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April 28, 1995

**MEMORANDUM**

**TO:** Michael Podolsky, Clare Lindsay, Brett Van Akkeren

**FROM:** Joanne Colt, William Driscoll, and Randy Freed

**SUBJECT:** Work Assignment 239, Task 2: Carbon Sequestration in Landfills

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This memorandum revises and refines the preliminary estimates of landfill carbon sequestration presented in ICF's January 12 memorandum (prepared under Work Assignment 210). It covers both mixed MSW and each of several materials present in MSW: food waste, three types of yard waste (grass, leaves, and branches), three types of paper waste (newspaper, office paper, and corrugated boxes), and three types of plastics (HDPE, LDPE, and PET). Our revised analysis indicates that the proportion of carbon sequestered in MSW landfills is considerably higher than estimated in our previous memorandum.

**BACKGROUND**

In our January 12 memorandum, we presented a "back-of-the envelope" calculation of the amount of landfilled carbon that enters long-term (i.e., hundreds of years) sedimentary storage. The analysis, which was based on a conceptual approach developed by Jean Bogner of Argonne National Laboratory and data derived from laboratory studies conducted in 1989 by Dr. Morton Barlaz of N.C. State, indicated that about 45 percent of the carbon placed in landfills does not degrade. We stated in the memorandum that the proportion sequestered might actually be considerably higher than 45 percent, and suggested performing a mass balance on landfilled carbon to make sure that our estimates of methane yields and sequestered carbon are consistent with one another.

In this memorandum, we take a different approach to estimating landfill carbon sequestration. Basically, we employ a mass balance approach that partitions the carbon placed into a landfill into each of its three major fates: bioconversion to methane, bioconversion to carbon dioxide, and long-term sequestration. Most of the inputs to the mass balance are derived from Dr. Barlaz's most recent (1994) laboratory studies<sup>1</sup> — the same laboratory studies that we have used, in previous memoranda, as a basis for estimating the methane yields of different materials in MSW. We discussed our analysis with both Dr. Barlaz and Kurt Spokas of Argonne National Laboratory (who works with Jean Bogner), and, although

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<sup>1</sup> Dr. Barlaz's work was funded by EPA's Air and Energy Engineering Research Laboratory under the supervision of Susan Thorneloe.

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they have not seen all of our detailed results, both researchers agree that the basic methodology is sound and the results are reasonable.

Our approach and results are discussed in detail below.

## APPROACH

### General Approach

Under this task, we estimated the amount of carbon sequestered when various materials in MSW are landfilled (and when mixed MSW is landfilled). To do so, we used the simplifying assumption that all carbon that enters a landfill is either (1) converted to methane or carbon dioxide (i.e., biogas), or (2) sequestered. This assumption is reasonable because the other fates of carbon entering a landfill are probably much less significant on a mass basis. These other fates include conversion to biomass (e.g., bacterial cells) and dissolution in leachate.<sup>2</sup>

We estimated the amount of carbon in each material in MSW as it enters the landfill, and the amount of carbon converted to methane or carbon dioxide as the material decomposes anaerobically. We then estimated the amount of carbon sequestered by calculating (1) the amount of carbon "in" minus (2) the amount of biogas carbon (i.e., CO<sub>2</sub> and CH<sub>4</sub>) "out." We first conducted this carbon balance for each carbon-containing material in MSW (e.g., food waste, newspapers). To obtain carbon balance estimates for mixed MSW, we used the values for carbon "in," carbon "out," and carbon sequestered for each material, combined with estimates of the proportion of each material in mixed MSW.

### Approach to Estimating the Amount of Carbon "In"

Our first step was to estimate the amount of carbon in each carbon-containing material in MSW as it enters a landfill. For food waste, yard waste, office paper, newspaper, and corrugated boxes, we used data from unpublished research by Dr. Barlaz.<sup>3</sup> We estimated the amount of carbon present in other carbon-containing materials (e.g., plastics, rubber, leather) using chemical formulas and data published by George Tchobanoglous et al.<sup>4</sup>

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<sup>2</sup>Because the results of our analysis indicate that landfilling may be much more favorable in GHG terms than previously thought, our findings may be controversial. Thus, some follow-up work to estimate carbon flows through these fates may be worthwhile.

<sup>3</sup> M. Barlaz, "Measurement of the Methane Potential of the Paper, Yard Waste, and Food Waste Components of Municipal Solid Waste," unpublished paper, Department of Civil Engineering, North Carolina State University, 1994.

<sup>4</sup> Tchobanoglous, George, Hilary Theisen, and Rolf Eliassen, *Solid Wastes: Engineering Principles and Management Issues* (New York: McGraw-Hill Book Company) 1977, p. 61.

Food Waste, Yard Waste, Office Paper, Newspaper, and Corrugated Boxes

Dr. Barlaz measured the amounts of several carbon-containing components (cellulose, hemicellulose, lignin, and total volatile solids) in samples of food waste, yard waste, paper waste, and mixed MSW. Dr. Barlaz's data are shown in Appendix A. The first column (column "a") shows the materials in MSW that Dr. Barlaz investigated. For each of these materials, Dr. Barlaz dried the material and analyzed the amount of cellulose, hemicellulose, lignin, and total volatile solids in one pound of dried material (protein measurements are from earlier work published by Dr. Barlaz<sup>5</sup>). These data are shown in columns "b" through "f" of Appendix A. We assume that the "total volatile solids" consist almost entirely of the four named components (cellulose, hemicellulose, lignin, and protein) plus all other carbon-containing components (e.g., waxes and tannins).<sup>6</sup>

We used Dr. Barlaz's data on the amount of cellulose, hemicellulose, etc., per pound of dry material (Appendix A) to estimate the amount of cellulose, hemicellulose, etc., per pound of wet material (Exhibit 1-A). (We converted to wet weight because MSW is typically measured by its wet weight, not its dry weight.) We did this based on the estimated solids content of each material, shown in column "b" of Exhibit 1-A. Our source for most of the data on solids content Tchobanoglous et al.<sup>7</sup> Dr. Barlaz provided us with the solids contents of grass, leaves, and branches in our telephone call earlier this week.

Columns "c" through "h" of Exhibit 1-A show the amounts of each carbon-containing component in one wet pound of each material. To determine the values for "other carbon-containing materials" in column "h," we simply subtracted (1) all of the measured carbon-containing components (cellulose, hemicellulose, lignin, and protein) from (2) the total volatile solids.

Note that Dr. Barlaz presented separate data for grass, leaves, and branches, rather than data for yard waste as a whole. We used Dr. Barlaz's data to estimate the values for yard waste, by assuming that yard waste is composed of 50 percent grass, 40 percent leaves, and 10 percent branches (all on a wet basis).

Finally, we converted the amounts of carbon-containing components per pound of wet material to the amount of carbon per pound of wet material. For cellulose, we used the chemical formula to determine that cellulose is 44.4 percent carbon (on a mass basis). Because there are various types of hemicellulose, we used a composite chemical formula to estimate that hemicellulose is 45.5 percent

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<sup>5</sup> Barlaz, Morton A. and Robert K. Ham, "The Use of Mass Balances for Calculation of the Methane Potential of Fresh and Anaerobically Decomposed Refuse," in *Proceedings from the GRCDA 13th Annual International Landfill Gas Symposium, March 27-29, 1990* (Silver Spring, MD: GRCDA -- The Association of Solid Waste Management Professionals) 1990, p. 235.

<sup>6</sup>The laboratory procedure for total volatile solids involves heating a sample to 550°C to determine the fraction of the solids that are driven off. This temperature is high enough to volatilize virtually all organic compounds but not high enough to volatilize most inorganics.

<sup>7</sup> Tchobanoglous et al, Op. cit., p. 57.

carbon. Our source for the chemical formulas for cellulose and hemicellulose was an EPA report.<sup>8</sup> For lignin, we used a carbon content of 63.8 percent, as reported in the "average elementary analysis of wood lignin" for coniferous species, in a chemical encyclopedia.<sup>9</sup> Because there are many types of protein, we used a carbon content of 53.8 percent from a composite composition for protein from a paper by Barlaz and Ham.<sup>10</sup> For "other carbon-containing components" (e.g., waxes and tannins), we used a value of 50 percent carbon, based on available data on the carbon content of waxes and tannins.<sup>11</sup>

### Plastics

Dr. Barlaz did not analyze the composition of plastics in his laboratory work. Therefore, for the three types of plastic, we used a different approach to determine the amount of carbon "in." For LDPE, HDPE, and PET plastics, we used the chemical formula for each type of plastic to determine the percentage of carbon per pound of plastic (on a dry basis), and then converted this to the pounds of carbon per wet pound of plastic. The resulting values are shown in columns "h," "m," and "n" of Exhibit 1-A (because plastics have a very low moisture content [about 2 percent] the dry-basis percentages are equal to the wet-basis percentages).

### **Approach to Estimating the Amount of Carbon "Out"**

To estimate the amount of carbon generated by each material in the form of biogas, we first determined the amount of carbon generated in the form of methane, and then estimated the amount generated in the form of carbon dioxide.

We started with Dr. Barlaz's unpublished data on the amount of methane generated by various materials when decomposed anaerobically under ideal conditions. These data (in units of milliliters of methane per dry gram of material, as given in Dr. Barlaz's paper) are provided in the last column (column "g") of Appendix A. In Exhibit 1-B, we convert these values to cubic feet of methane per pound of wet material (column "q") and then to pounds of carbon in the methane, per pound of wet material (column "r").

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<sup>8</sup> U.S. Environmental Protection Agency, *Estimate of Methane Emissions from U.S. Landfills* (Washington, D.C.: U.S. EPA) September 1994, p. 6.

<sup>9</sup> Kirk-Othmer, *Encyclopedia of Chemical Technology, Third Edition* (New York: John Wiley & Sons) 1981, Vol. 14, p. 298

<sup>10</sup> Barlaz, Morton A. and Robert K. Ham, "The Use of Mass Balances for Calculation of the Methane Potential of Fresh and Anaerobically Decomposed Refuse," in *Proceedings from the GRCDA 13th Annual International Landfill Gas Symposium, March 27-29, 1990* (Silver Spring, MD: GRCDA -- The Association of Solid Waste Management Professionals) 1990, p. 232.

<sup>11</sup> Different types of plant waxes have varying chemical compositions. We estimated the carbon content of Douglas fir bark wax at 59 percent, based on the components of the wax as reported in the Kirk-Othmer chemical encyclopedia cited above. Tannic acid has a carbon content of 54 percent. Thus, we believe that an estimated 50 percent carbon content for other carbon-containing compounds in materials in MSW is a conservative estimate, that will lead to a lower-bound estimate of the amount of carbon sequestered.

To estimate the amount of carbon released as carbon dioxide, we simply assumed that decomposition results in equal masses of methane-carbon and carbon dioxide-carbon (column "r" = column "s" of Exhibit 1-B). This is based on the stoichiometry of the anaerobic decomposition of carbohydrates and proteins, which generates equal moles of CO<sub>2</sub> and CH<sub>4</sub>. Our approach assumes that aerobic decomposition (which generates CO<sub>2</sub> but no CH<sub>4</sub>) plays a minimal role in the carbon balance.

The total amount of carbon released as biogas (methane plus carbon dioxide) is shown in column "t" of Exhibit 1-B.

To estimate the amount of carbon sequestered per wet pound of material landfilled, we subtracted (1) the amount of carbon released as biogas (the carbon "out") from (2) the amount of carbon "in." The results are shown in column "u" of Exhibit 1-B.

Exhibit 1-B also shows, in columns "v" and "w," the estimated percentage of carbon in each material that is released as biogas, and the percentage that is sequestered.

#### **Approach to Estimating the Carbon Balance for Mixed MSW**

Next we estimated the carbon balance for mixed MSW. To do so, we used (1) the carbon balance for each material in MSW (on a wet basis) from Exhibits 1-A and 1-B (supplemented by additional data on other carbon-containing materials in MSW), and (2) data from Franklin Associates on the composition of mixed MSW. Our spreadsheet analysis is shown in Exhibits 2-A and 2-B. The row labeled "Total" shows the values for mixed MSW, based on our calculations.

Exhibits 2-A and 2-B show additional rows for three materials (rubber, leather, and textiles) that were not considered in Exhibits 1-A and 1-B. Because rubber, leather, and textiles are carbon-containing materials in mixed MSW, we estimated the amount of carbon sequestered in these materials when mixed MSW is landfilled. For rubber and leather, we assumed that much more rubber than leather is discarded, and used the estimated carbon percentage (on a dry basis) for rubber from Tchobanoglous et al. For textiles, we again used data on carbon percentage (on a dry basis) from Tchobanoglous et al.<sup>12</sup> We assumed that all carbon in these materials, including textiles, would be sequestered. (For textiles, the petrochemical fibers such as polyester would be expected to be sequestered, but some natural fibers such as cotton might degrade.)

Because we did not have data on the composition of "other paper and paperboard" or of "wood," we estimated the carbon-containing components of these materials based on the composition of similar materials. We approximated the amount of each the carbon-containing component contained in "other paper and paperboard" by using the weighted averages of the amounts of each component in office paper, newsprint, and corrugated boxes. We used the composition of branches (on a dry basis) as the composition of wood (also on a dry basis).

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<sup>12</sup> Tchobanoglous et al, Op. cit.,p. 61.

We estimated the composition of an average wet pound of mixed MSW based on data on materials discarded in the municipal waste stream in 1993, developed by Franklin Associates.<sup>13</sup> The resulting values are shown in column "b" of Exhibit 2-A. The remaining columns of the spreadsheet were derived using the approach described above, in the discussion of Exhibit 1-A. Column "o" shows the total carbon "in" for the amount of each material contained in an average wet pound of mixed MSW.

Exhibit 2-B shows the amount of carbon "out" when a pound of mixed MSW (wet basis) is landfilled. We estimated methane generation based on the values in Exhibit 1-B, and the amounts of each material in one pound of mixed MSW; the resulting values are shown in column "q." (Note that no decomposition is expected for plastics, rubber and leather, and textiles over a timeframe of hundreds of years; thus the methane generation for these materials is assumed to be zero.) As before, the methane generation values are converted to methane carbon in column "r," and the remainder of the spreadsheet was derived as described above in the discussion of Exhibit 1-B.

The bottom row of Exhibits 2-A and 2-B shows the carbon balance based on Dr. Barlaz's analysis of the methane generated by anaerobic decomposition of mixed MSW under ideal conditions (Dr. Barlaz's data for mixed MSW are shown in the bottom row of Appendix A). Dr. Barlaz's data are empirical data, but are based on mixed MSW in a single truckload of refuse. In contrast, our estimate for the carbon balance for mixed MSW is based on the national average composition of mixed MSW, as estimated by Franklin Associates.

## RESULTS

The amount of carbon sequestered when one pound of each material (wet basis) is landfilled is shown in column "u" of Exhibit 1-B. The percentages of carbon sequestered for each material are shown in the last column of the exhibit. These percentages range from 30 percent for food waste to more than 80 percent for newsprint, leaves, and branches. We estimate carbon sequestration of 100 percent for plastic.

The amount of carbon sequestered when one pound of mixed MSW (wet basis) is landfilled is shown at the bottom of column "u" of Exhibit 2-B. Again, the percentages of carbon sequestered are shown in the last column of the exhibit. The percentages for each material are the same as in Exhibit 1-A. The percentage for mixed MSW is shown in the "Total" row: 81.4 percent. The bottom row shows similar results based on data collected by Dr. Barlaz on methane generation from one pound of mixed MSW: 74.5 percent. Although Dr. Barlaz's results are based on a single sample, and our estimate is based on aggregation of dozens of individual data points, each with some component of error, there is remarkable agreement between our "bottom-up" estimate of biogas generation and carbon sequestration for mixed MSW and his "top-down" measurement for mixed MSW.

Both of the results are consistent with estimates of carbon sequestration presented in a paper authored by Jean Bogner and Kurt Spokas of Argonne National Laboratory.<sup>14</sup> Bogner and Spokas

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<sup>13</sup> U.S. EPA, Office of Solid Waste and Emergency Response, *Characterization of Municipal Solid Waste in the United States: 1994 Update*, November 1994, pp. 31, 62, and 71.

<sup>14</sup> Bogner, J., and K. Spokas. 1993. "Landfill CH<sub>4</sub>: Rates, Fates, and Role in the Global Carbon Cycle." *Chemosphere*, Vol. 26, Nos. 1-4, pp. 369-386.

conducted original laboratory research on biogas production during the decomposition of mixed refuse and compared their results to others presented in the literature, including earlier work by Dr. Barlaz. The authors concluded that "...in general, more than 75 percent of the carbon deposited in landfills remains in sedimentary storage." The authors believe that the percentage sequestered may be even higher in field conditions that are not conducive to biodegradation.

## CAVEATS AND LIMITATIONS

This section discusses the major caveats and limitations associated with the analysis.

Probably the most important caveat is that the analysis is based on only one set of laboratory experiments. While researchers other than Dr. Barlaz have conducted laboratory studies tracking the degradation of MSW, Dr. Barlaz is the only researcher we have been able to identify who has tested different materials individually. Among the people we have spoken to over the past few months, Dr. Barlaz is recognized as the expert on the degradation of different fractions of MSW under anaerobic conditions. Moreover, as discussed below, his findings with respect to the methane potential of mixed MSW are well within the range used by landfill gas developers.

Both Dr. Barlaz and Kurt Spokas made the point that Dr. Barlaz's work was conducted under "ideal" conditions favoring biogas production. As a result, one would expect the carbon balances derived from Dr. Barlaz's work to overestimate the amount of biogas that would be generated in the field, and therefore to underestimate the amount of carbon sequestered in landfills. The extent to which this is true is unclear. Dr. Barlaz's observed methane yield for mixed refuse was about 1 ft<sup>3</sup>/lb, which is actually toward the lower end of the range used by landfill gas developers (0.8 - 1.8 ft<sup>3</sup>/lb). This indicates that Dr. Barlaz probably has not systematically overestimated methane yields, and that the carbon balances based on his work therefore do not systematically underestimate carbon sequestration in landfills. However, it is also likely that the 0.8 - 1.8 ft<sup>3</sup>/lb range used by landfill gas developers overestimates methane generation for certain landfills, particularly those located in arid climates or designed to be kept dry (i.e., in accordance with the recent revisions to the Subtitle D criteria). The bottom line is that the carbon balances presented in this memorandum probably underestimate carbon sequestration for dry landfills, and might overestimate carbon sequestration for landfills located in climates that favor biogas production.

It is also important to mention that when Dr. Barlaz conducted a carbon mass balance for his laboratory experiments, he was unable to account for 100 percent of the carbon. The carbon recovery calculations presented in his paper reflect the extent to which measured losses in cellulose and hemicellulose are consistent with the amount of methane actually produced. Carbon recovery was 87.5% for mixed refuse and ranged from 75.1% to 98.3% for the different materials. Dr. Barlaz believes that the "missing" carbon was mostly in the form of biogas and is currently recalculating methane yields under that assumption. The effect will be to slightly increase biogas yields, which would — in our analysis — result in slightly decreased sequestration estimates.

As mentioned earlier, our analysis ignores the aerobic phase of degradation that occurs directly after organic wastes are placed in a landfill. We asked both Kurt Spokas and Dr. Barlaz whether they believed this to be a significant limitation, and they both stated that it is not, because the vast majority of organic carbon placed in landfills degrades under anaerobic conditions.

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Finally, our spreadsheet analysis is subject to limitations introduced by the assumptions that were made at various steps in the analysis, as described in the "approach" section of this memorandum.

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We look forward to hearing your comments on this analysis. Please call Joanne at (703) 934-3284 with questions or comments.

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