Chemosphere 183 (2017) 257–265

Contents lists available at ScienceDirect

Chemosphere

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

Occurrence, distribution and bioaccumulation behaviour of hydrophobic organic contaminants in a large-scale constructed wetland in Singapore

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HIGHLIGHTS

- Muti-class hydrophobic organic contaminants were detected in different compartments of a large-scale constructed wetland.
- Root concentration factors (log RCFs) were positively correlated with octanol-water partition coefficients (log Kow's).
- Positive linear relationship between root concentration factor (log RCF) and octanol-water partition coefficient (log K_{OW}).
- Translocation factors (log TFs) were negatively correlated with log K_{OW}.
- Dechlorane plus isomers exhibited a high degree of accumulation in plant leaves.
- Relatively high concentrations of triclosan degradation products (MTCS and 2,8 DCDD) observed in plant roots and leaves.

ARTICLE INFO

Article history:
Received 20 March 2017
Received in revised form
18 May 2017
Accepted 19 May 2017
Available online 20 May 2017

Handling Editor: Myrto Petreas

Keywords:
Synthetic musks
Triclosan
Halogenated flame retardants
Constructed wetland
Typha angustifolia
Bioaccumulation
Translocation

ABSTRACT

This study involved a field-based investigation to assess the occurrence, distribution and bioaccumulation behaviour of hydrophobic organic contaminants in a large-scale constructed wetland. Samples of raw leachate, water and wetland plants, *Typha angustifolia,* were collected for chemical analysis. Target contaminants included polychlorinated biphenyls (PCBs), organochlorine pesticides (OCP), as well as several halogenated flame retardants (HFRs) and personal care products (triclosan and synthetic musks). In addition to PCBs and OCPs, synthetic musks, triclosan (TCS) and dechlorane plus stereoisomers (*syn*- and *anti*-DPs) were frequently detected. Root concentration factors (log RCF L/kg wet weight) of the various contaminants ranged between 3.0 and 7.9. Leaf concentration factors (log LCF L/kg wet weight) ranged between 2.4 and 8.2. *syn*- and *anti*-DPs exhibited the greatest RCF and LCF values. A strong linear relationship was observed between log RCF and octanol-water partition coefficient (log K_{OW}). Translocation factors (log TFs) were negatively correlated with log K_{OW}. The results demonstrate that more hydrophobic compounds exhibit higher degrees of partitioning into plant roots and are less effectively transported from roots to plant leaves. Methyl triclosan (MTCS) and 2,8-dichlorodibenzo-p-dioxin (DCDD), TCS degradation products, exhibited relatively high concentrations in roots and leaves., highlighting the importance of degradation/biotransformation. The results further suggest that *Typha angustifolia* in this constructed wetland can aid the removal of hydrophobic organic contaminants present in this landfill leachate. The findings will aid future investigations regarding the fate and bioaccumulation of hydrophobic organic contaminants in constructed wetlands.

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1. Introduction

Constructed wetlands are commonly used as an alternative nature-assisted water treatment technology, which can be employed to substitute and/or supplement conventional wastewater treatment plants. In particular, constructed wetlands can be useful for treatment of landfill leachate, which may contain a variety of potentially hazardous chemicals. Previous studies have demonstrated the occurrence of heavy metals, organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), perfluroalkyl substances (PFASs), pharmaceuticals and personal care products

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http://dx.doi.org/10.1016/j.chemosphere.2017.05.113
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(PPCPs), endocrine disrupting chemicals (EDCs) and halogenated flame retardants (HFRs) in landfill leachate (Baun et al., 2004; Osako et al., 2004; Slack et al., 2005; Eggen et al., 2010; Andrews et al., 2012; Madera-Parra et al., 2015; Dan et al., 2017).

Constructed wetlands have been shown to exhibit good removal efficiencies of COD, BOD, nutrients and heavy metals (Juwarkar et al., 1995; Cheng et al., 2002). However, relatively few studies have focused on assessing the removal of hydrophobic organic chemicals. Previous studies have shown that PCBs discharged into a wetland were removed by sorption processes, with removal efficiency ranging between 71% and 91% (Wojciechowska, 2013). Synthetic musk compounds were found to be quantitatively eliminated by hydrophobic interactions in constructed wetlands, with removal efficiency ranging between 30% and 80% (Matamoros and Bayona, 2006). Studies have also demonstrated that pharmaceuticals can be removed effectively in constructed wetlands, with removal efficiency ranging between 23% and 90% (Li et al., 2014). Different constructed wetland systems may exhibit different removal capabilities, due to differences in temperature, hydraulic retention time (HRT), depth and plants species utilized. Further, removal efficiency for a given chemical is highly dependent on physicochemical properties and resistance to abiotic and biotic degradation processes.

The process of bioaccumulation in aquatic plants is undoubtedly important for understanding the attenuation and removal of contaminants in constructed wetland systems. Waterborne chemicals can sorb to plant roots, as well as translocate and partition into stem and leaf tissue during evapotranspiration (Létondor et al., 2015; Hurtado et al., 2016). In addition, symbiotic bacteria associated with plant roots can result in biotransformation of the chemical, typically resulting in more water soluble metabolites (Singer et al., 2003; Chaudhry et al., 2005). Photodegradation of chemicals within the plant can also occur (Katagi, 2004). Common aquatic plants utilized in constructed wetlands include *Typha latifolia, Typha angustifolia, Cyperus papyrus, Alternanthera philoxeroides, Colocasia esculenta, Scirpus validus, Acorus calamus, Chrysopogon zizanioides* and *Pontederia cordata* (Zarate et al., 2012; Deng et al., 2016).

The present study involves a field investigation to assess the occurrence, distribution and bioaccumulation behaviour of various hydrophobic organic compounds in a large-scale constructed wetland in Singapore. The studied wetland, Lorong Halus wetland, is a horizontal subsurface flow constructed wetland, which receives leachate from a decommissioned landfill. The studied chemicals include legacy POPs, such as PCBs, OCPs and polybrominated diphenyl ethers (PBDEs), along with several alternative flame retardants, synthetic musks, as well as triclosan (TCS) and its degradation products.

2. Methodology

2.1. Field sampling

The present study involved a field investigation of the Lorong Halus wetland, a large-scale constructed wetland in northwest Singapore that treats leachate associated with an adjacent decommissioned landfill. The original landfill had a designed capacity of up to 3000 m^3/d and received untreated municipal solid waste, as well as construction and demolition waste between 1970 and 1999 (Yin et al., 2016). A schematic illustration of the different wetland components and water flow directions is shown in Fig. 1. The overall retention time of this constructed wetland was designed to be approximately 10.2 d, including 0.3 d for equalization tank (volume: 2630 m^3), 3.4 d for aeration lagoon (total area 2453 m^2), 1.3 d for sedimentation tank (volume: 7350 m^3), 3.2 d for reed beds (total area 38,000 m^2) and 2 d for the polishing ponds (total area 13,000 m^2), respectively (Sim et al., 2013). There are approximately 160,000 plants consisting of eight different species within the wetland reed beds, with cattail *Typha angustifolia* being the most abundant species. The leachate inflow is relatively low and is diluted with reservoir water prior to the entering the equalization tank. The mixing ratio (raw leachate:reservoir water) prior to the equalization tank is typically between 1:1.5 to 1:3. The flow rate of this diluted leachate entering the constructed wetland system is on average 300 m^3/d.

1 L amber glass bottles were used to collect replicate samples ($n = 3$) of the leachate (S1), as well as water outflow from the sedimentation tank (S2), water outflow from reed bed number 3 (S3), water outflow from the polishing pond (S4) on the 8th October 2015 and 11th February 2016. The leachate and other water samples were collected at the corresponding outlet pipes and processed immediately after they were transported back to the lab. Suspended particulate matter (SPM) was obtained by filtrating the water using glass fibre filter disks (47 mm, pore size 1 μm). During the same sampling events, *Typha angustifolia* were also collected ($n = 4$ per site) randomly at reed bed 3 (S3). The plants are maintained in a gravel substrate (10–30 cm depth). The selected plants were mature (≥4 months), ranging in height between 1.5 and 2 m, with a total biomass between 0.5 and 1 kg per plant. Plant roots and leaves were and washed with MiliQ water prior to extraction. The moisture content of the roots and leaves were 93.8% and 82.5%, respectively.

2.2. Chemicals and reagents

The extraction solvents were HPLC grade dichloromethane (DCM), hexane and isooctane, all purchased from Fisher Scientific (Loughborough, UK). Granular anhydrous sodium sulfate and florisil (60–100 mesh) were obtained from Sigma (St Louis, MO, USA). Information regarding the target analytes studied is summarized in Tables S1–S4. Chlorinated pesticides, PCBs congeners, PBDEs and chlorobenzenes were purchased from AccuStandard (New Haven, CT, USA). Pentabromoethylbenzene (PBEB), tribromophenoxyethane (BTBPE), hexabromobenzene (HBB) and dechlorane plus isomers (*anti*-DP and *syn*-DP) were purchased from Wellington Laboratories (Ontario, Canada). Musk ketone (MK) was obtained from Sigma (St Louis, MO, USA). Polycyclic synthetic musks, including celestolide (ADBI), galaxolide (HHCB), tonalide (AHTN) and Traseolide (ATII), were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). TCS, as well as methyl triclosan (MTCS) and 2,8-dichlorodibenzo-p-dioxin (2,8-DCDD) were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany) as well. 1,2,4,5-tetrabromobenzene and a mixture of tetrabromoethylcyclohexane (TBECH) isomers were from Sigma (St Louis, MO, USA). 1,3,5-tribromobenzene was purchased from Tokyo Chemical Industry (Tokyo, Japan).

Carbon-13 ($^{13}C_x$) and deuterium (d_x) labelled compounds were purchased in order to enable isotope dilution quantification of target analytes in field samples. These isotopically labelled Internal surrogate standards, included musk xylene-d_{15}, tonalide-d_3, $^{13}C_6$-α-hexachlorocyclohexane (HCH), $^{13}C_6$-γ-HCH (Dr. Ehrenstorfer GmbH, Augsburg, Germany), a mixture of $^{13}C_{12}$-PCBs, $^{13}C_6$-1,2,3,4-tetrachlorobenzene ($^{13}C_6$-1,2,3,4-TeCBz), $^{13}C_6$-1,2,3,4-hexachlorobenzene ($^{13}C_6$-HxCBz), 1,2,4-trichlorobenzene-d3 (1,2,4-TriCBz-d_3, Cambridge Isotope Laboratories, Andover, MA, US), a mixture of $^{13}C_{12}$-polybrominated diphenyl ethers ($^{13}C_{12}$-PBDEs), $^{13}C_6$-HBB, $^{13}C_{12}$-MTCS (Wellington Laboratories, Ontario, Canada). $^{13}C_{12}$-$^{13}C_{12}$-PCB-111 and $^{13}C_{12}$-BDE-77, used as a recovery standards, were purchased from Cambridge Isotope Laboratories (Andover, MA, US) and Wellington Laboratories (Ontario, Canada),

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