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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

A characterization scheme based on landfill leachate chemical signatures could support 27 studies of leachate evolution over time, liner performance, and help confirm or disprove potential 28 leachate contamination of groundwater. Wide variations in single constituents across time, sites, 29 and site practices, and inconsistencies related to common bivariate measures suggest a robust, 30 31 multivariate analysis could be useful. A variant Stiff diagram approach (a subjective analytical comparison of soluble salts) has been developed, and supports graphical depictions of multiple 32 samples. The hypothesis is that leachates with similar chemistry form clusters, and this was 33 34 tested using a data set of 652 samples from 26 distinct liner systems collected from a Long Island (New York, USA) landfill over more than 20 years. Most (75%) of diagrams were classified into 35 three general leachate groupings that associated with the kinds of wastes received in the 36 particular landfill module (90% if "early" leachate results are not considered). 37 38

39

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

43 Introduction

Reliable, meaningful landfill leachate contaminant signatures could support analyses of 44 landfill processes, and help determine liner performance and identify groundwater contamination 45 sources, depending on the signature relationship to landfill sections, waste types, or leachate age. 46 Univariate characterizations (e.g., ammonia, COD, or chloride concentrations), while often 47 48 useful, necessarily oversimplify leachate variability (Majone et al. 1998; Marttinen et al. 2002), and are subject to large concentration ranges (Kjeldsen and Christopherson 2001). Bivariate 49 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. 2002). 53

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

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

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

70 Materials and Methods

71 Sampling and Analysis

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

79 **Data Analysis**

Stiff diagrams (Stiff, 1951) were drawn using Grapher (Golden Software, Golden, CO) to 80 determine similarities and differences among the sample results. Stiff diagrams depict major 81 cations and anions from aqueous samples, measured in meq. The cations (Na^++K^+ , Ca^{2+} , Mg^{2+} , 82 NH4⁺) and anions (HCO₃⁻, SO₄⁻², Cl⁻, NO₃⁻) selected here follow mid-1980s USGS practices 83 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 values of ~1 (using two significant figures, meaning maximum values range 0.95-1.05) (Tonjes 86 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.

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

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

99 **Results**

Samples tended to be classified consistently, so that nearly 75% (482 of 652) were 100 101 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 signature patterns. Four of 16 remaining systems had only 1 or 2 mature samples that did not 103 share a single signature pattern. The liner systems where only inert materials (mostly C&D) were 104 105 landfilled at first and then ash was added (Cell 5 Phases 4, 5, 8 and 9 and Cell 6) tended to first be classified with the C&D pattern, and then the ash pattern. Only one-third (54 of 170) of 106 107 "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).

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

123 Discussion

The most widely referenced classification process for leachates, BOD:COD ratios, 124 125 reflects the overall state of biological degradation with a low BOD:COD ratio (<0.4) defining "mature" or "aged" leachate in this accounting (Kjeldsen et al. 2002). Molecular weights of 126 organic compounds has also been used to define similar classes (Calace et al. 2001). Chian and 127 128 DeWalle (1976) defined bivariate ratio sets to create leachate groupings, using broad bands instead of linear trends due to large variabilities and changes in ratios with landfill age, creating 129 130 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

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

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

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

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

154 **Conclusions**

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

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

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223 Table 1. Leachate Classifications

Classification	Diagram Characteristics
MSW Signature	$Na^{+}+K^{+}, HCO_{3}^{-} \sim 1$
-	Cl ⁻ , NH ₄ ⁺ major constituents (but $<$ HCO ₃ ⁻ , Na ⁺ +K ⁺)
	$Ca^{+2} \sim Mg^{+2}, << Na^{+}+K^{+}$
	$NO3^{-}, SO_{4}^{-2} \sim 0$
Ash Signature	Cl ⁻ ~1
	$Ca^{+2} \sim Na^{+}+K^{+}$, major constituents but <1 (usually <<1)
	Mg^{+2} , NH_4^+ , $HCO_3^- NO3^-$, $SO_4^{-2} \sim 0$
C&D Signature	Cl ⁻ ~1
	$Na^{+}+K^{+}>Ca^{+2}$ but <1
	NH_4^+ , HCO_3^- noticeably >0
	Mg^{+2} , NO3 ⁻ , SO4 ⁻² ~ 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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