Bioresource Technology 200 (2016) 795–803

Contents lists available at ScienceDirect

Bioresource Technology

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

Evaluation of herbicide (persistent pollutant) removal mechanisms through hybrid membrane bioreactors

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HIGHLIGHTS

- Removal of Ametryn in an MBR is mainly due to biodegradation.
- Adsorption of Ametryn by activated sludge and membrane surface are insignificant.
- 46% of Ametryn is removed due to biodegradation alone by the MBR system.

ARTICLE INFO

Article history:
Received 2 September 2015
Received in revised form 14 October 2015
Accepted 14 October 2015
Available online 21 October 2015

Keywords:
Ametryn
Persistent Organic Pollutants (POPs)
Membrane bioreactor (MBR)
Biodegradation
Adsorption

ABSTRACT

A laboratory-scale membrane bioreactor (MBR) combined with ultraviolet (UV) disinfection and granular activated carbon (GAC) adsorption was researched for over seven months to evaluate the removal efficiencies and mechanisms of a moderately persistent s-triazine herbicide (Ametryn), which is commonly used in Australian sugarcane farmlands. Long-term experiments showed that MBR alone (*15 h hydraulic retention time* (*HRT*)) can remove 65% of Ametryn from its influent which had a concentration of 1–2 mg/L. A batch study was carried out to assess the mechanisms of removal of Ametryn through MBR and found that 0.1186 mg of Ametryn/g-VSS is adsorbed onto sludge particles when 1 mg/L of Ametryn is added to the mixed liquor and showed a 64% removal after 12 h. This experiment confirmed that 99%, 92% and 83% removal of Ametryn could be achieved only from biodegradation, if the MBR maintains a HRT of 7.5, 2.5 and 1.5 days respectively.

1. Introduction

Pesticides and herbicides are categorised as Persistent Organic Pollutants (POPs) which are carbon-based chemical substances that persist in the environment, bio-accumulate through the food web, capable of long-range transport and pose a risk of causing adverse effects to human health and to the environment at large. Herbicide and pesticide contaminated surface water is mainly discharged from the agricultural lands during wet season, and at the same time a significant amount of herbicide and pesticide residues are discharged into the environment through the existing wastewater treatment plants all over the world unintentionally. As it was found that there are many adverse impacts to human life by the consumption of pesticide/herbicide and POP contaminated water for a long time, most of the major drinking water treatment plants have been upgraded in developed countries with suitable advanced treatment methods such as reverse osmosis (RO) or nano-filtration (NF) Yüksel et al. (2013). However, the rapid deterioration to the global ecosystem and to the marine life due to the deposition of these organic pollutants including pesticide and herbicide residues has now been recognised as a major problem but ignored for a long period of time. Hybrid wastewater treatment systems are defined as combination of two or more individual treatment processes (*combination of different biological, adsorption, wetland, or membrane processes*). These hybrid systems perform better than a single treatment process. Recent research studies have found that these hybrid systems could improve the treatment of micropollutants (Li et al., 2011). Membrane bioreactor (MBR) technology, which is a combination of biological and membrane filtration processes, is an ideal example for a popular hybrid wastewater treatment system. Recently, many researchers have studied MBR to improve its performance and to reduce its drawbacks in industrial applications. It is a known fact that MBR is a better treatment process than Activated Sludge Process (ASP) for the treatment of micropollutants and POPs (Radjenović et al., 2008).

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http://dx.doi.org/10.1016/j.biortech.2015.10.041
0960-8524/

A significant number of MBR related research studies have been conducted during the past decade to investigate the treatment performance of wastewater consisting Persistent Organic Pollutants such as pesticides, herbicides and pharmaceuticals. Most of the studies confirmed that MBR is more superior in the treatment of persistent and toxic substances in waste streams compared to that of the other conventional wastewater treatment processes (Fazal et al., 2015). It was found that in most cases, MBR alone cannot be applied for the total removal of such substances and MBR is combined with advanced treatment technologies (hybrid MBR) such as powdered or granular activated carbon (PAC/GAC) adsorption, ozonation and advanced oxidation processes and high pressure membrane systems (Alturki et al., 2010), or modified the system by introducing specific microorganism cultures or adopting improved features (Nguyen et al., 2013). A double membrane system comprising a MBR combined with a NF membrane was investigated by (Wang et al. (2015)) for the treatment of antibiotic production wastewater and found that MBR–NF process produces excellent quality treated water with a high yield of 92 ± 5.6%. A two-stage MBR with anoxic/aerobic reactors (Boonyaroj et al., 2012) operated for 300 days to treat municipal solid waste leachate and recorded a 77–96% removal of phenolic compounds. They found that both biodegradation and adsorption mechanisms were responsible for removal of phenolic compounds from leachate. In another study (Luo et al., 2014) with moving bed bioreactor (MBBR) using polyurethane sponge-growth carrier, it was noted that biodegradation served as a major removal pathway for most of the micropollutants, but some persistent compounds such as carbamazepine, ketoprofen and pentachlorophenol were removed through sorption to sludge.

During this study, a laboratory-scale hybrid MBR combined with Ultra-Violet (UV) disinfection system and a GAC was evaluated for the treatment and operating performance under toxic conditions. Ametryn, which is a commonly used herbicide in the sugarcane farmlands located in the Great Barrier Reef (GBR) catchments – Queensland, Australia, was selected for assessing the hybrid treatment system. Different quantities of Ametryn were mixed to the influent as the target compound. The technical details of Ametryn are described in a different article (Navaratna et al., 2010). The hybrid MBR project was originally commenced in April 2009 and evaluated the performance extensively under distinct environmental/operational conditions. The hybrid MBR was researched for 744 days totally and operated in two different environments (Supplementary Fig. 1); Phase 1 at tropical (Townsville, Queensland) for 530 days (Navaratna et al., 2012a) and Phase 2 at subtropical (Geelong, Victoria) for 214 days. The main objective of this current study (Phase 2) was to consolidate and confirm the previous findings and identify the possible mechanisms of removal of Ametryn through the hybrid MBR system.

2. Methods

2.1. Experimental setup

A hybrid laboratory-scale MBR system, which is shown in Fig. 1, was operated to evaluate the efficiencies and mechanisms of removal of Ametryn from its influents. The hydraulic capacities of the feed tank and the MBR were 40 L and 13 L respectively. A hollow fibre polyethylene (PE) membrane module (pore size 0.4 μm, effective area 0.2 m^2) was submerged in the MBR reactor. Activated sludge (approximately 6000 mg/L) was collected from the Anglesea Wastewater Treatment Plant in Geelong to acclimatise the bioreactor. Same feed recipe in Navaratna and Jegatheesan (2011) was used to prepare the synthetic wastewater and fed to the MBR through the feed tank. Estimated quantities of Ametryn stock solution (160 mg/L) were mixed with synthetic wastewater to obtain the required concentrations (1–4 mg/L). Ametryn stock solution was prepared using the method stated in (Navaratna et al., 2012a). COD concentration of synthetic feed wastewater was maintained around 800 ± 100 mg/L.

The upper limit of the trans-membrane pressure (TMP) was restricted to 20 kPa and the membrane module was cleaned chemically using 3 g/L of NaOCl solution, when the TMP reached to its maximum level, as stated by the manufacturer. Temperature of the mixed liquor was maintained at 20 ± 2 °C using an immersed thermostat in the bioreactor. Compressed air was supplied through a perforated manifold at a rate of 10 L/min (for a hydraulic volume of 13 L) to maintain the dissolved oxygen (DO) concentration around 3.5 ± 1.0 mg/L. MBR sludge was not wasted intentionally to provide a sound environment for slow growing bacteria which is important for the biodegradation of persistent micropollutants. However, small amounts of MBR sludge were collected weekly for experiments and considering this sludge removal, the average sludge retention time (SRT) was estimated as 180 days.

In this study, temperature of the mixed liquor in the bioreactor was maintained at 20–21 °C. MBR was adjusted to operate at a uniform flow rate of 20 L/day with intermittent suction (12 min ON and 3 min OFF) of permeate. UV disinfection unit, which consists of an UV-C lamp (wavelength: 254 nm; total UV dosage: 6.602 W s/cm^2; 20.3 W) and a stainless steel body (made at UVS

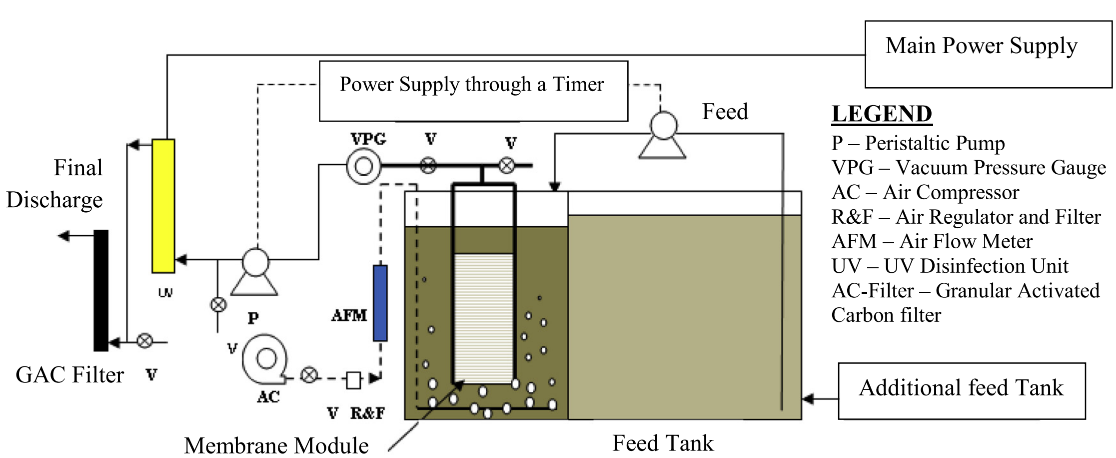

Fig. 1. Experimental setup.