Chemosphere 127 (2015) 269–275

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Investigating landfill leachate as a source of trace organic pollutants

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HIGHLIGHTS

- Landfill leachates were analyzed to pollutants commonly associated with wastewater effluents.
- Concentration of pollutants ranged several orders of magnitude (ng mg L^{-1}).
- Preliminary mass balance completed to estimate contribution of leachate to WWTPs.
- Landfill leachate maybe an significant source of environmental pollution.

ARTICLE INFO

Article history:
Received 8 December 2014
Received in revised form 5 February 2015
Accepted 6 February 2015
Available online 6 March 2015

Handling Editor: Keith Maruya

Keywords:
Landfill leachate
Contaminants of emerging concern
DEET
Sucralose
Primidone

ABSTRACT

Landfill leachate samples ($n = 11$) were collected from five USA municipal solid waste (MSW) landfills and analyzed for ten trace organic pollutants that are commonly detected in surface and municipal wastewater effluents (viz., carbamazepine, DEET, fluoxetine, gemfibrozil, PFOA, PFOS, primidone, sucralose, sulfamethoxazole and trimethoprim). Carbamazepine, DEET, PFOA and primidone were detected in all leachate samples analyzed and gemfibrozil was detected in samples from four of the five-landfill sites. The contaminants found in the highest concentrations were DEET (6900–143 000 ng L^{-1}) and sucralose (<10–621 000 ng L^{-1}). Several compounds were not detected (fluoxetine) or detected infrequently (sulfamethoxazole, trimethoprim and PFOS). Using the average mass of DEET in leachate amongst the five landfills and scaling the mass release from the five test landfills to the USA population of landfills, an order of magnitude estimate is that over 10 000 kg DEET yr^{-1} may be released in leachate. Some pharmaceuticals have similar annual mean discharges to one another, with the estimated annual discharge of carbamazepine, gemfibrozil, primidone equating to 53, 151 and 128 kg $year^{-1}$. To the authors knowledge, this is the first time that primidone has been included in a landfill leachate study. While the estimates developed in this study are order of magnitude, the values do suggest the need for further research to better quantify the amount of chemicals sent to wastewater treatment facilities with landfill leachate, potential impacts on treatment processes and the significance of landfill leachate as a source of surface water contamination.

1. Introduction

Landfills are the primary repository for the disposal of a wide array of consumer products at the end of their useful life. Products that are routinely disposed to landfills include textiles and carpeting that may be contaminated with fluorinated compounds (Busch et al., 2010); containers that may contain residual quantities of pesticides, insecticides or solvents (Brown and Donnelly, 1988; Öman and Hynning, 1993); and unused pharmaceuticals (Buszka et al., 2009). Landfill leachates are formed by the percolation of water through landfills, where water interacts with waste in an anaerobic environment, concentrating organic matter, inorganic elements and anthropogenically-produced organic compounds. It is thus no surprise that municipal solid waste (MSW) and landfill leachate has been reported to contain a wide variety of organic compounds (Christensen et al., 1994; Öman and Junestedt, 2008; Musson and Townsend, 2009; Busch et al., 2010).

Modern landfills in the United States of America (USA) permitted under Subtitle D of the Resource Conservation and Recovery Act are constructed with a low permeability barrier layer to

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http://dx.doi.org/10.1016/j.chemosphere.2015.02.030
0045-6535/

contain leachate that is collected for treatment (US Congress, October 21, 1976; Benson et al., 2007). Leachate is typically treated at local wastewater treatment facilities where pollutants that are not biologically degraded may be concentrated in the biosolids (Harrison et al., 1999; Clarke and Smith, 2011) or if not attenuated by the treatment process, released into the environment in wastewater effluent (Vanderford et al., 2003). While a broad range of organic chemicals are essential to modern society, many ultimately find their way into the environment where they can cause adverse effects on both living organisms and the environment (Jobling et al., 1998; Daughton and Ternes, 1999). Indeed, the contamination of surface water bodies with persistent organic pollutants (POPs), endocrine disrupting chemicals (EDCs), pharmaceuticals and personal care products (PPCPs) or other 'contaminants of emerging concern' (CECs) is well-documented (Ternes, 1998; Kolpin et al., 2002). Some organic contaminants present in water bodies a threat to natural aquatic ecosystems, but they can also contaminate potable water supplies and pose a risk to public health (Hölzer et al., 2009). The main source of organic pollutants to surface water bodies is thought to be wastewater discharges from domestic and industrial sources; however, landfill leachates treated at municipal wastewater treatment plants are another potential source of environmental pollutants that has received little attention (Eggen et al., 2010).

In 2011, there were an estimated 1908 municipal solid waste (MSW) landfills operating in the USA, as well as thousands of closed landfills, many of which do not have leachate collection systems (US EPA, 2013). While there has been considerable effort to divert materials from landfills to recycling or biological treatment processes (composting, anaerobic digestion), landfills remain the dominant disposal alternative for MSW in the USA. The volume of landfill leachate generated at each landfill will depend upon its surface area and topography, regional climatic conditions such as rainfall, and the cover type. No reliable estimate of the annual volume of landfill leachate generated in the USA is currently available, though a life-cycle model estimated 40 L of leachate could be attributed to each metric ton of MSW disposed, which excludes leachate from older unlined landfills that may be released to groundwater directly (Camobreco et al., 1999).

The composition of waste present in landfills coupled with the quantity of leachate generated will determine the type and concentration of chemicals present in landfill leachate. Many early studies focused on toxic and carcinogenic organic compounds in landfill leachate including BTEX (benzene, toluene, ethylbenzene and xylenes), chlorinated hydrocarbons, polycyclic aromatic hydrocarbons (PAHs) and organochlorine pesticides (Brown and Donnelly, 1988; Öman and Hynning, 1993). Subsequently, hundreds of individual chemicals from a variety of classes has been detected in landfill leachates; including pharmaceuticals (Andrews et al., 2012) , fluorinated surfactants (Busch et al., 2010), phosphate based flame retardants (Yasuhara, 1994), phthalate acid esters (Bauer et al., 1998), bisphenol A (Yamamoto et al., 2001), personal care products (Eggen et al., 2010), polybrominated diphenyl ethers (Öman and Junestedt, 2008), chlorinated dioxins/furans (Öman and Junestedt, 2008) and organotin compounds (Pinel-Raffaitin et al., 2008). A recent publication has highlighted the occurrence of a large number of pharmaceuticals, personal care products, and other substances in US landfill leachates (Masoner et al., 2014). Examples of detected contaminants and their concentrations in landfill leachates are presented in Table 1. The number of contaminants detected in landfill leachate is higher than in other environmental compartments (i.e., soil, surface water, wastewater, air, biosolids) and is likely due to concentration of contaminants in a central location.

Landfill leachates are acutely toxic and the parameters most correlated with toxicity are ammonia, alkalinity and COD (Schrab et al., 1993; Bernard et al., 1996, 1997; Baun et al., 2000). Furthermore, the isolated organic fractions of the leachate using solid-phase extraction (SPE) techniques have also shown toxicity associated with organic pollutants (Baun et al., 2000; Baderna et al., 2011). Studies have demonstrated the genotoxicity (Gajski et al., 2012) and cytotoxicity (Baderna et al., 2011) and a survey of leachates ($n = 58$) from the USA found toxic and cancer causing chemicals irrespective of the type of waste received (Brown and Donnelly, 1988). The environmental impacts of future landfill leachates warrants monitoring as there are currently 90 million organic and inorganic substances identified in the CAS database and over 100000 chemicals routinely used in industrial applications in the U.S. and Europe (ECHA, 2009).

The aim of this study is to measure the concentration of indicator wastewater organic pollutants in landfill leachates and these data will serve as a starting point for an assessment of the significance of landfill leachate as a source of water contamination. The compounds selected for inclusion in this study cover a variety

Table 1
List of organic contaminants reported in landfill leachates and their concentrations.

Contaminants	Country	n	nd	Range ng L^{-1}	References
Bisphenol A	Japan	10		1300–17200000	Baun et al. (2000)
Brominated fire retardants	Sweden	22		<dl–39000	Öman and Junestedt (2008)
	Canada				Li et al. (2012)
Chlorinated solvents	USA			<dl–4616000	Schrab et al. (1993)
1,4-Dioxane	Japan	8		14–109000	Yasuhara et al. (1997)
	Japan	11		<dl–198000	Yasuhara et al. (1999)
Organochlorine pesticides	USA			3–647	Murray and Beck (1990)
Perfluoroalkyl compounds	Germany	20		4–12819	Busch et al. (2010)
Phthalate acid esters	USA			29000–58000	Schrab et al. (1993)
	Japan	8		900–1100	Yasuhara et al. (1997)
	Germany	36		100–240000	Bauer et al. (1998)
	Denamrk	10		1000–340000	Baun et al. (2004)
	Sweden	22		39000	Öman and Junestedt (2008)
Organophosphates	Japan	4		137000–5430000	Yasuhara (1994)
Polyaromatic hydrocarbons	USA			301–1220000	Murray and Beck (1990)
	Sweden	22		8000	Öman and Junestedt (2008)
	Poland	22	0	0.06–77	Matejczyk et al. (2011)
Volatile and semi-volatile compounds	USA			<dl–10200000	Schrab et al. (1993)
	Denmark	31		<1000–15000000 (BTEX)	Kjeldsen et al. (1998)
	Denmark	10		21500–354900 (BTEX)	Baun et al. (2004)
	Poland	22	7	0.16–7.48 (Benzene)	Matejczyk et al. (2011)
	Poland	22	1	4.2–197 (DCM)	Matejczyk et al. (2011)

n = Number of samples analyzed; nd = number of samples below the detection limit; range = only samples above the detection limit.