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A Classification Methodology for Landfill Leachates

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1 **A Classification Methodology for Landfill Leachates**

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26 **Abstract**

27 A characterization scheme based on landfill leachate chemical signatures could support
28 studies of leachate evolution over time, liner performance, and help confirm or disprove potential
29 leachate contamination of groundwater. Wide variations in single constituents across time, sites,
30 and site practices, and inconsistencies related to common bivariate measures suggest a robust,
31 multivariate analysis could be useful. A variant Stiff diagram approach (a subjective analytical
32 comparison of soluble salts) has been developed, and supports graphical depictions of multiple
33 samples. The hypothesis is that leachates with similar chemistry form clusters, and this was
34 tested using a data set of 652 samples from 26 distinct liner systems collected from a Long Island
35 (New York, USA) landfill over more than 20 years. Most (75%) of diagrams were classified into
36 three general leachate groupings that associated with the kinds of wastes received in the
37 particular landfill module (90% if "early" leachate results are not considered).

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40 **Subject headings:** sanitary landfills, monitoring, liners, chemical compounds, waste
41 management

42

43 **Introduction**

44 Reliable, meaningful landfill leachate contaminant signatures could support analyses of
45 landfill processes, and help determine liner performance and identify groundwater contamination
46 sources, depending on the signature relationship to landfill sections, waste types, or leachate age.
47 Univariate characterizations (e.g., ammonia, COD, or chloride concentrations), while often
48 useful, necessarily oversimplify leachate variability (Majone et al. 1998; Marttinen et al. 2002),
49 and are subject to large concentration ranges (Kjeldsen and Christopherson 2001). Bivariate
50 comparisons, especially BOD:COD ratios, have been used to classify leachate types (beginning
51 with Chian and DeWalle 1976); some find these measures inconsistent (Lo 1996; Armstrong and
52 Rowe 1999; Statom et al. 2004), and limited to descriptions of waste stability (Kjeldsen et al.
53 2002).

54 Stiff diagrams (Stiff 1951) subjectively describe ionic solutions, and have been used in
55 investigations of landfill-related groundwater contamination (beginning with Kimmel and Braids
56 1980). I have developed a multivariate analysis that modifies traditional Stiff diagrams, and
57 applies principal component analysis (PCA) to the resulting diagram components. This creates a
58 unique analytical approach that supports grouping of leachate results by major ion chemistry,
59 which appears to be a function of waste type and age.

60 **Setting**

61 The Town of Brookhaven landfill on Long Island, New York has operated since 1976.
62 All of its cells have liner systems with leachate collection. Cell 5 (1995-2003) and Cell 6
63 (2003+) were constructed in phases with “double composite” liner systems. Wastes received
64 have varied, affected mostly by a ban on landfilling of MSW (enacted in 1983, effective 1990,
65 fully enforced 1995). Landfilling post-1995 has been restricted to construction and demolition

66 debris ("C&D"), "car fluff," waste-to-energy incinerator ash, and some other materials classified
67 by New York State as either "products of resource recovery" or "inert" (Tonjes and Swanson
68 1994). Disposal rates have increased from ~250,000 tonnes yr⁻¹ (1976) to 1.1 million tonnes yr⁻¹
69 (including wastes used as cover).

70 **Materials and Methods**

71 **Sampling and Analysis**

72 Unfiltered leachate samples are collected either from manholes or taps on pump lines.
73 Samples were collected irregularly until 1993 when semi-annual sampling was established.
74 Samples are analyzed by contract laboratories holding the highest New York state certifications
75 (at that time). Beginning in 1993, analytes included "leachate indicators" (nutrients, salts, and
76 general water quality indicators), metals, volatile organic compounds (VOCs), semi-volatile
77 organic compounds, herbicides, pesticides, and dioxins and furans. Prior to 1993, samples
78 typically were analyzed for leachate indicators, and sometimes metals and VOCs.

79 **Data Analysis**

80 Stiff diagrams (Stiff, 1951) were drawn using Grapher (Golden Software, Golden, CO) to
81 determine similarities and differences among the sample results. Stiff diagrams depict major
82 cations and anions from aqueous samples, measured in meq. The cations (Na⁺+K⁺, Ca²⁺, Mg²⁺,
83 NH₄⁺) and anions (HCO₃⁻, SO₄⁻², Cl⁻, NO₃⁻) selected here follow mid-1980s USGS practices
84 (e.g., Wexler 1988). These Stiff diagrams were modified, however, using a normalization
85 process based on the greatest value for any diagram parameter, so that diagrams are bounded by
86 values of ~1 (using two significant figures, meaning maximum values range 0.95-1.05) (Tonjes
87 et al. 1995), with the intention that concentration differences do not obscure diagram shape
88 similarity. Diagram shapes were classified (Table 1) using exemplars of primary leachate types.

89 Figure 1 is an example of the signature shape associated with landfilled MSW, illustrating a
90 shape archetype found for 65 other leachate samples.

91 PCA using correlational matrices was applied to confirm diagram classifications
92 (Statistix 9, Analytical Software, Tallahassee, FL). The value of the PCA is its potential for
93 identification of similar diagram shapes, insofar as each diagram vertex is in a somewhat similar
94 position relative to the same parameter vertex in other diagrams. PCA can collapse
95 dimensionality, and so mapping the transformed data points into a 2-dimensional PCA graph
96 approximates distance relations in a theoretical 8-dimensional space -- similar shapes should plot
97 close together. The percent of variance explained is the conservation of overall distance
98 relations.

99 **Results**

100 Samples tended to be classified consistently, so that nearly 75% (482 of 652) were
101 identified as a signature pattern or similar to one. Liner systems eventually tended to be
102 classified consistently: 9 of 26 systems had “mature” results classified into only one of the
103 signature patterns. Four of 16 remaining systems had only 1 or 2 mature samples that did not
104 share a single signature pattern. The liner systems where only inert materials (mostly C&D) were
105 landfilled at first and then ash was added (Cell 5 Phases 4, 5, 8 and 9 and Cell 6) tended to first
106 be classified with the C&D pattern, and then the ash pattern. Only one-third (54 of 170) of
107 “other” patterns occurred after an identification of a signature leachate pattern (a “mature” liner
108 system), and 16 had relatively low concentrations (<100 meq. for a sum of the 9 Stiff diagram
109 parameters), lower than associated with the signature patterns. Thus, once a signature pattern was
110 detected, 93% of following results were also a signature pattern (except where contamination
111 decreased below signature pattern levels).

112 A single graph of all 652 results in the PCA is difficult to interpret (Figure 2). Although
113 Figure 2 is not a random scatterplot, and results concentrate in certain areas and are sparse in
114 others, no clear clusters emerge. However, mapping the signature classifications alone (Figure 3
115 illustrates the MSW signature set) appears to justify the subjective results groups. The MSW-
116 signature results in Figure 3 create a fairly tight cluster in the lower left-hand corner of the plot,
117 and a halo of other results, described as varying from signature plot in one characteristic
118 description, is associated with the cluster. When similar isolations are made of the other two
119 signature shapes, the C&D signature, not as precisely defined as the other two, is more diffuse,
120 and overlaps considerably with the ash classifications (as it should, as many characteristics are
121 the same in Table 1). “Other” diagrams almost all map away from the signature centroids. The
122 two-dimensional PCA accounts for nearly 60% of the variance.

123 **Discussion**

124 The most widely referenced classification process for leachates, BOD:COD ratios,
125 reflects the overall state of biological degradation with a low BOD:COD ratio (<0.4) defining
126 “mature” or “aged” leachate in this accounting (Kjeldsen et al. 2002). Molecular weights of
127 organic compounds has also been used to define similar classes (Calace et al. 2001). Chian and
128 DeWalle (1976) defined bivariate ratio sets to create leachate groupings, using broad bands
129 instead of linear trends due to large variabilities and changes in ratios with landfill age, creating
130 classes of leachate sorted by age. Gibbons et al. (1999) created a discriminant function to
131 separate VOCs results for MSW, hazardous waste, and co-disposal facilities. Isotopes have been
132 used to differentiate leachate contamination from other impacts on groundwater (beginning with
133 Baedecker and Back 1979) but although Bogner et al. (1996) used deuterium and stable C

134 isotopes to understand methane formation in fills, there has not been any use of isotopes to
135 distinguish leachate types to date.

136 Kylefors (2003) analyzed all data from 5 rounds (one year) of leachate sampling using
137 PCA, which echoes the approach used here. She did not normalize her parameters. Three
138 samples grouped nicely, and two others did not. Others (such as Barker et al. 1988) have used
139 PCA as a means of discerning leachate impacts from other contaminant sources, with a variety of
140 successes and failures.

141 Here, I found patterns in leachate quality despite its chemical complexity and variability,
142 and the changes in leachate quality were associated with differing waste streams for different
143 landfill systems. Although Stiff diagram analyses have always depended upon subjective
144 comparisons, use of principal component analysis and normalized diagrams can make the
145 process more quantitative and objective, and provide better justifications for determinations of
146 diagram shape similarity.

147 The diagram shapes and their clustering in the PCA depiction has been previously used to
148 track groundwater contamination and to identify contamination sources (Tonjes et al. 1995).
149 Close analysis of changes in diagram shapes may be useful in determining relationships between
150 dissolution (and other chemical reactions) and decomposition, and changes that occur with aging
151 of the landfilled material. Differences in diagram shapes and scaling factors appear to distinguish
152 primary and secondary liner chemistries and concentrations, in many cases, which holds promise
153 for understanding liner performance.

154 **Conclusions**

155 More than 600 leachate samples collected over 20 years from 26 different liner systems
156 were analyzed using a Stiff diagram variant, and further compared using principal component

157 analysis. These data were parsed so that 75% of all results were classified into three general
158 categories; once a landfill system leachate was classified as belonging to one group, 90% of
159 subsequent samples also fell into the same groupings. Thus, this method sorted monitoring data
160 samples into like and unlike sets. The data are generated from types of chemical analyses that
161 can be accurately and consistently accomplished with less technical requirements than some
162 other approaches, and the data processing is not very difficult to accomplish. Although
163 BOD:COD ratios are especially useful in determining treatment options (Renou et al. 2008), the
164 signature pattern approach clearly has more utility to track contamination in groundwater. The
165 subjectivity of signature pattern identification is offset by validation associated with waste
166 inputs, and clustering in PCA diagrams. Use of PCA can be daunting, but growing use of
167 statistical packages makes this feasible to consider.

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222

223 Table 1. Leachate Classifications

Classification	Diagram Characteristics
MSW Signature	$\text{Na}^+ + \text{K}^+, \text{HCO}_3^- \sim 1$ $\text{Cl}^-, \text{NH}_4^+$ major constituents (but $< \text{HCO}_3^-, \text{Na}^+ + \text{K}^+$) $\text{Ca}^{+2} \sim \text{Mg}^{+2}, \ll \text{Na}^+ + \text{K}^+$ $\text{NO}_3^-, \text{SO}_4^{-2} \sim 0$
Ash Signature	$\text{Cl}^- \sim 1$ $\text{Ca}^{+2} \sim \text{Na}^+ + \text{K}^+$, major constituents but < 1 (usually $\ll 1$) $\text{Mg}^{+2}, \text{NH}_4^+, \text{HCO}_3^-, \text{NO}_3^-, \text{SO}_4^{-2} \sim 0$
C&D Signature	$\text{Cl}^- \sim 1$ $\text{Na}^+ + \text{K}^+ > \text{Ca}^{+2}$ but < 1 $\text{NH}_4^+, \text{HCO}_3^-$ noticeably > 0 $\text{Mg}^{+2}, \text{NO}_3^-, \text{SO}_4^{-2} \sim 0$
MSW-similar	One characteristic rule not followed (completely) but diagram appears similar
Ash-similar	One characteristic rule not followed (completely) but diagram appears similar
C&D similar	One characteristic rule not followed (completely) but diagram appears similar
Other	Distinct from the 3 major signature shapes

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226 List of Figures

227 Figure 1. MSW Stiff diagram example; the y-axis indicates the relative scaling factor for each
228 cation and anion. The maximum strength of any one ion was 82 meq.

229 Figure 2. Scatterplot of the first two axes of the principal component analysis of the 8 modified
230 Stiff diagram parameters from 652 leachate samples from the Brookhaven landfill

231 Figure 3. Clustering of the 66 MSW (◆) and 179 MSW-similar diagrams (◇) from the
232 scatterplot of the first two axes of the principal component analysis of the 8 modified Stiff
233 diagram parameters from 652 leachate samples from the Brookhaven landfill (see Figure 2)