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Removal of polycyclic musks by anaerobic membrane bioreactor: Biodegradation, biosorption, and enantioselectivity



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HIGHLIGHTS

• Anaerobic MBR was effective for removing PCMs from a wastewater solution.

• Biotransformation was the dominant removal mechanism for all five PCMs.

• PCMs were significantly partitioned to the biosolids phase in the anaerobic reactor.

• Negligible enantioselectivity was observed in the removal of chiral PCMs in AnMBR.

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ABSTRACT

This study aims to investigate the performance of anaerobic membrane bioreactor (AnMBR) for removing five polycyclic musks (PCMs), which are common active ingredients of personal care and household cleaning products. A laboratory scale AnMBR system was used in this investigation. Concentrations of the PCMs in both the liquid and biosolids phase were measured to conduct a mass balance analysis and elucidate their fate during AnMBR treatment. The AnMBR was effective for removing PCMs from the aqueous phase by a combination of biotransformation and sorption onto the biosolids. However, biotransformation was observed to be the dominant removal mechanism for all five PCMs. Enantioselective analysis of the PCMs in influent, effluent and biomass samples indicated that there was negligible enantioselectivity in the removal of these PCMs. Accordingly, all enantiomers of these PCMs can be expected to be removed by AnMBR with similar efficiency.

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1. Introduction

Reclaimed municipal effluent is an increasingly important water resource used in many countries for a diverse range of applications including agricultural irrigation, industrial processes, non-potable usage and even to supplement potable water supplies. As a consequence, there has been an increasing attention to the elimination of trace organic chemicals (TrOCs) during the wastewater treatment and reclamation processes. Conventional wastewater treatment processes were not specifically developed for removing TrOCs (Le-Minh et al., 2010b; Rivera-Utrilla et al., 2013). Thus, the removal of some TrOCs can be quite low or highly variable. In recent years, membrane bioreactors (MBRs) have been shown to improve the removal of refractory trace chemicals as a consequence of extended biosolids retention times and high biomass concentrations (Alturki et al., 2010; Le-Minh et al., 2010a, 2010b). Many studies have shown the effective removal of TrOCs including pharmaceuticals and personal care products (PPCPs), pesticides, and endocrine disrupting chemicals by MBRs (Coleman et al., 2009; Nghiem et al., 2009; Tadkaew et al., 2011; Trinh et al., 2012). In particular, MBRs have been shown to achieve improved removal of some contaminants, which have otherwise been considered to be relatively persistent and recalcitrant compounds during treatment (Clara et al., 2005; De Wever et al., 2007; Radjenovic et al., 2009; Sipma et al., 2010; Tambosi et al., 2010).

In addition to the more established aerobic MBR systems, there is a growing interest in of the deployment of anaerobic MBR (AnMBR) systems for municipal wastewater treatment (Lew et al., 2009). Compared to aerobic MBR, AnMBRs can be much more energy efficient but can also maintain a high effluent quality suitable for environmental discharge and water reuse. Other



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advantages of AnMBRs include the reduction in chemical consumption and sludge production (Ozgun et al., 2013; Gao et al., 2014). In addition, AnMBR can convert the organic content in wastewater to biogas, which is a renewable fuel (Visvanathan and Abeynayaka, 2012). The further optimisation of these advantages may see the implementation of AnMBR systems as a cost-effective option for municipal wastewater treatment plants in the coming years.

Several studies have previously been conducted to investigate the removal efficiencies of micropollutants using AnMBRs (Xu et al., 2008; Monsalvo et al., 2014). Most of these have focused on high strength organic industrial wastewater such as alcoholdistillery and brewery wastewater (Choo and Lee, 1998; Ince et al., 1998). More recently, there has been a focus on the use of AnMBRs for treating municipal wastewater at centralised (Saddoud et al., 2007; Baek et al., 2010; Martinez-Sosa et al., 2011) and decentralised (Wen et al., 1999; Lew et al., 2009) facilities. The potential to apply AnMBR for municipal wastewater treatment is the development in sewer mining, in which, clean water is extracted from the sewer at source (Butler and MacCormick, 1996; Xie et al., 2013). The remaining wastewater is of much higher wastewater strength and is suitable for anaerobic treatment. However, while information about the removal of TrOCs by AnMBRs is still limited, little is known about the fate of polycyclic musks

(PCMs) during AnMBR treatment. PCMs are commonly used ingredients in personal care and household cleaning products. They have been reported to be resistant to biodegradation under aerobic conditions, which has led to their detection at high concentrations in wastewater treatment plant effluents and in effluent impacted water bodies (Ricking et al., 2003; Yang and Metcalfe, 2006; Clara et al., 2011; Wang and Khan, 2014).

Most PCMs are chiral chemicals. For examples, tonalide (AHTN), phantolide (AHDI), and cashmeran (DPMI) have one chiral centre. Some PCMs such as galaxolide (HHCB) and traseolide (ATII) have two chiral centres. As such, AHTN, AHDI and DPMI may occur in two enantiomeric forms, while HHCB and ATII have four stereoisomers. However, commercial formulations of ATII tend to produce only the 'trans' configurations (Gatermann et al., 2002). Consistent with this, only two enantiomers of ATII were detected in analytical standards and in environmental samples. Our previous research has shown that these chemicals are used and occur in municipal wastewater as an even composition of each of the possible enantiomers (Wang and Khan, 2014). However, it is known that the enantiomeric fractions (EF) of some chiral chemicals may be changed during biological wastewater treatment processes (Hashim and Khan, 2011; Hashim et al., 2011). Accordingly, this investigation was undertaken using an enantiospecific analytical method to enable observation of any changes in EF during AnMBR treatment.

Table 1

Chemical name, common trade names and molecular structures of five PCMs.

Abbreviation	Chemical name	Trade name	Structure	
ННСВ	4,6,6,7,8,8-Hexamethyl-1,3,4,6,7, 8-hexahydrocyclopenta[g]isocromene	Galaxolide, abbalide	$H_{AC} CH_{3}H_{3}C CH_{3}$ $H_{3}C CH_{3}$ $H_{3}C CH_{3}$ $H_{3}C CH_{3}$ $H_{3}C CH_{3}$ $H_{4}CH_{3}H_{3}C CH_{3}$ $H_{3}C CH_{3}$ $H_{3}C CH_{3}$ $H_{3}C CH_{3}$ $H_{3}C CH_{3}$	$H_{3}C CH_{3}H_{3}C H$ $H_{3}C CH_{3}H_{3}C H$ $H_{3}C CH_{3}$ $(4S, 7S)-galaxolide$ $H_{3}C CH_{3}H_{3}C H$ $H_{3}C CH_{3}H_{3}C H$ $H_{3}C CH_{3}H_{3}C H$
AHTN	7-Acetyl-1,1,3,4,4,6-hexamethyl-tetraline	Tonalide, fixolide	(4R, 7S)-galaxolide H_3C H_3C CH_3 H_3C H_3C H_3 H_3C H_3 H_3C CH_3 (3R)-tonalide	(4S, 7R)-galaxolide H_3C CH_3 CH_3 H_3C CH_3 CH_3 H_3C CH_3 CH_3 H_3C CH_3 CH_3 (3S)-tonalide
AHDI	5-Acetyl-1,1,2,3,3,6-hexamethylindane	Phantolide	$H_{3}C H_{3}C $	$(35) \text{ tormate}$ $H_3C \xrightarrow{CH_3} CH_3$ $H_3C \xrightarrow{CH_3} CH_3$ $H_3C \xrightarrow{CH_3} CH_3$ $(25) \text{-phantolide}$
ATII	5-Acetyl-1,1,2,6-tetramethyl-3-isopropylindane	Traseolide	$(2R, 9)$ $H_{3}C \qquad H_{3}C \qquad CH_{3} \qquad $	$(2S, 3S) - traseolide$ $H_{3C} - H_{3} - CH_{3} - CH_{3} - H_{3} - CH_{3} - CH_{3}$
DPMI	1,1,2,3,3,-Pentamethyl-1,2,3,5,6, 7-hexahydro-4H-inden-4-one	Cashmeran	H_3C CH_3 H_3C CH_3 H_3C CH_3 H_3C CH_3 (2S)-cashmeran	H_{3C} H

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