



Municipal landfill leachates: A significant source for new and emerging pollutants

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ABSTRACT

Landfills have historically remained the most common methods of organized waste disposal and still remain so in many regions of the world. Thus, they may contain wastes resulting from several decades of disposal and decomposition with subsequent release of organic compounds that may have environmental, wildlife and human health consequences. Products containing different types of additives with unique beneficial improvement properties are in daily use. However, when these products are decomposed, additives are release into the environment, some of which have been shown to have negative environmental impacts, resulting in the ban or at least restricted application of some chemicals. New and emerging compounds are continuously discovered in the environment. Herein, we report qualitative and quantitative data on the occurrence of new and emerging compounds with increasing environmental and public health concern in water- and particle phase of landfill leachates. Under normal environmental conditions, several of these chemicals are persistent high-volume products. Identified chemicals in the leachates at nanogram (ng) or microgram (μg) per liter levels include – chlorinated alkylphosphates such as tris(1-chloro-2-propyl) phosphate (TCPP), N-butyl benzensulfonamide (NBBS), the insect repellent diethyl toluamide (DEET) and personal care products such as the non-steroidal anti-inflammatory drug ibuprofen and polycyclic musk compounds. Among new and emerging contaminants, perfluorinated compounds (PFCs) were measured in the water phase at concentrations up to 6231 ng/L. Compared with the other chemicals, PFCs were primarily distributed in water phase. An effective removal method for PFCs and other polar and persistent compounds from landfill leachates has been a major challenge, since commonly used treatment technologies are based on aeration and sedimentation. Thus, the present study has shown that municipal landfill leachates may represent a significant source of concern for legacy, new and emerging chemicals in groundwater.

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1. Introduction

Municipal landfills may be sources of a wide range of compounds with environmental, wildlife and human health concern. Several studies have been performed in order to understand the chemical processes in a landfill and identifying potentially environmentally hazardous compounds in leachates (Paxéus, 2000; Schwarzbauer et al., 2002; Öman and Hynning, 1993). In recent decades, the content of wastes in landfills has changed, mainly as a result of higher demand and better quality of consumer products. For example, batteries, paints, oils, electrical products and pharmaceuticals that are potentially deposited at landfills contain additives that might have a negative impact on environmental and human health. Flame-retardants, plasticizers, surface-active substances and fragrances are chemicals commonly used to improve certain properties of consumer products. Previous studies on landfill leachates have evaluated parameters such as organic matter, chemical- and

biological oxygen demand, nutrients and metal ions. Since the 1990s, extended studies that include organic chemical groups such as phenolic compounds, aromatic acids, chlorinated aromatic compounds, and polycyclic aromatic compounds, have been reported (Paxéus, 2000; Schwarzbauer et al., 2002; Welander, 1997; Öman and Hynning, 1993).

New and emerging groups of compounds such as perfluorinated compounds (PFCs), are now detectable in most environmental matrix (Schultz et al., 2006; Sinclair and Kannan, 2006; Woldegiorgis et al., 2006) and in biota (Kannan et al., 2002; Martin et al., 2004). Particularly, perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) have received attention because of their unique chemistry (as both water and oil repellent and reduction of surface tension), persistence, bioaccumulation and toxic effects. Thus, PFCs have a widespread application in a myriad of products, including household cleaning agents, carpets, textiles, paper coatings, cosmetics, fire-fighting foams and packaged-food containers (Hekster et al., 2003). Presently, few data on landfill leachates have been reported showing that these may represent a significant source of PFCs (Bossi et al., 2008; Kallenborn et al., 2004; Woldegiorgis et al., 2006; Busch et al., 2010). Contrary to many persistent organic pollutants with a

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medium to high hydrophobicity, many PFCs are mainly found in the water phase (Schultz et al., 2006; Woldegiorgis et al., 2006), and efficient removal of these compounds becomes a challenging task during leachate treatment. Many municipal landfills discharge directly into waste water treatment plants (WWTPs) without treatment, resulting to environmental spreading of PFCs, polar and persistent emerging compounds such as N,N-diethyl toluamide (DEET), alkylphosphates, pharmaceuticals and personal care products (PPCPs).

Recently, a wide range of PPCPs has been detected in the environment (Kolpin et al., 2002; Rimkus, 1999). Domestic pathway through municipal sewage treatment plants (STPs) has been recognized as the main route of human pharmaceutical substances into the aquatic environment (Ternes, 1998). Polycyclic musk substances are active ingredients in cosmetics and household products that are used worldwide in large quantities and subsequently ending up in landfills (Salvito, 2005). For example, 1,3,4,6,7,8-hexahydro-4,6,6,7,8,8-hexamethylcyclopenta- γ -2-benzopyran (HHCB or Galaxolide®) and 6-acetyl-1,1,2,4,4,7-hexamethyltetraline (AHTN or Tonalide®) seem to represent the most abundant musk fragrances in the environment that potentially accumulate in sewage sludge (Mogensen et al., 2004). Many of the emerging compounds with suspected risks to organisms and human health include, the insect repellent, DEET (Corbel et al., 2009). Although most of these compounds are present at trace levels in the environment, harmful effects to the ecosystem and to the food web cannot be excluded. Persistent drugs such as carbamazepin, is not effectively removed from wastewater and may therefore enter, at least, the aquatic environment with unknown implications (Andreozzi et al., 2002; Isidoria et al., 2005; Zurita et al., 2007). Regulatory consequences on the risks associated with the presence of these compounds in the environment have been the subject of continuous discussion for decades (Ternes, 2001).

The aims of the present were, firstly to investigate the levels of emerging organic compounds in untreated landfill leachates using a non-target full scan screening analysis (landfill A, B and C) and secondly perform quantitative analysis of selected PPCPs (landfill A) and PFC (landfill B and C). In order to get an overall knowledge about the distribution patterns and suitable treatment processes for removing such compounds in untreated leachates, the quantification of emerging compounds were performed separately in water- and particle phases.

2. Materials and methods

2.1. Leachate sampling

Three differently engineered landfills are included in this study. Landfill A is located in an abandoned sand/gravel pit without bottom liner. The bottom of the old part of the landfill is approximately at the groundwater level and the landfill is further limited to the east and north by solid bedrock, granite gneiss. The landfill is limited on the Westside by a ridge of massive rock, overlaid by a thin layer of compact moraine, sand/gravel and locally marine clay. Leachate from this site enters the groundwater through the sand/gravel aquifer and follows a well-defined path to a concentrated spring located 200 m downstream before a treatment system containing an aerated lagoon and a wetland filter. Five leachate samples were collected at and downstream of landfill A; i) a well located just below the old landfill site (W-old), ii) a well in the lower part of the currently active landfill (W-active), iii and iv) wells located 50 (W-50) and 100 (W-100) downstream of the landfill, respectively, and v) inlet water to the treatment system, taken in the pumping house, 200 m downstream of the landfill (W-200) (Fig. 1). Both landfills B and C had clay liners and a tubing system for collecting leachates and a direct discharge of the leachates without treatment to the municipal wastewater net. Groundwater (W-old, W-

active, W-50 and W-100) for non-target screening was sampled with the use of a peristaltic pump with Teflon tubing. The samples were pumped into autoclaved acidified (10 mL 3 M H₂SO₄) N₂-purged 1-liter glass flasks. Leachates from landfills B, C and W-inlet at landfill A, were sampled directly into pre-prepared 1 L sampling bottles. Non-target screening samples and quantitative PPCP analysis were sampled in glass bottles while samples for quantitative PFC analysis was sampled into methanol washed PE quality plastic bottles. The flasks were filled completely and stored in dark conditions, at 4 °C until analysis. Information on the landfills is summarized in Table 1.

2.2. Non-target screening and target analysis of selected emerging compounds

Qualitative screening for organic compounds were performed only with leachates from landfills A, B and C. Sample preparation by solid phase extraction (SPE) was performed essentially as previously described by Braun et al. (2003). Briefly, 1-L of each water sample was filtered and thereafter Soxhlett-extracted with toluene. From a final volume of 500 μ L, a 1 μ L aliquot was analyzed by GC-MS. The water samples (adjusted to pH 3 and spiked with the internal standard) were extracted by SPE on a mixed sorbent bed (LiChrolut® EN and LiChrolut® C18; both from Merck, Darmstadt, Germany) and eluted with acetone. A cleanup on silica gel accomplished the sample preparation prior GC-MS analysis. A GC-MSD instrument (6890 HP-MSD; Agilent Technology, Waldbronn, Germany) equipped with a 30 m HP-5MS capillary (0.25 mm I.D., 0.25 μ m film) was used for analysis. Full scan mode (50–500 amu) was applied for qualitative screening (non-target analysis) and selected ion monitoring (SIM) mode for quantification of selected emerging substances. The carboxylic drugs were analyzed after derivatization with GC-negative chemical ionization mass spectrometry in accordance to Möder et al. (Möder et al., 2007). Quantitative analysis of the PPCPs, were performed with samples from landfill A and the limits of quantification (LOQ) are specified in Table 4.

2.3. Determination of ionic perfluorinated substances and telomers

Quantitative analyses of PFCs in water- and particle phases were performed for leachates from landfills B and C, and sediment samples from landfill D. All sampling devices were free of perfluorinated polymers to avoid cross-contamination. About 2 g of dry sample was spiked with 20 μ L internal standard, ¹³C PFOS and ¹³C PFOA (0.5 ng/ μ L). 1 mL of 200 mM NaOH in methanol was added and soaked for 30 min. Then, 100 μ L 2 M HCl in methanol was added followed by another 9 mL of methanol. The mixture was thoroughly mixed and then extracted on a wrist-action-shaker for 30 min. After centrifugation (2000 rpm, 5 min), 1 mL of supernatant methanol extract was treated with 25 mg ENVI-Carb mixed in 50 μ L glacial acetic acid. After thorough mixing and centrifugation (10,000 rpm, 10 min), 20 μ L recovery standard, 0.1 ng/ μ L 3,5-bis(trifluoromethyl)phenyl acetic acid, and 0.5 mL of 4 mM NH₄ OAc in water were added to the final extract prior to analyses by the HPLC/MS system (binary pump 1525 μ with a Waters 2777 Sample Manager coupled to a Micromass (Q)-TOF-MS (QTOF micro)).

2.4. Determination of fluorotelomer alcohols

Wet sediment samples were spiked with internal standard, 7:1 FA (7:1 fluorinated alcohol, 98% purity) (ABCR Karlsruhe, Germany), and extracted with ethyl acetate three times for 30 min on a wrist-action shaker. Centrifugation and cleanup on ENVI-Carb was performed as described above, without the addition of glacial acetic acid. Recovery standard, 1,2,3,4-tetrachloronaphthalene (1,2,3,4-TCN, 10 μ g/mL in isooctane) (Dr.Ehrenstorfer, Augsburg, Germany), was added prior to

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