMP) does not correlate well with cellulose or volatile solids, is a more expensive test and the results are more variable than cellulose. BMP may be best used in conjunction with cellulose or volatile solids to determine waste bio-stability.

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