Water Research 121 (2017) 46–60

Contents lists available at ScienceDirect

Water Research

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

# Removal of selected PPCPs, EDCs, and antibiotic resistance genes in landfill leachate by a full-scale constructed wetlands system

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ARTICLE INFO

*Article history:*
Received 24 February 2017
Received in revised form
27 April 2017
Accepted 4 May 2017
Available online 8 May 2017

*Keywords:*
Antibiotic resistance genes (ARGs)
Constructed wetlands (CWs)
Emerging contaminants (ECs)
Landfill leachate
Pharmaceuticals and personal care products (PPCPs)

ABSTRACT

Landfill leachate could be a significant source of emerging contaminants (ECs) and antibiotic resistance genes (ARGs) into the environment. This study provides the first information on the occurrence of selected ECs and ARGs in raw leachate from 16-year old closed landfill site in Singapore. Among the investigated ECs, acetaminophen (ACT), bisphenol A (BPA), clofibric acid (CA), caffeine (CF), crotamiton (CTMT), diclofenac (DCF), *N,N*-diethyl-m-toluamide (DEET), gemfibrozil (GFZ), lincomycin (LIN), salicylic acid (SA), and sulfamethazine (SMZ) were the most frequently detected compounds in raw landfill leachate. The concentrations of detected ECs in raw landfill leachate varied significantly, from below quantification limit to 473,977 ng/L, depending on the compound. In this study, Class I integron (*intl1*) gene and ten ARGs were detected in raw landfill leachate. Sulfonamide resistance (*sul1, sul2*, and *dfrA*), aminoglycoside resistance (*aac6*), tetracycline resistance (*tetO*), quinolone resistance (*qnrA*), and *intl1* were ubiquitously present in raw landfill leachate. Other resistance genes, such as beta-lactam resistance (*blaNMD1*, *blaKPC*, and *blaCTX*) and macrolide-lincosamide resistance (*ermB*) were also detected, detection frequency of <50%. The removal of target ECs and ARGs by a full-scale hybrid constructed wetland (CW) was also evaluated. The vast majority of ECs exhibited excellent removal efficiencies (>90%) in the investigated hybrid CW system. This hybrid CW system was also found to be effective in the reduction of several ARGs (*intl1*, *sul1*, *sul2*, and *qnrA*). Aeration lagoons and reed beds appeared to be the most important treatment units of the hybrid CW for removing the majority of ECs from the leachate.
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## 1. Introduction

In recent years, emerging contaminants (ECs), such as pharmaceuticals and personal care products (PPCPs) and endocrine disrupting chemicals (EDCs), have increasingly gained attention due to their omnipresence in the environment and their potential to cause undesirable ecological effects (Daughton and Ternes, 1999; Terzic et al., 2008; Kummerer, 2009a, 2009b; Verlicchi et al., 2010). It has been documented that PPCPs enter the aquatic environment via a number of pathways, including wastewater treatment plant (WWTP) effluent (Nakada et al., 2008; Kuroda et al., 2012; Tran et al., 2016b), discharge of wastewater from hospitals or pharmaceutical industrial zones, direct discharge of animal wastewater from livestock breeding, poultry processing, aquaculture, septic system, etc. (Kummerer, 2001; Bound and Voulvoulis, 2005; Verlicchi et al., 2010; Tran et al., 2014b, 2015). So far, most of the studies on the occurrence, fate and pollution control and management of PPCPs and EDCs have focused on wastewater, surface water and groundwater compartments, while little information has been reported for landfill sites.

Landfills have historically remained the most common method for disposal of municipal solid waste and still remain so in many countries (Eggen et al., 2010; Clarke et al., 2015; Lu et al., 2016). After several decades of disposal and decomposition, landfill leachates are sources of a wide range of chemical and microbial pollutants (Eggen et al., 2010; Clarke et al., 2015; Wang et al., 2015; Song et al., 2016). In an earlier study, Eggen et al. (2010) found that a large number of ECs, such as PPCPs, chlorinated alkyl phosphates, perfluorinated chemicals (PFCs), were detected in municipal

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http://dx.doi.org/10.1016/j.watres.2017.05.008
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landfill leachates. Similarly, in recent studies, it has been realised that municipal landfill leachates may represent a significant source of concern for emerging contaminants, including pharmaceutically active compounds, antibiotics, and personal care products in the aquatic environment (Clarke and Smith, 2011; Clarke et al., 2015; Lu et al., 2016; Sui et al., 2017).

Apart from the presence of chemical pollutants in landfill leachates, microbial contaminants (i.e. pathogens, enteric viruses, antibiotic resistant bacteria and/or antibiotic resistance genes) caused by leachates is another important aspect of concern (Tigini et al., 2014; Wang et al., 2015; Song et al., 2016). For example, Wang et al. (2015) proved that the occurrence of high levels of antibiotic resistant bacteria (ARB) and antibiotic resistance genes (ARGs) in landfill leachate, indicating that landfill and landfill leachate are considered as antibiotic resistance reservoirs. To date, it is unclear whether the presence of ARB/ARGs in the landfill leachate is associated with the migration of unused and unwanted PPCPs, particularly in terms of antibiotics and antimicrobial agents. The greatest concern for the release of PPCPs and ARGs from landfill leachate into the water environment is the evolution of antibiotic resistant bacteria via continuous exposure to antimicrobials in the leachates even at very low concentration (i.e. ng/L or μ g/L) over the long-term (Martínez, 2008; Gullberg et al., 2011). For example, Martínez (2008) proposed that anthropogenic variation of the environment might enrich the population of resistant bacteria and facilitate the transmission of ARGs to human pathogens. In another study, Cattoir et al. (2008) found that plasmid-encoded *qnr* genes present in waterborne bacteria were enriched after being exposed to quinolones in river waters. It is noted that *qnr* genes are normally found in the chromosomes of waterborne bacteria where it has an unknown function (Poirel et al., 2005). However, after being integrated into plasmids, bacteria carrying such plasmids exhibit a resistance to quinolones (Poirel et al., 2005). As such, the enrichment of plasmid-encoded *qnr* genes in waterborne bacteria in river water can be considered as a first step in the transfer of this ARG to human pathogens (Cattoir et al., 2008; Martínez, 2008).

Till now, there has been still insufficient evidence to conclude that the presence of ARGs in nonclinical environments results in the development of clinical antibiotic resistance pathogens, since the transfer of ARGs from reservoirs to other bacteria is probably a rare and random event (Manaia, 2017). However, it is widely believed that the transmission of ARGs from nonclinical environment to bacterial pathogens can take place via horizontal gene transfer by mobile genetic elements, such as integrons, transposons and plasmids (Martínez, 2008; Pal et al., 2016; Manaia, 2017). In particular, the high abundance of ARGs under a particular environment, such as the co-occurrence of antimicrobial biocides and heavy metals, may also promote the selection of antibiotic resistance (Pal et al., 2016).

For these reasons, the removal of PPCPs, EDCs, and ARGs from landfill leachate is critically needed to protect aquatic ecosystems and reduce the risk of antibiotic resistance transmission from the environment to humans. Hitherto, numerous wastewater treatment technologies have been developed for removing both chemical and microbial pollutants in landfill leachate (Guo et al., 2008; Bove et al., 2015; Gao et al., 2015; Wu et al., 2015b; Lu et al., 2016; Sui et al., 2017). For instance, membrane bioreactors and membrane separation or advanced chemical oxidation processes (AOPs) are reported to efficiently remove both chemical and microbial pollutants, but these technologies are rather expensive and not entirely feasible for widespread application in rural areas (Guo et al., 2008; Wu et al., 2015b; Lu et al., 2016; Sui et al., 2017). Among the developed wastewater treatment technologies, constructed wetlands (CWs) are considered to be reasonable options for treating landfill leachate due to its lower cost, easy operation and less maintenance requirements (Wu et al., 2015b; Liu et al., 2016). Up to now, only two studies in Spain have reported about the removal of PPCPs by full-scale horizontal subsurface flow (HSSF) constructed wetlands (CWs) (Matamoros et al., 2009; Hijosa-Valsero et al., 2010). Similarly, in relation to removal of ARGs using CWs, it remains a relatively new treatment method. Some studies in China and Europe have investigated the removal of ARGs by CWs, but these studies showed diverse results and conclusions on the removal performance, probably due to their different CW configurations, operating conditions, and influent compositions (Liu et al., 2013; Nõlvak et al., 2013; Berglund et al., 2014; Lv et al., 2015). Therefore, the first objective of this study was to provide the first data on the occurrence of selected PPCPs, EDCs, and ARGs in raw landfill leachate collected from a closed landfill site in a tropical region (Singapore), where climate condition and age of the landfill site are different from those reported in other countries (China, Japan, Spain, and U.S). The second objective of this study was to evaluate the removal performance of PPCPs, EDCs, and ARGs by a full-scale hybrid CWs in Singapore. This hybrid CW system contains different treatment units (equalization tanks, aerobic lagoons, reed beds, and polishing ponds).

## 2. Materials and methods

### 2.1. Target compounds, solvents and other chemical reagents

The target compounds monitored in this study were 29 selected PPCPs and EDCs belonging to different therapeutic classes.

(i) Anticonvulsants: carbamazepine (CBZ) and gabapentin (GBP).
(ii) Anti-itching: crotamiton (CTMT).
(iii) Antimicrobial agents: triclosan (TCS) and triclocarban (TCC).
(iv) Antipsychotic drug: sulpiride (SPR).
(v) Beta-blocker: atenolol (ATN).
(vi) Chloramphenicol (CAP).
(vii) Glycopeptide antibiotic: vancomycin (VCM).
(viii) Lipid regulating drugs: clofibric acid (CA) and gemfibrozil (GFZ).
(ix) Lincosamide antibiotics: clindamycin (CLI) and lincomycin (LIN).
(x) Macrolide antibiotics: azithromycin (AZT), clarithromycin (CLAR), erythromycin (ERY), and tylosin (TYL).
(xi) Nonsteroidal anti-inflammatory drugs (NSAIDs): acetaminophen (ACT), diclofenac (DCF), fenoprofen (FEP), indomethacin (IDM), naproxen (NPX), and salicylic acid (SA).
(xii) Plasticizer: bisphenol A (BPA).
(xiii) Reductase inhibitor: trimethoprim (TMP).
(xiv) Repellent: N, *N*-diethyl-m-toluamide (DEET).
(xv) Stimulant: caffeine (CF).
(xvi) Sulfonamide antibiotics: sulfamethoxazole (SMX) and sulfamethazine (SMZ).

The physicochemical properties of the target antimicrobials are presented in Table S1 (Supplementary Information). All the target antimicrobials as well as other chemical reagents/solvents are of high purity grade (>99%) and were purchased from Sigma–Aldrich (Sigma–Aldrich, Singapore). Twenty-one $^{2}H$ and $^{13}C$ isotope labeled internal/surrogate standards (ILISs) were purchased from Toronto Research Chemicals (Toronto, Canada), including acetaminophen-$d_4$ (ACT-$d_4$), bisphenol A-$d_6$ (BPA-$d_6$), carbamazepine-$d_8$ (CBZ-$d_8$), caffeine-$d_9$ (CF-$d_9$), diclofenac-$d_4$ (DCF-$d_4$), *N*,*N*-diethyl-m-toluamide-$d_{10}$ (DEET-$d_{10}$), gemfibrozil-$d_6$ (GFZ-$d_6$), indomethacin-$d_4$ (IDM-$d_4$), naproxen-$d_3$ (NPX-$d_3$), salicylic acid-$d_4$ (SA-$d_4$), azithromycin-$d_3$ (AZT-$d_3$), chloramphenicol-$d_5$ (CAP-$d_5$),

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