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Composition and variability of leachate from recent and aged areas within a municipal landfill

Nancy Ragle, John Kissel, Jerry E. Ongerth, Foppe B. DeWalle

ABSTRACT: A leachate study was conducted at a large, operating, regional municipal solid waste (MSW) landfill near Seattle, Washington, to examine differences in composition and emission rates between old and new areas of the fill. The landfill began operation in 1966, was receiving approximately 2000 tpd of MSW and had $30 \times 10^6 \text{ m}^3$ ($\sim 12 \times 10^6$ tonnes) waste in place at the time of this study. The two areas studied had average ages of 3.7 and 16 years, and contained 5.7×10^6 and 2.04×10^6 tonnes of MSW, respectively. Variations in flow rate, total dissolved solids (TDS), chemical oxygen demand (COD), total organic carbon (TOC), Fe, and Mn were monitored over a 3-month period in the winter and spring of 1992. Increases in flow driven by precipitation caused gradually increasing leachate mass emissions from the aged fill. The rate of mass emission increase with increasing flow from the new fill was more than three times higher than that from the old fill. Leachate flow through the old fill appeared more channelized, resulting in diluting effects with increasing percolation. In leachate from the new fill concentrations were essentially independent of flow. Overall, mass emissions per unit waste mass in place decreased with increasing waste age for TDS and Mn, indicating that these components were leachable independent of degradation processes. Mass emissions per unit waste mass in place increased with increasing waste age for COD, TOC, and Fe, which typify components that increase in availability for leaching with increasing age and progressing stage of decomposition. *Water Environ. Res.*, 67, 238 (1995).

KEYWORDS: composition, emissions, landfill, leachate, solid waste.

Assessment of present and future solid waste landfill design requirements and contaminant impacts on adjacent surface and ground waters requires accurate characterization of the quality and quantity of leachate produced (Tchobanoglous, 1993). Previous studies at a variety of landfills and historic data from the study site taken at widely spaced intervals show wide variations in individual leachate component concentrations [King Co. Solid Waste Div. (KCSWD), 1989]. Although the literature suggests that leachate quality changes as a landfill ages (Chian and DeWalle, 1976), evidence of a clear distinction between leachates from areas of different ages within a single landfill was not apparent in historical data from the site studied here (KCSWD, 1992). Whether such variability is due to random or to systematic factors is at present not adequately defined.

The principal objective of this study was to determine if, in a municipal solid waste (MSW) landfill, the composition of leachate from areas of aged decomposed waste differs from that of leachate from areas of recently deposited waste. The study was designed to investigate the short-term variability of leachate quality at a single municipal landfill and to determine whether leachate from fill areas of different ages could be distinguished.

Prior to data collection to test for age dependence of leachate composition, definition of the variability in leachate components over short-to-long time scales was essential. Initially, continuous monitoring of specific conductance was used to assess composition variations over the shortest time scale. Modest variations in this period led to subsequent hourly and then daily sampling in periods of dry weather and of rainfall. During the main sampling period, chemical oxygen demand (COD), total organic carbon (TOC), and total recoverable Fe and Mn, in addition to specific conductance, total dissolved solids (TDS), and flow rate, were monitored for 22 days at hourly, daily, and weekly intervals.

Methods

Sampling sites. Samples were collected at two sampling sites: site 1 was at a flow monitoring station (FS 3, Figure 1) at the downstream end of the collection system draining the old waste area [referred to subsequently as area 1 (Figure 1, "Main Hill")]; site 2 was at a manhole (MH46N, Figure 1) at the downstream collection point for the new waste area [referred to subsequently as area 2 (Figure 1, "Central Pit")].

Sample collection. Iron, manganese, COD, and TOC, and specific conductance were analyzed in samples collected at hourly intervals for 5 to 6 hours per day twice weekly, April 1 through April 22, 1992. Metal samples were collected in 250 mL high-density polyethylene (HDPE) bottles containing 2 mL of low trace metal grade concentrated nitric acid and were stored at room temperature until laboratory analysis. Organic samples were collected in 250-mL amber glass bottles containing 2 mL of 9 N reagent grade sulfuric acid and were immediately placed in a cooler of approximately 7°C, then transferred to a refrigerator of the same approximate temperature. The average pH of the COD and TOC samples was 4 before analysis.

Flow determination. At site 1, flow data were collected by timing the opening and closing of a valve that cycled after 200 gal had entered the manhole. Each flow value represents an average over the 200-gal cycle. At site 2, flow was measured using the Parshall flume permanently installed in the manhole.

Rainfall data were obtained from a Met-1 model 099R Tipping Bucket Rain Gauge operated routinely by landfill technicians.

Sample analysis. Analytical methods used in the study conformed to previously described standard methods (USEPA, 1986) as described briefly below.

Specific conductance was measured using a multichannel Solomat MPM 4000 Matrix Processor and 4007 Chemistry Module with a 1.0 K cell encompassing four carbon graphite ring electrodes in an epoxy body, with a 25-ft cable encased in a polyethylene jacket. Temperature compensation was provided in-

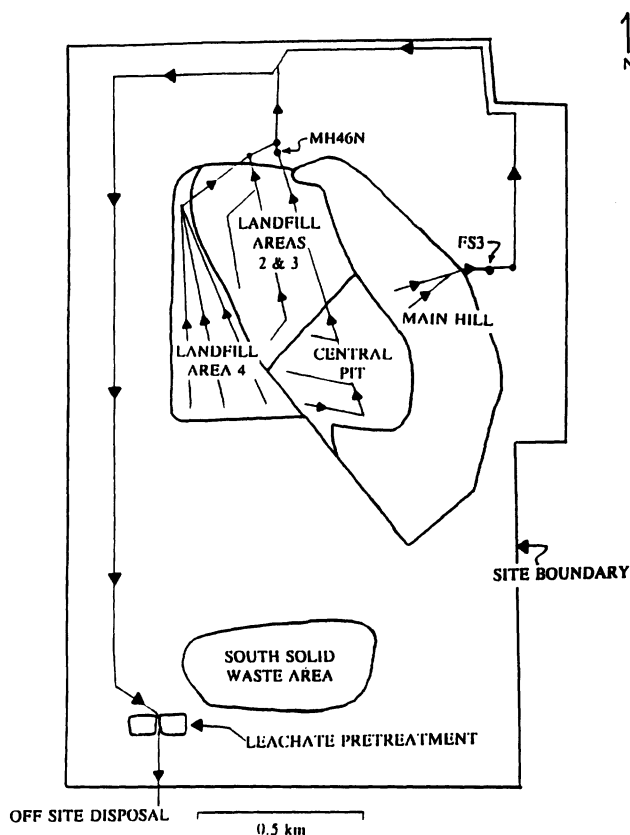


Figure 1—Site map.

ternally, and the instrument was standardized using potassium chloride solutions prepared with analytical grade KCl and deionized water from a Barnstead NANOpure ultrapure water system. The probe was cleaned with reagent grade acetone between measurements.

COD and TOC analyses were carried out by a professional laboratory. For COD, the analysis followed EPA method 410.4, which entails an oxidation of the sample using potassium dichromate, sulfuric acid, and mercuric sulfate catalyst. After 2 hours at 150°C and then cooling, absorbance is measured using a MR-401 spectrophotometer at 620 nm to gauge the amount of chromic ion formed during the redox reaction. The TOC analysis followed EPA method 415.1, which involves an oxidation of the reduced organic carbon using ultraviolet radiation and persulfate. The evolved CO₂ gas is then measured by a non-dispersive infrared detector.

Metals analysis was carried out by the author following EPA method 3005 with slight modifications. After mixing, 50 mL of the sample was transferred to a 100-mL Griffin beaker, 1 mL trace metal grade concentrated nitric acid and 2.5 mL analytical grade concentrated hydrochloric acid were added. The samples were covered with watch glasses and heated on a hot plate at approximately 95 to 100°C until the volume decreased to approximately 10 mL. After cooling, the samples were quantitatively transferred to 50-mL volumetric flasks and brought to volume using deionized water. Blanks, spiked blanks, primary standards, and laboratory duplicates were prepared in the same manner. Before spectroscopy, sample dilutions and dilution duplicates of 1:9, 1:49, and 1:99 were prepared, as necessary, using

a 1% nitric acid solution, which also served as the instrument blank.

Atomic absorption spectroscopy was performed using a Perkin-Elmer model 5100 instrument, with a Perkin-Elmer Intensitron Hollow Cathode lamp for manganese, and a Hamamatsu Photonics K.K. Hollow Cathode lamp for iron. For Mn, the wavelength used was 279.5 nm with a slit width of 0.2 nm. For Fe, the wavelength was 248.3 nm with the same slit width. For both Fe and Mn, the instrument was calibrated with 1.0, 2.0, 5.0, and 10.0 ppm standards using a nonlinear calibration method.

Results

Study site description. The work described here was conducted at a large regional landfill in central western Washington. The location is in the foothills of the Cascade mountains where the annual precipitation was 137 cm/year. The landfill has been in operation since 1966 and was currently receiving approximately 2000 tons/day of MSW. The construction of the landfill and its environmental control systems permit dividing it into subunits that function independently.

Two subunits were selected for this study (Figure 1). Area 1, the old area, had a surface of 21.8 ha with approximately 2.04×10^6 T waste in place. Area 2, the new area, had a surface area of 41.3 ha with approximately 5.7×10^6 T waste in place. The areas are contiguous but the old one is no longer active. Initial waste placement in the old area began in 1966, resulting in an average age of 16 years. It is unlined, but covered with a sophisticated clay and membrane system. The new area is in current operation. It contains waste having an average age of 3.7 years. It is constructed with an advanced clay and synthetic membrane liner and has an integral leachate and gas collection system. Because it is in active use, it does not have an impervious cover in place. At the time of this study, filling had been completed in the new area and it had been graded and covered with a well-compacted low-permeability soil cover, including a surface drainage system. The final synthetic membrane cover was added after conclusion of the sampling period.

Site hydrology. Rainfall totaling 36 cm occurred on the study areas during the January-to-April study period interspersed with periods of dry weather ranging from 1 to 5 days. By trial and error comparison, the time lags between rainfall and leachate appearance giving the highest correlation coefficient were 4 days for the old area ($R^2 = 0.736$) and 30 days for the new site ($R^2 = 0.81$).

Leachate composition. Records of leachate composition in the 4 years before this study showed higher values in the older fill than in the newer (Table 1). The COD/TOC ratios were approximately 4.6 for both old and new areas. The COD/TDS ratios were 1.6 for both old and new areas, using TDS values derived from the conductivity measurements. The Fe/Mn ratio for the old fill was as high as 23 and only 7 for the new area. Comparison of these historic data with those collected during this study showed slightly lower median and maximum values in the current period. The concentrations measured in leachate from the old area were generally higher than from the new area, based on comparison between samples taken from 1988 to 1991 and during this study, the spring of 1992. The median flow from the new fill was more than three times higher than from the old fill, corresponding approximately to the 2.8 times higher tonnage-in-place ratio and the 2 times higher surface area ratio. This was

Table 1—Leachate characteristics during study and preceding period.

Parameter	Old site		New site	
	Median	Range (SD)	Median	Range (SD)
Preceding period				
COD (mg/L)	9 100	1 200–75 000	7 050	220–21 600
TOC (mg/L)	2 000	79–81 000	1 500	31–6 800
TDS (mg/L)	5 460	840–12 600	4 730	858–10 120
Fe (mg/L)	350	140–600	200	90–680
Mn (mg/L)	15	5–27	28	15–28
Flow (l/hr)	—	95–4 604	—	0–14 509
Study period				
COD (mg/L)	6 544	2 538–11 117 (1820)	3 277	2 795–4 166 (376)
TOC (mg/L)	1 956	605–3 429 (563)	867	794–1 046 (71)
TDS (mg/L)	4 449	2 268–6 668 (1 217)	6 113	5 310–6 415 (459)
Fe (mg/L)	266	93–543 (86)	85	76–98 (6)
Mn (mg/L)	9.1	4.4–18.0 (3.1)	14.2	12.7–15.4 (0.9)
Flow (l/hr)	1 094	506–3 406 (762)	3 409	295–4 178 (1 359)

reflected in greater dilution and lower concentrations in leachate from the new fill.

Variability. The hourly variations in composition of leachate from the old fill (Figure 2), indicated by data from April 10, 1992, showed a flow decrease to 63% of the initial value during a 5-hour time period and an increase of 47% in COD and of 74% in iron from the initial values. The corresponding increases for Mn and TDS were 53 and 20%, respectively. Leachate from the new area (Figure 3) showed less variability compared to that of the old area. On April 7, during which leachate flow was nearly constant, an increase of 30% in COD and of 3% in iron was observed.

Analytical precision for each parameter as indicated by the coefficient of variation ranged from 0.29 to 2.9% (Table 3). As indicated by continuous conductivity measurements little minute to minute variation in composition was observed. This was corroborated by observed variation in each of the five leachate components measured between field duplicates taken a few minutes apart. For the old area maximum percent variation between duplicates was greatest for COD, 6.1%, (Table 3) and

least for Mn, 0.22%. For the new area maximum percent variation between duplicates was also greatest for COD, 24%, (Table 3) and also least for Mn, 0.72%. For each parameter at both areas, the maximum hour-to-hour variation in concentrations was comparable to day-to-day variations (Table 3). Maximum hourly and daily variations for each parameter at the old area were at least four times those observed at the new area. At the old area highest hourly and daily variations were observed in TOC and Fe, >200%, the lowest in TDS, 50%. At the new area highest hourly and daily variations were observed in COD and Fe, 30 to 50%, the lowest in Mn, 6%.

Relation between leachate components. In old fill leachate strong associations were observed between the concentration of different leachate components as indicated by the correlation coefficients, with R^2 values ranging from 0.85 to 0.99 (Table 4). New fill leachate showed high correlations only between TOC, conductivity, and manganese. For old leachate the wide range of concentrations in each of the parameters compared with that of new fill leachate contributed to the higher correlation coefficients. The COD/TOC ratio from the regression slope was 3.2

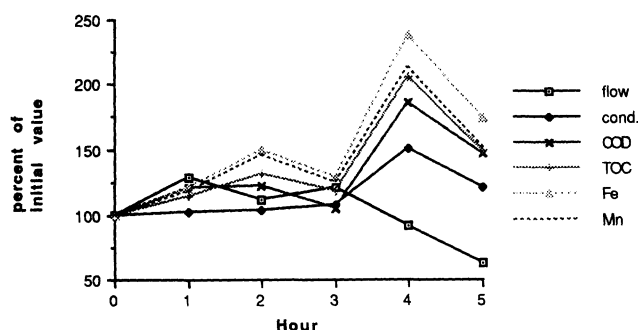
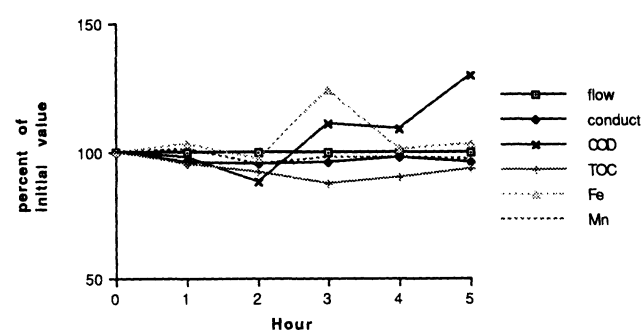
**Figure 2—Hourly leachate at old site.****Figure 3—Hourly leachate at new site.**

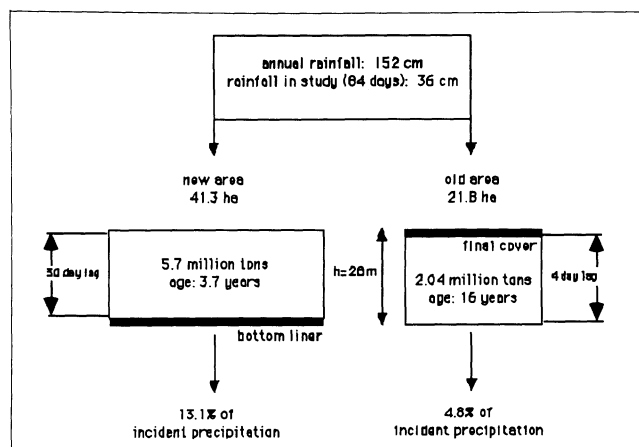
Table 2—Leachate characteristics and their flow dependency.

Ordinate	Abscissa	Linear relation	R ²	Site
TOC (g/hr)	Flow (L/hr)	$y = 1\,714 + 0.13x$	0.017	Old
		$y = 1\,042 + 0.61x$	0.887	New
TDS (g/hr)	Flow (L/hr)	$y = 3\,395 + 1.2x$	0.797	Old
		$y = 14\,444 + 5.3x$	0.965	New
COD (g/hr)	Flow (L/hr)	$y = 5\,593 + 0.6x$	0.040	Old
		$y = -1\,786 + 3.9x$	0.756	New
Fe (g/hr)	Flow (L/hr)	$y = 240 + 4.3e^{-2x}$	0.257	Old
		$y = 2.7 + 1.3e^{-2x}$	0.986	New
Mn (g/hr)	Flow (L/hr)	$y = 6.9 + 2.8e^{-3x}$	0.648	Old
		$y = 2.7 + 1.3e^{-2x}$	0.986	New

(Table 4) for old leachate and 3.3 for new leachate. The COD/conductivity ratio was 1.6 in the old leachate and 0.65 in new leachate indicating that the new leachate contained almost three times as much ions as the old leachate at a given organic strength. The COD/Fe ratio in the new leachate was almost twice as high as in the old leachate possibly resulting from the greater availability and greater oxidation of the metals in the old fill.

Mass emission rates. In the old fill leachate parameter concentrations decreased rapidly with increasing flow. The new fill leachate concentrations were almost independent of flow. At high flow rates concentrations of COD, TOC, and Fe were similar in leachate from both sites. Under similar high flow conditions conductivity and Mn were lower in the old fill leachate. Mass emission rates increased only slightly with increasing flowrate for the old fill and more rapidly for the new fill (Table 2). Most parameters showed a three to six times greater rate of increase with increasing flow in leachate from the new fill compared to the old fill.

For leachate of comparable strength from both areas the major difference between them was higher salt content and lower iron concentration from the new area. Also a more rapid increase of the mass emission was observed with increasing flow rates in the new area. The median concentrations in old leachate were

**Figure 4—Characteristics of old and new sites.**

twice as high as in new leachate, whereas the flowrate was three times lower.

Discussion

Examination of data presented above describing leachate characteristics from old and new areas of the landfill permits synthesis of a conceptual model relating concentrations, flow, and fill conditions (Figure 4). The smaller old area with a final synthetic membrane cover produced less leachate (1/3) of greater strength (2×). The leachate produced was approximately 5% of incident rainfall. The 4-day percolation lag time of the old area was very short compared with the 30-day lag of the new area. Given the observed rapid response to rainfall and the dilution effect observed in the concentration-flow relationships (Table 2), the short lag of the old fill was apparently a reflection of a relatively high degree of channelization within the fill accompanied by increased flow through the established channels. The lower strength leachate produced in the new area appeared after a 30-day lag and constituted approximately 13% of incident rainfall. Together with the observed increasing mass emission with increasing flow, these factors suggest that as an increasing

Table 3—Minimum and maximum % variation over an hour and a day as defined by [(high - low)/low]100.

Parameter	% Variation over a day		% Variation over an hour		% Variation of field duplicates		% coefficient of variation lab duplicates	
	Min	Max	Min	Max	Min	Max	Min	Max
Old site								
COD (mg/L)	14.6	182	0	183	4.9	6.1	0.27	1.6
TOC (mg/L)	10.4	241	0	241	0.16	0.37	0.18	0.67
TDS (mg/L)	3.2	50.7	2.8	40	—	—	0.25	2.9
Fe (mg/L)	18.5	268	0.1	267	0.56	2.8	1.3	1.3
Mn (mg/L)	8.5	149	0.09	149	0.1	0.22	0.29	0.29
New site								
COD (mg/L)	2.1	48	0.56	46	8.5	24.0	see above	
TOC (mg/L)	4.0	15	0	13	1.4	15.0		
TDS (mg/L)	1.3	13	0	11	—	—		
Fe (mg/L)	2.9	28	0	28	0.24	2.7		
Mn (mg/L)	1.3	6.3	0.17	6	0.67	0.72		

Table 4—Correlation coefficients (R^2) and linear relations between different leachate parameters ($y = a + bx$).

R^2	(COD)		(TOC)		(Cond.)		(Fe)		(Mn)	
	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)
COD	—	—	0.95	0.07	0.85	0.09	0.91	0	0.94	0.07
TOC	—	—	—	—	0.87	0.88	0.96	0.27	0.99	0.80
Cond.	—	—	—	—	—	—	0.87	0.48	0.87	0.71
Fe	—	—	—	—	—	—	—	—	0.98	0.46
Mn	—	—	—	—	—	—	—	—	—	—

Coefficient <i>b</i> (Ordinate) (y)	Abcissa (x)									
	COD		TOC		Cond.		Fe		Mn	
	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)
COD	—	—	3.2	3.0	1.6	0.6	21.2	38.7	664	25
TOC	—	—	—	—	0.5	0.2	6.6	6.1	207	81
Cond.	—	—	—	—	—	—	13.2	51.0	417	379
Fe	—	—	—	—	—	—	—	—	31.6	4.1
Mn	—	—	—	—	—	—	—	—	—	—

(1) old site, (2) new site

amount of rainfall enters the fill it flushes an increasing volume of fill rather than remaining channelized leading to dilution as in the old fill.

When leachate mass emissions were computed per ton of waste in place it was apparent that the concentrations of COD, TOC, and Fe increased from the 3.7 year average age new fill to the 16 year average age old fill. Compared on the same basis, con-

centrations of TDS and Mn decreased over the time span of the new to old fill (Figure 5). Comparison suggests that the increasing trend over this time span in components of the first group was due to their increasing availability for leaching through decomposition of the waste in place, whereas components of the second group were relatively available as placed in the fill and were gradually flushed out over time. The patterns observed (Figure 4) were consistent with those described previously (Chian and DeWalle, 1976).

Conclusions

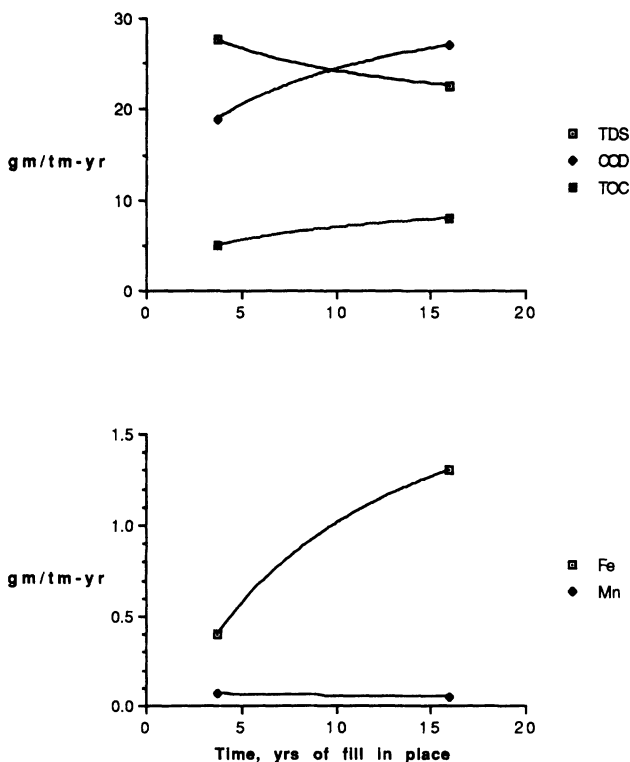
Observations on variations in leachate composition over time scales ranging from minutes to days indicated that minute-to-minute variations in gross organic and inorganic composition parameters were small, <10%. Hour-to-hour and day-to-day variations in the same gross composition parameters were large, that is, two to three times.

Observations on variations in leachate composition with variations in leachate flow indicated that mass emission from areas of old fill increased slowly with increasing flow in comparison to the more rapid, that is, three to six times, increase from areas of new fill.

Comparison of mass emissions per unit waste mass in place indicated that leachate composition parameters could be divided into at least two groups. One group included inorganic parameters TDS and Mn, which exhibited high initial availability and a decrease in emission rate over years of flushing. A second group includes the decomposition-dependent parameters COD, TOC, and Fe, which appeared at relatively low rates initially but increased over a period of at least 16 years as waste components degrade producing leachable products.

Acknowledgment

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**Figure 5—Leachate characteristics.**

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