

Emerging Contaminants at a Closed and an Operating Landfill in Oklahoma

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Abstract

Landfills are the final depositories for a wide range of solid waste from both residential and commercial sources, and therefore have the potential to produce leachate containing many organic compounds found in consumer products such as pharmaceuticals, plasticizers, disinfectants, cleaning agents, fire retardants, flavorings, and preservatives, known as emerging contaminants (ECs). Landfill leachate was sampled from landfill cells of three different age ranges from two landfills in Central Oklahoma. Samples were collected from an old cell containing solid waste greater than 25 years old, an intermediate age cell with solid waste between 16 and 3 years old, and operating cell with solid waste less than 5 years old to investigate the chemical variability and persistence of selected ECs in landfill leachate of differing age sources. Twenty-eight of 69 analyzed ECs were detected in one or more samples from the three leachate sources. Detected ECs ranged in concentration from 0.11 to 114 $\mu\text{g/L}$ and included 4 fecal and plant sterols, 13 household/industrial, 7 hydrocarbon, and 4 pesticide compounds. Four ECs were solely detected in the oldest leachate sample, two ECs were solely detected in the intermediate leachate sample, and no ECs were solely detected in the youngest leachate sample. Eleven ECs were commonly detected in all three leachate samples and are an indication of the contents of solid waste deposited over several decades and the relative resistance of some ECs to natural attenuation processes in and near landfills.

Introduction

There are 90,000 to 100,000 closed municipal landfills and about 3100 operating landfills (Zero Waste America 2011) in the United States. The closed landfills, many of which are unlined and poorly capped, may be sources of a large number of organic compounds known as emerging contaminants (ECs) to surrounding groundwater and surface water. ECs consist of household and industrial compounds in wastes and consumer products that include fecal and plant sterols, pharmaceuticals, food additives, soaps and detergents, solvents, cleaning agents, fire retardants, plasticizers, perfumes, and pesticides. ECs, although they generally occur in small concentrations in water ($<1 \text{ mg/L}$), may singly or in aggregate cause health problems for humans and wildlife ingesting water containing these compounds.

In 2008, approximately 135 million tons of municipal solid waste (MSW) was deposited in landfills in the United States, making landfilling the most common method of MSW disposal (U.S. Environmental Protection Agency 2009). Landfills are the final depositories for a large number of anthropogenic organic compounds, including

ECs (Slack et al. 2005). Pharmaceutical compounds may occur in concentrations of approximately 8.1 mg/kg in typical MSW (Musson and Townsend 2009). Having been discarded in landfills, ECs may be degraded/metabolized, adsorbed to solids, or dissolved in leachate (Musson and Townsend 2009). Anaerobic conditions in landfills and nearby groundwater receiving organic-rich leachate from landfills (Cozzarelli et al. 2011) are likely to slow metabolism/breakdown of organic compounds in leachate and groundwater compared to aerobic conditions that are more common in shallow groundwater (Bedient et al. 1997). In groundwater downgradient from an abandoned unlined landfill near Elkhart, Indiana, detergent metabolites, plasticizers, disinfectants, fire retardants, pharmaceuticals, and an antioxidant were detectable at concentrations in the low parts-per-billion range (Buszka et al. 2009). Huset et al. (2011) reported on detection of 24 fluorochemicals in landfill leachates, primarily short-chain (C4-C7) carboxylates or sulfonates associated with paper, textiles, and carpets.

Leaching of organic chemicals from both old and modern landfills to groundwater and surface water is a potentially important environmental problem, with such chemicals potentially being toxic, estrogenic, and carcinogenic to both terrestrial and aquatic organisms (Cozzarelli et al. 2011; Huset et al. 2011; Matejczyk et al. 2011). Some reports have indicated that mixtures of dilute concentrations of ECs in water may deleteriously affect human health, as had been

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previously shown for aquatic biota (Sharpe and Skakkebaek 1993; Lye et al. 1999; Pomati et al. 2006; Tillitt et al. 2010).

Many landfills constructed and operated in the United States prior to the 1990s were not designed to contain leachate, having no or insufficient liners, faulty caps, and insufficient or no leachate-collection systems due to the lack of regulations requiring those features at the time of operation of those landfills. Thus, tens of thousands of abandoned landfills may contribute ECs to adjoining groundwater and surface water, as has been reported in a few studies (Barnes et al. 2004; Buszka et al. 2009; Eggen et al. 2010). Most modern landfills have leachate-collection and pumping systems that collect leachate into lined holding pits or lagoons. However, leaks through liners, caps, and leachate-collection systems may allow some leachate to escape from containment systems into the surrounding environment. Furthermore, leachate collected in containment systems typically is drained into sewer lines that discharge to municipal waste water treatment plants. Municipal waste water treatment plants may not be designed to remove some ECs and varying proportions of ECs could be discharged with treated waste water back into the receiving streams (Ternes 1998; Snyder et al. 2007).

To investigate chemical changes and persistence of ECs in landfill leachate with landfill age, this paper reports on the occurrence and distribution of selected ECs in leachate from landfill cells of three different ages: (1) an old landfill cell containing solid waste greater than 25 years old, (2) an intermediate age landfill cell with solid waste between 16 and 3 years old, and (3) an operating landfill cell with solid waste less than 5 years old. Comparisons of ECs in leachate from landfill cells containing MSW of differing ages may provide information about leaching and degradation of ECs in landfill leachate with time and changes in MSW content with time, although the lack of control of MSW content in sampled landfill cells can complicate such analysis. Such data can be compared to the occurrence of ECs in leachate from landfills in other parts of the world, such as those reported by Buszka et al. (2009), Musson and Townsend (2009), Huset et al. (2011), and Matejczyk et al. (2011). Because analyses of EC contents of MSW at the time of burial were not available, this paper does not describe a controlled experiment, but rather represents a reconnaissance sampling of leachate and groundwater from a closed landfill and leachate from an operating landfill to gather information about possible changes in leachate composition with time and transport of ECs in groundwater.

Materials and Methods

Description of Field Sites

Landfill 1 (Figures 1 and 2) is a closed municipal landfill that served about 100,000 residents from the cities of Norman, Moore, Noble, and Town Hall Park in Central Oklahoma from the early 1900s until 1985 (Dixon and Popoola 1992). No liners or leachate-collection systems were installed at Landfill 1 (Christenson et al. 1999), which was typical prior to the passage of regulations in Subtitle D of the Resource Conservation and Recovery Act (RCRA) in the 1990s. The MSW in this landfill is primarily residential and commercial in origin, but small amounts of household hazardous wastes also may have been discarded in this landfill. This landfill



Figure 1. Map showing the locations of Landfills 1 and 2. Landfill 2 is an active landfill, one of four major landfills operating in the Oklahoma City metropolitan area, which has a population of approximately 560,000 residents (U.S. Census Bureau 2010). Landfill 2 is equipped with a liner and leachate-collection system. The MSW deposited in this landfill is non-hazardous and is from residential and commercial sources. Two cells were sampled at Landfill 2—one that had been closed for 3 years, and a cell that had been operating for 5 years and was being filled with MSW.

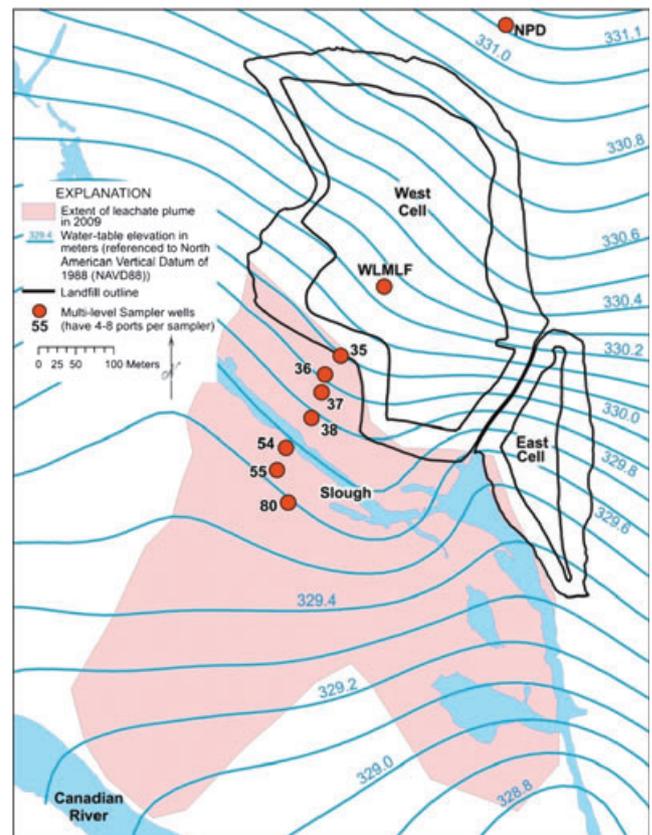


Figure 2. Map showing well locations and water-table elevations of Landfill 1 (Cozzarelli et al. 2011).

was constructed in sandy alluvium of the Canadian River and covered with a vegetated clay cap in the mid-1980s. Landfill 1 has been the site of extensive research by the U.S. Geological Survey (USGS) (<http://ok.water.usgs.gov/projects/norlan/index.html>), U.S. Environmental Protection Agency, and numerous universities (Christenson et al. 2003).

Sampling

To evaluate differences in landfill-leachate composition, one monitoring well was sampled at Landfill 1 in February 2009 (water-level monitoring landfill [WLMLF] well). The WLMLF well was screened between 12.63 and 14.15 m below land surface in Landfill 1, which had been closed for more than 25 years (oldest age cell). Two leachate-collection drains were sampled at Landfill 2 in February 2009. Sample 2-S was collected from a leachate-collection drain in a capped landfill cell that received MSW from 1993 to 2006 (intermediate age cell). Sample 2-N was collected from a leachate-collection drain in a landfill cell that was not yet capped, and had received MSW from 2005 to 2009 (youngest age cell).

To evaluate changes in leachate composition with transport in an alluvial aquifer, three monitoring wells were sampled at Landfill 1 in February 2009 (WLMLF, 35-5, and 38-6), with wells 35-5 and 38-6 being sampled in September 2000 (as described by Barnes et al. 2004). Wells 35-5 and 38-6 were parts of multilevel wells shown in Figure 2, screened between 5.52 and 5.64 m below land surface and 6.95 and 7.07 m below land surface, respectively. Those wells were selected for sampling because they are screened in the landfill, near the toe of the landfill, and downgradient near the center of the leachate plume, respectively, enabling qualitative evaluation of fate and transport of ECs in groundwater at the oldest landfill sampled for this paper. Well 35-5 is 100 m downgradient from well WLMLF and near the toe of the landfill. Well 38-6 is about 90 m further downgradient from well 35-5 (Figure 2). These wells were positioned on a transect parallel to the local groundwater flowpath determined from aquifer properties and contaminant plume morphology (Lucius and Bisdorf 1995; Scholl and Christenson 1998).

Groundwater and leachate samples were filtered in the field using a pre-cleaned aluminum plate-filter assembly with a glass-fiber filter having a 0.7-mm nominal pore size, prebaked at 450 °C. The filtered samples were collected in 1-L baked amber glass bottles sealed with Teflon™-lined caps, immediately chilled on ice upon collection, and shipped overnight to USGS laboratories, as described in USGS sampling protocols (Wilde et al. 1998a and 1998b).

Sample Analyses

In water samples collected at the two landfills in February 2009, concentrations of 69 selected ECs (Table 1) and percent recoveries of three surrogate isomers of ECs were analyzed by solid-phase extraction (SPE), capillary-column gas chromatography/mass spectrometry (GC/MS), or high-performance liquid chromatography and mass spectrometry (HPLC/MS) (Zaugg et al. 2006; Furlong et al. 2008) at the USGS National Water Quality Laboratory in Denver, Colorado. For water samples collected in September 2000,

Analyzed Emerging Contaminant	Ranges of Reporting Limits (September 2000)	Ranges of Reporting Limits (February 2009)
Acetaminophen	0.00900	—
Acetophenone	0.150	0.650
Acetyl-hexamethyl-tetrahydro-naphthalene	—	0.500
Anthracene	0.0500	0.0400
Anthraquinone	—	0.160
3-Beta-coprostanol	0.00500–0.600	2.00
Beta-sitosterol	—	2.00
Benzophenone	—	0.220
Benzo[a]pyrene	0.0700	0.0800
Bis(2-ethylhexyl) phthalate	2.50	—
Butylated hydroxyl toluene	0.0800	—
Bromacil	—	1.00–11.2
Bromoform	—	0.100
Butalbital	—	0.400
Caffeine	0.0140–0.0800	0.100–0.300
Camphor	—	0.0440
Carbaryl	0.0600	0.610–0.730
Carbodox	0.100	—
Carbazole	—	0.0400–0.320
Cholesterol	0.00500–1.50	2.00
Chlorpheniramine	—	0.0400
Chlorpyrifos	0.0200	0.120
Chlortetracycline	0.0500	—
Cimetidine	0.00700	—
Ciprofloxacin	0.0400	—
Cis-chlordane	0.0400	—
Codeine	0.100–0.240	0.400
Cotinine	0.0230–0.0800	0.800
4-Cumylphenol	—	0.100–0.300
Dehydronifedipine	0.0100	—
Diazepam	—	0.0400
Diazinon	0.0300	0.0800
1,4-Dichlorobenzene	0.0300	0.0400
Dieldrin	0.0800	—
Diethoxynonyl phenol	—	1.48–10.0
Diethylphthalate	0.250	—
Digoxigenin	0.00800	—
2,6-Dimethylnaphthalene	—	0.120
Diltiazem	0.0120	—

Table 1 Continued

Analyzed Emerging Contaminant	Ranges of Reporting Limits (September 2000)	Ranges of Reporting Limits (February 2009)
1,7-Dimethylxanthine	0.0180	—
2,6-Di-tert-butylphenol	0.0800	—
2,6,D-tert-butyl-1,4-benzoquinone	0.500	—
d-Limonene	—	1.75
Doxycycline	0.100	—
Enrofloxacin	0.0200	—
Erythromycin-H ₂ O	0.0500	—
Ethanol, 2-butoxy-phosphate	0.200	0.800–0.840
Fluoranthene	0.0300	0.0400
Fluoxetine	0.0180	—
Gemfibrozil	0.0150	—
Hexahydro-hexamethyl-cyclopentabenzopyran	—	0.500
Hydrocodone	—	0.400
Ibuprofen	0.0180	—
Indole	—	0.0800–15.7
Isoborneol	—	1.13–5.26
Isophorone	—	0.120–3.00
Isopropylbenzene (cumene)	—	0.300
Isoquinoline	—	0.220–9.90
Lincomycin	0.0500	—
Lindane	0.0500	—
Menthol	—	0.819–3.52
Metalaxyl	—	0.120–1.26
Metaxalone	—	4.00
Methadone	—	0.400
5-Methyl-1H-benzotriazole	0.100	1.10–1.90
3-Methyl-1H-indol (skatol)	—	0.0400
1-Methylnaphthalene	—	0.0220
2-Methylnaphthalene	—	0.0360
Methyl parathion	0.0600	—
4-Methyl phenol	0.0400	—
Methyl salicylate	—	0.100
Metolachlor	—	0.0800–0.400
Naphthalene	0.0200	0.0400
n,n,Diethyl-meta-toluamide (DEET)	0.0400	0.0600
4-Nonylphenol	0.500	—
4-Nonylphenol monoethoxylate	1.00	—
4-Nonylphenol diethoxylate	1.10	—
Norfloxacin	0.0200	—

4-Octylphenol monoethoxylate	0.100	0.430–1.00
4-Octylphenol diethoxylate	0.200	1.00
Oxycodone	—	0.400
Oxytetracycline	0.100	—
4-Tert-octylphenol	—	1.00
Paranonylphenol	—	1.48–4.88
p-Cresol	—	0.180
Phenanthrene	0.0600	0.0400
Phendimetrazine	—	0.0400
Phenol	0.250	0.500
Prometon	—	0.200–0.380
Pyrene	0.0300	0.040
Ranitidine	0.0100	—
Roxithromycin	0.0300	—
Salbutamol	0.0290	—
Sarafloxacin	0.0200	—
Stigmastanol	2.00	2.00
Sulfadimethoxine	0.0500	—
Sulfamerazine	0.0500	—
Sulfamethazine	0.0500	—
Sulfamethizole	0.0500	—
Sulfamethoxazole	0.0230–0.0500	—
Sulfathiazole	0.100	—
2-Tert-butyl- and 3-tertbutyl-4-hydroxyanisole	—	1.65
3-Tert-butyl-4-hydroxy anisole	0.120	—
Tetrachloroethylene	0.030	0.120
Tetracycline	0.050–0.100	—
Tri(2-butoxyethyl) phosphate	—	0.800
Tri(2-chloroethyl) phosphate	0.0400	0.100–0.442
Tributyl phosphate	—	0.160
Triclosan	0.0500	0.200–0.210
Tri(dichloroisopropyl) phosphate	0.100	0.100–0.120
Triethyl citrate	—	0.390–1.17
Trimethoprim	0.0140–0.0300	—
Triphenyl phosphate	0.100	0.120
Tylosin	0.0500	—
Virginiamycin	0.100	—
Warfarin	0.00100	—

Notes: —, not analyzed; reporting limits in micrograms per liter.

as described by Barnes et al. (2004), 75 ECs were analyzed using three analytical methods: SPE and single quadrapole, liquid chromatography/mass spectrometry with electrospray ionization set in positive mode, and selected ion monitoring (SIM) (Meyer et al. 2000); SPE and measurement by HPLC/MS using a polar reverse-phase octylsilane (C8) HPLC

column (Cahill et al. 2004); and continuous liquid-liquid extraction and measurement by GC/MS (Brown et al. 1999) (Table 1).

Results and Discussion

Emerging Contaminants Detected in Landfill Leachate

Twenty-eight of the 69 analyzed ECs were detected in one or more of the samples collected from the three landfill cells in February 2009 (Figure 3). Detected ECs included: 4 fecal and plant sterols, 13 household and industrial compounds (including a detergent, a fragrance, plasticizers, cleaning agents, a prescription analgesic, wood preservatives, and fire retardants), 7 hydrocarbons (mostly polycyclic aromatic hydrocarbons (PAHs) associated with combustion of organic matter), 3 pesticides, and the insect repellent *n,n*-diethyl-*meta*-toluamide (DEET) (Figure 3, Table 2). Bisphenol-A, a component of polycarbonate plastics and epoxy resins was detected in all of the water samples collected in February 2009, but negligible recoveries of the surrogate compound bisphenol-A-d3 precluded quantitation of that compound in those samples. The detected ECs were similar to the types of compounds detected in leachate at a landfill in Indiana (Buszka et al. 2009) and to the phenolic compounds and pesticides reported in landfill leachate in Denmark by Christensen et al. (2001). Most studies in this field have used different analytical methods with different EC analytes and reporting limits, making comparison of results between studies

difficult, but all studies reporting on ECs in or near landfills have reported numerous detections of these compounds in landfill leachate and downgradient groundwater. Recent examples of studies using other methods to analyze ECs in landfill leachate include the use of high-pressure liquid chromatography with fluorescence detector (HPLC/FLD) to detect PAHs (Matejczyk et al. 2011) and detection of short-chain carboxylate and sulfonate compounds using SPE followed by liquid chromatography and tandem mass spectrometry (Huset et al. 2011).

ECs detected in leachate at these landfills ranged in concentration from 0.11 to 114 mg/L (Figure 3). The greatest number of ECs (24) was detected in leachate from the cell of intermediate age (2-S, 3 to 16 years old), whereas the least number of ECs (16) was detected in leachate from the operating cell (2-N, less than 5 years old, Figure 3).

Four ECs were detected solely in the oldest leachate sample collected at Landfill 1 (WLMLF) (Figure 3): isoborneol, tri(dichloroisopropyl) phosphate, triphenyl phosphate, and carbaryl. Isoborneol is a bicyclic terpene derived from pines and other conifers that is a precursor to camphor. Isoborneol may leach from components of yard waste and paper, two of the most abundant types of materials in MSW in the United States (U.S. Environmental Protection Agency 2009). Tri(dichloroisopropyl) phosphate is a flame retardant used in fabrics that is chemically similar to the flame retardant tris(1-chloro-2-propyl) phosphate reported in landfill leachate in Norway by Eggen et al. (2010), but similar compounds were not detected downgradient from a landfill in Indiana (Buszka et al. 2009). Triphenyl phosphate is a widely used plasticizer. Carbaryl is a carbamate insecticide that was detected in half of the urban streams sampled for the National Water Quality Assessment Program of the USGS (Gilliom et al. 2006). Two ECs were detected solely in leachate of intermediate age collected at site 2-S: fluoranthene and pyrene (Figure 3), both of which are PAHs produced by the low-temperature, incomplete combustion of hydrocarbons and may be leached from the breakdown of yard waste, paper, or ashes in MSW. PAHs and other organic compounds in landfill leachate are likely to be toxic, persistent, and bioaccumulative, posing risks to the health of humans and aquatic and terrestrial wildlife, according to Matejczyk et al. (2011). No EC was detected solely in leachate from the newest landfill site 2-N (Figure 3). Detection of fewer ECs in leachate collected from the operating cell may have been caused by fewer types of ECs in MSW discarded in recent years due to recycling and hazardous-waste collection programs (U.S. Environmental Protection Agency 2009) or to less time for ECs to dissolve and leach from recently buried MSW.

Some commonality of EC contents of MSW deposited over several decades and the relative recalcitrance of selected ECs to breakdown in landfills were indicated by ECs detected in all three of the leachate samples: the household/industrial compounds camphor, isopropylbenzene (cumene), 4-*tert*-octylphenol, tri(2-butoxyethyl) phosphate, and tributyl phosphate; the hydrocarbons 1-methylnaphthalene, 2-methylnaphthalene, and naphthalene; and the pesticides anthraquinone, 1,4-dichlorobenzene, and DEET (Figure 3, Table 2).

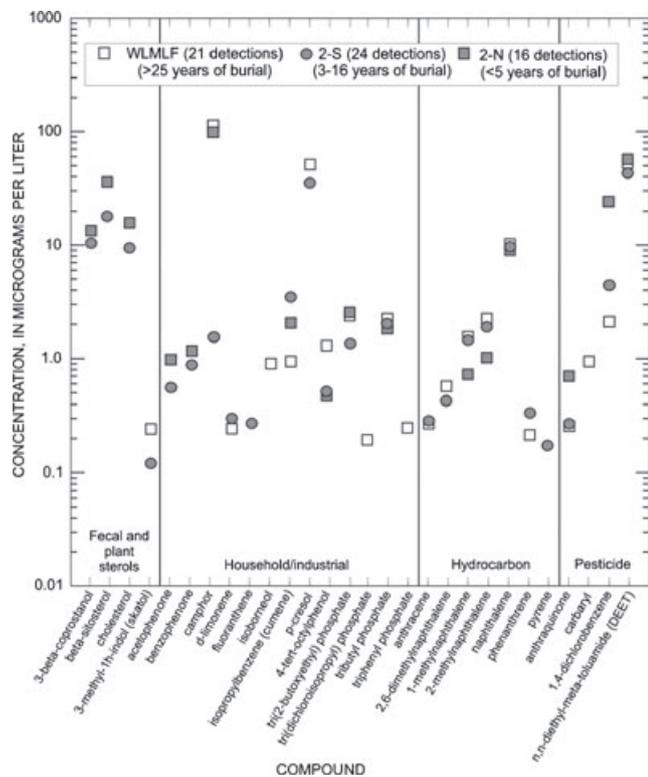


Figure 3. Concentrations of detected emerging contaminants in three leachate samples of different ages collected at two landfills in Central Oklahoma in February 2009.

Table 2
Uses, Sources, and Ranges of Concentrations of Emerging Contaminants Detected in Leachate Samples Collected from Three Different Landfill Cells in Central Oklahoma, February 2009

Detected Emerging Contaminant	Uses/Sources of Detected Emerging Contaminant	Concentrations of Emerging Contaminant (mg/L) by Site and Leachate Age		
		WLMLF (>25 Years of Burial)	Site 2-S (3–16 Years of Burial)	Site 2-N (<5 Years of Burial)
3-Methyl-1h-indol (skatol)	Mammalian feces and coal tar	0.242	E0.12 ¹	<0.0400
3-Beta-coprostanol	Carnivore feces	<2	E10.41	E13.41
Cholesterol	Sterol produced by plants and animals	<2	E9.42 ¹	E15.7 ¹
Beta-sitosterol	Sterol produced by plants	<2	E17.7 ¹	E35.8 ¹
4-Tert-octylphenol	Detergent	E1.24 ¹	E0.486 ¹	E0.463 ¹
Acetophenone	Fragrance	<0.649	E0.516 ¹	E0.906 ¹
Benzophenone	Cosmetics, UV stabilizer	<0.216	E0.807 ¹	E1.07 ¹
Camphor	Plasticizer, fumigant	E114 ¹	E1.55 ¹	E98.8 ¹
d-Limonene	Citrus-based solvent	E0.245 ¹	E0.302 ¹	<1.75
Fluoranthene	Wood preservative	<0.0400	E0.273 ¹	<0.0400
Isoborneol	Plasticizer, adhesive	0.903	<1.13	<5.26
Isopropylbenzene (cumene)	Industrial intermediate	E0.945 ¹	E3.48 ¹	E2.06 ¹
p-Cresol	Wood preservative	E51.2 ¹	E35.2 ¹	<0.180
Tri(2-butoxyethyl) phosphate	Fire retardant	E2.43 ¹	E1.34 ¹	E2.54 ¹
Tri(dichloroisopropyl) phosphate	Fire retardant	0.195 ¹	<0.120	<0.100
Tributyl phosphate	Plasticizer	E2.25 ¹	E2.04 ¹	E1.83 ¹
Triphenyl phosphate	Fire retardant	0.249	<0.120	<0.120
1-Methylnaphthalene	Combusted hydrocarbons	1.59	E1.45 ¹	E0.728 ¹
2,6-Dimethylnaphthalene	Combusted hydrocarbons	0.572 ¹	E0.426 ¹	<0.120
2-Methylnaphthalene	Combusted hydrocarbons	2.25	E1.90 ¹	E1.02 ¹
Anthracene	Combusted hydrocarbons and asphalt	0.271	E0.286 ¹	<0.0400
Naphthalene	Combusted hydrocarbons	9.53	E9.91 ¹	E9.07 ¹
Phenanthrene	Combusted hydrocarbons	0.215	E0.338 ¹	<0.0400
Pyrene	Combusted hydrocarbons	<0.0400	E0.174 ¹	<0.0400
1,4-Dichlorobenzene	Fumigant, herbicide intermediate	E2.11 ¹	E4.41 ¹	E24.0 ¹
Anthraquinone	Pesticide, dye	0.260 ¹	E0.271 ¹	E0.702 ¹
Carbaryl	Insecticide	E0.942 ¹	<0.610	<0.726
n,n-Diethyl-meta-toluamide (DEET)	Insect repellent	E52.6 ¹	E43.7 ¹	E52.8 ¹

¹Estimated concentration less than the least-concentrated calibration standard but greater than the instrument detection limit or greater than the most-concentrated calibration standard.

The fecal and plant sterols 3-beta-coprostanol, beta-sitosterol, and cholesterol were detected in leachate from both the landfill site of intermediate age (2-S) and the operating landfill (2-N), but were not detected in leachate from WLMLF, the oldest landfill cell (Figure 3, Table 2), indicating that leaching of those sterols may be limited to more recently discarded MSW (less than 25 years before present). The fragrance and resin precursor compound acetophenone and the cosmetic and ultraviolet (UV)-stabilizing compound benzophenone also were detected only in leachate from MSW at the site of

intermediate age (2-S) and the operating site (2-N) (Figure 3, Table 2). Those two ECs may be more common in recently deposited MSW, or they may be more readily leached or biodegraded with time than some of the other analyzed ECs.

Concentrations of ECs in landfill leachate provide qualitative information about the EC content of MSW and the attenuation of some ECs in landfill leachate with time. Of the 28 ECs detected in the leachate samples, 12 ECs (3-methyl-1h-indol (skatol), camphor, isoborneol, p-cresol, 4-tert octylphenol, tri(2-chloroisopropyl) phosphate, tributyl phosphate,

triphenyl phosphate, 2,6-dimethylnaphthalene, 1-methylnaphthalene, 2-methylnaphthalene, and carbaryl) were detected in greater concentrations or were only detected in leachate from the oldest age source (WLMLF) than in leachate from the intermediate age source (2-S) and the youngest age source (2-N). The ECs detected in greatest concentrations or only detected in the oldest leachate from WLMLF were monoaromatic, bicyclic, heterocyclic aromatic, polycyclic aromatic, and organophosphate compounds that may have been contributed from a variety of sources/uses, including feces, fumigant/plasticizer, wood preservative, combustion by-products, plasticizer/adhesive, fire retardants, and a pesticide. Many of those compounds continue to be produced and used in commercial products as of 2010 (Table 2).

ECs detected in the greatest concentrations or only detected in landfill leachate of intermediate age at site 2-S included: d-limonene (a natural citrus-based cleaning compound), fluoranthene (an industrial intermediate), isopropylbenzene (cumene), and the hydrocarbons anthracene, naphthalene, phenanthrene, and pyrene (Figure 2, Table 2). ECs detected in greatest concentration in the newest leachate sample collected from site 2-N included: the fecal and plant sterols 3-beta-coprostanol, beta-sitosterol, cholesterol; the household chemicals acetophenone, benzophenone, and tri(2-butoxyethyl)phosphate; and the pesticides 1,4-dichlorobenzene, anthraquinone, and DEET (Figure 2, Table 2). Those ECs are more likely to be parent compounds (unmodified ECs) than the PAHs which dominated the ECs detected in the leachate samples collected at the landfill cell of intermediate age (2-S).

Because many of the ECs detected in the three leachate samples are industrial intermediates and PAHs, some of these ECs may be metabolites of other ECs created by the

processes such as oxidation in water containing oxygen and other oxidizing ions such as nitrate and sulfate, hydrolysis, and reactions such as methylation and demethylation taking place in a range of redox conditions (Cozzarelli et al. 2011). The EC analysis method used for this paper primarily analyzed parent compounds found in commercial products, limiting the possibility of more fully evaluating metabolism of ECs in leachate and groundwater.

Changes in EC Detections and Concentrations with Time and Place at Landfill 1

Occurrence of ECs in landfill leachate and groundwater at Landfill 1 in September 2000 (Barnes et al., 2004) and February 2009 (Table 2) provides qualitative information about the fate and transport of ECs in groundwater near landfills in the decades after closure. Several factors may affect the occurrence and distribution of ECs in groundwater near landfills, including: (a) concentrations of an EC in the MSW, (b) EC chemical properties (e.g., solubility, volatility, and distribution coefficient), (c) aquifer properties (e.g., permeability, dispersivity, groundwater seepage velocity, redox condition, and organic carbon content), and (d) biological properties (numbers of bacterial and fungal species and strains capable of metabolizing a specific EC) (Barber 2008).

ECs detected in both wells 35-5 and 38-6 at Landfill 1 in September 2000 included: 3-beta-coprostanol, cholesterol, 2,6-di-tert-butyl-1,4-benzoquinone, 4-methyl phenol, 4-nonylphenol, 4-nonylphenol monoethoxylate, 4-octylphenol diethoxylate, 4-octylphenol monoethoxylate, tri(2-chloroethyl) phosphate, naphthalene, and DEET (Figure 4, Table 3). Most of those ECs are fecal sterols and phenols that may be related to detergents. ECs detected in all three

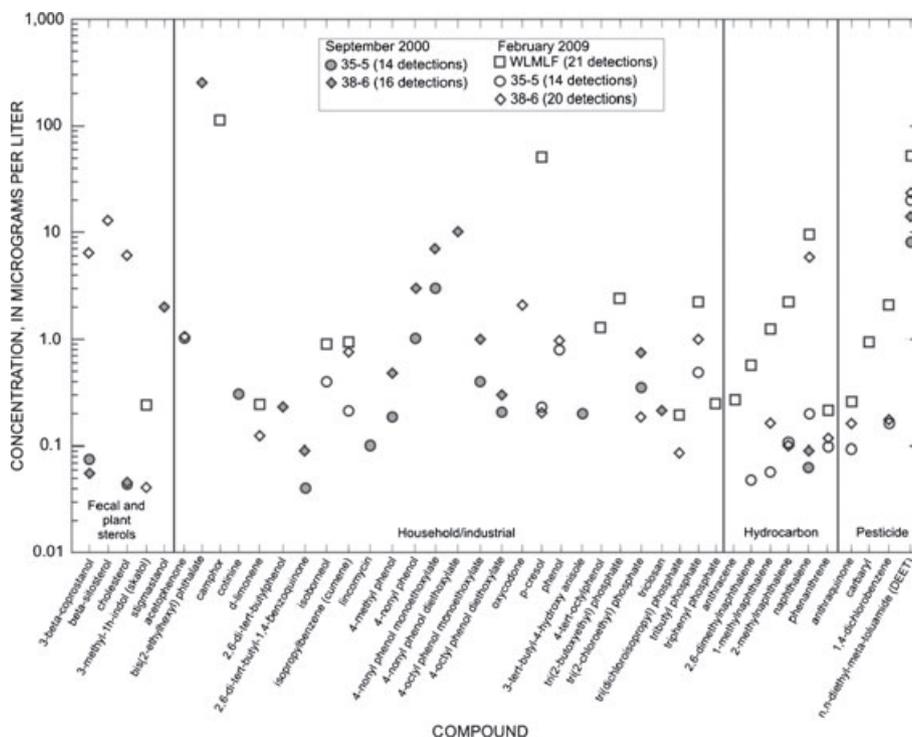


Figure 4. Concentrations of detected emerging contaminants in leachate and downgradient groundwater samples collected from wells at Landfill 1 in central Oklahoma, September 2000 and February 2009.

Table 3
Detection of Emerging Contaminants Detected in Leachate and Groundwater Samples Collected at Landfill 1 in Central Oklahoma in September 2000 and February 2009

Emerging Contaminant	Well Sample in Which Emerging Contaminant was Detected (September 2000)	Well Sample in Which Emerging Contaminant was Detected (February 2009)
Acetophenone	—	35-5, 38-6
Anthracene	—	WLMLF
Anthraquinone	NA	WLMLF, 35-5, 38-6
3-Beta-coprostanol	35-5, 38-6	38-6
Beta-sitosterol	NA	38-6
Bis(2-ethylhexyl) phthalate	38-6	NA
Camphor	NA	WLMLF
Carbaryl	—	WLMLF
Cholesterol	35-5, 38-6	38-6
Cotinine	35-5	—
1,4-Dichlorobenzene	—	WLMLF, 35-5, 38-6
2,6-Dimethylnaphthalene	NA	WLMLF, 35-5
d-Limonene	NA	WLMLF, 38-6
2,6-Di-tert-butylphenol	38-6	NA
2,6-Di-tert-butyl-1,4-benzoquinone	35-5, 38-6	NA
Isoborneol	NA	WLMLF, 35-5
Isopropylbenzene (cumene)	NA	WLMLF, 35-5, 38-6
Lincomycin	35-5	NA
3-Methyl-1h-indol (skatol)	NA	WLMLF, 38-6
1-Methylnaphthalene	NA	WLMLF, 35-5, 38-6

2-Methylnaphthalene	NA	WLMLF, 35-5, 38-6
4-Methyl phenol	35-5, 38-6	NA
Naphthalene	35-5, 38-6	WLMLF, 35-5, 38-6
n,n-Diethyl-methyl-toluamide	35-5, 38-6	WLMLF, 35-5, 38-6
4-Nonylphenol	35-5, 38-6	NA
4-Nonylphenol monoethoxylate	35-5, 38-6	NA
4-Nonylphenol diethoxylate	38-6	NA
4-Octylphenol monoethoxylate	35-5, 38-6	—
4-Octylphenol diethoxylate	35-5, 38-6	—
Oxycodone	NA	38-6
p-Cresol	NA	WLMLF, 35-5, 38-6
Phenanthrene	—	WLMLF, 35-5, 38-6
Phenol	—	35-5, 38-6
Stigmastanol	38-6	—
3-Tert-butyl-4-hydroxy anisole	35-5	NA
4-Tert-octylphenol	NA	WLMLF
Tri(2-butoxyethyl) phosphate	NA	WLMLF
Tri(2-chloroethyl) phosphate	35-5, 38-6	38-6
Tributyl phosphate	NA	WLMLF, 35-5, 38-6
Triclosan	38-6	—
Tri(dichloroisopropyl) phosphate	—	WLMLF, 38-6
Triphenyl phosphate	—	WLMLF

Notes: —, not detected; NA, not analyzed; note that the landfill-leachate well WLMLF had not been installed in September 2000.

of the leachate and groundwater samples collected from wells WLMLF, 35-5, and 38-6 at Landfill 1 in February 2009 included: isopropylbenzene, p-cresol, tributyl phosphate, 1-methylnaphthalene, 2-methylnaphthalene, naphthalene, phenanthrene, 1,4-dichlorobenzene, anthraquinone, and DEET (Figure 4, Table 3). Those mostly monoaromatic and polyaromatic hydrocarbon compounds are prevalent and persistent in the environment (Foght 2008; Seo et al. 2009). Only two ECs were detected in all five samples from Landfill 1, DEET, and naphthalene (Figure 4, Table 3). DEET is an insect repellent that is detected commonly in environmental samples (Kolpin et al. 2002). Naphthalene is an aromatic PAH, is an intermediate, used as manufacture surfactants, plasticizers, and pesticides and is most commonly known

for its use as a fumigant pesticide in mothballs and other products.

Cumulative concentrations of ECs also can provide qualitative information about leaching and biodegradation of those compounds in landfills and downgradient groundwater. At Landfill 1, in September 2000, cumulative EC concentrations were about 10 times greater in water sampled from well 38-6 than in well 35-5, indicating that the core of the organic matter content of the leachate plume had moved about 90 m from the downgradient edge of the landfill (Barnes et al. 2004). In the February 2009 sampling, cumulative concentrations of ECs were greatest in the leachate sampled from well WLMLF, which had not been installed at the time of the 2000 sampling (Figure 5). As with the sampling in

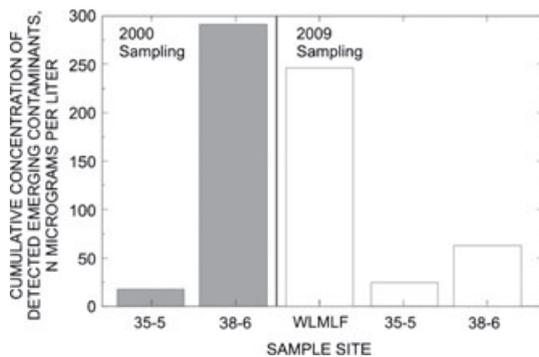


Figure 5. Cumulative-detected concentrations of ECs in the leachate and groundwater samples collected at Landfill 1 in Central Oklahoma in September 2000 and February 2009.

September 2000, greater concentrations of ECs were in water from well 38-6 than in water from well 35-5 at the toe of the landfill, indicating that the bulk of these organic compounds in groundwater may have been transported at least 90 m in the leachate plume seeping from that landfill in the decades after closure. Lesser cumulative EC concentrations in water from well 38-6 in February 2009 than in September 2000, however, may indicate that the core of the EC plume has moved further downgradient from that well. Because no wells further downgradient were sampled at that time, definitive conclusions about further transport of ECs in groundwater at Landfill 1 during that period cannot be made.

Conclusions

Twenty-eight ECs were detected in landfill-leachate samples collected from three landfill cells of different age classes (old, MSW >25 years; intermediate, MSW 16-3 years; and new, MSW <5 years) and included 4 fecal and plant sterols, 13 household/industrial, 7 hydrocarbon, and 4 pesticide compounds. The number of detections and concentrations of ECs from the oldest leachate sample indicate that closed, unlined landfills can be the sources of numerous ECs for many decades after closure. Repeated detection of ECs in leachate from old, intermediate, and new landfills indicate that some ECs have been the components of MSW for many decades and may be relatively resistant to natural attenuation processes under the anaerobic conditions typical in landfills. The large numbers of ECs in landfill leachate pose concerns about the potential discharge of these compounds to adjoining groundwater and surface water and the toxicity, estrogenic activity, carcinogenesis, and other deleterious health effects on aquatic and terrestrial organisms that may be exposed to landfill leachate.

Sampling of ECs from two wells downgradient from the landfill cell containing old MSW in 2000 and 2009 indicated that several ECs were detectable in samples from two downgradient wells during each sampling. The insect repellent DEET and the PAH naphthalene were detected in water samples collected from both wells at both samplings. Greater EC concentrations in samples from the furthest downgradient well from the landfill than in samples from the well at the toe of the landfill indicated that ECs may last for decades

in shallow groundwater and that the core of EC concentrations in the leachate plume may have migrated beyond the toe of the landfill after more than 25 years of closure. Lesser concentrations of ECs in the 2009 sampling than in the 2000 sampling indicate natural attenuation of these compounds caused by transport and degradation processes in the aquifer.

Identification of compounds at waste sites that appear to be persistent and occur at notable frequency is an important step in identifying compounds of emerging concern, as described by Ela et al. (2011). More research is needed regarding the persistence, bioaccumulation, and toxicity of individual ECs and mixtures of ECs in water to evaluate the risk posed to the surrounding environment. Controlled in situ experiments including long-term measurement of ECs and their metabolites in surface water and groundwater downgradient from closed landfills will provide more complete knowledge of EC natural attenuation processes.

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