



Research article

Simultaneous removal of atrazine and organic matter from wastewater using anaerobic moving bed biofilm reactor: A performance analysis



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ABSTRACT

In this study, an anaerobic moving bed biofilm reactor (AMBBR) was designed to biodegrade atrazine under mesophilic (32 °C) condition and then it was evaluated for approximately 1 year. After biofilm formation, acclimation, and enrichment of microbial population within the bioreactor, the effect of various operation conditions such as changes in the concentration of influent atrazine and sucrose, hydraulic retention time (HRT), and salinity on the removal of atrazine and chemical oxygen demand (COD) were studied. In optimum conditions, the maximum removal efficiency of atrazine and COD was 60.5% and 97.4%, respectively. Various models were developed to predict the performance of atrazine removal as a function of HRT during continuous digestion. Also, coefficients kinetics was studied and the maximum atrazine removal rate was determined by Stover - Kincannon model ($r_{max} = 0.223 \text{ kg}_{ATZ}/\text{m}^3\text{d}$). Increasing salinity up to 20 g/L NaCl in influent flow could inhibit atrazine biodegradation process strongly in the AMBBR reactor; whereas, the reactor could tolerate the concentrations less than 20 g/L easily. Results showed that AMBBR is feasible, easy, affordable, so suitable process for efficiently biodegrading toxic chlorinated organic compounds such as atrazine. There was no accumulation of atrazine in the biofilm and the loss of atrazine in the control reactor was negligible; this shows that atrazine removal mechanism in this system was due to co-metabolism.

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1. Introduction

Many chemical xenobiotic compounds entering the environment are aromatic compounds that used in industry and agriculture frequently (Boopathy, 2017). These compounds are entered into soil and aquatic environment through various resources and because of their specific characteristics, they accumulate in the environment (Pérez-Leblic et al., 2012). Atrazine (2-chloro-4-

ethylamino-6-isopropylamino-1,3,5-triazine) as a kind of s-triazine is one of the most commonly used herbicide in the worldwide (Baghapour et al., 2013). It's known as possible human carcinogens (Group 2B) and persistent organic pollutants (POPs) in the environment (Nasseri et al., 2014). Due to atrazine widespread use in several industrial and agricultural activity and physicochemical properties, is frequently detected in surface and subsurface water resources (Baghapour et al., 2013; Boopathy, 2017). United state environmental protection agency (EPA) and world health organization (WHO) have established the maximum contaminant level (MCL) of atrazine in drinking water as 3 and 2 $\mu\text{g/L}$, respectively. Similarly, in Iran the institute of standards and industrial research of Iran (ISIRI) has set a limit for atrazine as 2 $\mu\text{g/L}$ (Baghapour et al., 2013; Nasseri et al., 2014).

In order to protect human health and environment from the adverse effects of atrazine, the effluents containing it needed to be treated with suitable methods (Baghapour et al., 2013). Atrazine treatment is mainly carried out by various physicochemical methods. These methods of treatment are pricey, complicated and produce a lot of toxic intermediates (Boopathy, 2017). The biological method breaks and transforms the complex compounds to simple compounds, such as carbon dioxide, water, nitrogen and etc. (Nasseri et al., 2014). Atrazine removal thru biological methods is also employed both aerobically as well as anaerobically, but very few studies have done on atrazine biodegradation in anaerobic conditions versus aerobic conditions (Boopathy, 2017; Nasseri et al., 2014). Aerobic bioprocesses have been used to biodegrade atrazine but they have high aeration cost, are complex for operation and maintenance, generate high amount of biomass which poses economic and environmental challenges (Baghapour et al., 2013; Derakhshan et al., 2016). In anaerobic treatment process, converts the organic pollution to a small quantity of sludge and can produce a large amount of biogas (Ghosh and Philip, 2004).

In recent decades, anaerobic moving bed biofilm reactors (AMBBRs) technology in wastewater treatment of municipal, industrial, and agricultural has been used successfully (Wang et al., 2009). In AMBBRs reactor, the growth of microorganisms occurs on moving solid carriers resulting in the formation of stable biofilm (Kawan et al., 2016; Kermani et al., 2008). The biofilm carriers used in AMBBR usually are with density less than water and they can move in the fluid inside the reactor (di Biase et al., 2017; Kermani et al., 2008). The main advantage of AMBBRs along with increasing microbial activity is reduction of head losses, no filter channeling, operated easily, without biomass loss or temperature dependence, strong tolerance to loading impact and relatively smaller reactor (di Biase et al., 2017; di Biase et al., 2016; Kawan et al., 2016). Also, they have a simple design and not using a mechanical mixing, and therefore are economically affordable (di Biase et al., 2017; Wang et al., 2009). For those reasons, AMBBRs are appropriate for the treatment of a variety of wastewaters containing toxic and xenobiotic compounds (di Biase et al., 2017; Wang et al., 2009).

The main objective of this study was to evaluate the efficiency of AMBBR reactor as well as HRTs in atrazine and COD removal at different influent concentrations in order to examine the capability of this reactor in different loading rates. Secondly, we aimed to explore the effect of salinity on biodegradation of atrazine as a model compounded of chlorinated aromatic xenobiotic that are widely used in agriculture.

2. Materials and methods

2.1. Startup and operation of bioreactors

In this pilot-scale study, the effects of the input atrazine

concentration, HRT, and salinity were evaluated in an AMBBR reactor. As indicated in Fig. 1S, the AMBBR reactor is used that is made of Plexiglas (diameter = 20 cm, height = 50 cm, freeboard = 2 cm) with a working volume of 15 L. 70% of the reactor volume were filled with prepared media (diameter = 2 cm, height = 1 cm, and relative density = 0.98). The area of biofilm carriers available for pre-formed biofilm was about 410 m^2/m^3 (Baghapour et al., 2013; Nasseri et al., 2014). Continuous mixing reactor was done by a submerged pump installed in the reactor floor. Mixing time was adjusted for 2 to 8 times per hour based on organic loading rate increase during experiments. The duration of mixing lasted for 1.30 min. This duration could animate the bed in the reactor. Hydraulic retention times (HRTs) was set by controlling the flow rate of influent synthetic wastewater. To discharge the possible accumulated sludge, a drain valve was used at the bottom of the bioreactor. To control confounding variables, fluctuations in raw wastewater, and the system best operation, synthetic wastewater was used. pH level of raw sewage was set about 7.5 ± 0.1 using sodium bicarbonate (0.5 mol/L). Bicarbonate can act as a buffer to maintain pH and also as an electron receptor for hydrogenotrophic methanogenesis (Delgado et al., 2012; Paulo et al., 2003). The water needed for synthetic wastewater was provided from tap water. The synthetic wastewater had the following composition: $\text{NaHCO}_3 = 20 \text{ mg/L}$, $\text{MgSO}_4 \cdot 7\text{H}_2\text{O} = 5 \text{ mg/L}$, $\text{KH}_2\text{PO}_4 = 5 \text{ mg/L}$, $\text{CaCl}_2 \cdot 2\text{H}_2\text{O} = 5 \text{ mg/L}$, $(\text{NH}_4)_2\text{HP}_2\text{O}_4 = 50 \text{ mg/L}$, $\text{C}_{12}\text{H}_{22}\text{O}_{11}$ variable (300–1100 mg/L), atrazine variable (0.1, 1 and 10 mg/L) and trace element were: $\text{FeSO}_4 \cdot 7\text{H}_2\text{O} = 0.2 \text{ mg/L}$, $\text{ZnCl}_2 = 0.1 \text{ mg/L}$, $\text{CoCl}_2 = 0.1 \text{ mg/L}$, $\text{NiCl}_2 = 0.1 \text{ mg/L}$, $\text{CuSO}_4 \cdot 5\text{H}_2\text{O} = 0.001 \text{ mg/L}$, $\text{H}_3\text{BO}_3 = 0.2 \text{ mg/L}$, $\text{MnSO}_4 = 0.5 \text{ mg/L}$ (Nasseri et al., 2014). According to previous studies, the maximum removal efficiency of atrazine biodegradation occurs at 32 °C (Baghapour et al., 2013; Nasseri et al., 2014). In this study, the temperature was set at 32 °C in the feeder tank by an electric heater. Although the compositions of synthetic wastewater were completely soluble in water, a small electric mixer was used to return sewage from the floor to the top of the tank in order to prevent quality changes in the wastewater happened due to storage. The electric mixer rotated all the wastewater every 15 min (Fig. 1S). The operational scheme of the system for 9 phases is presented in Table 1.

2.2. Preparing and installing the reactor

To set up the system and initiate biological adaptation stage, a filter column with an approximate volume of 15 L as pilot was seeded by the mesophilic anaerobic sludge digester bacteria collected from Shiraz urban wastewater treatment plant which had no operational problem, the concentration suspended solids was 30 g/L, and VSS/TSS ratio was 0.8; the remained space inside the bioreactor was filled with the synthetic wastewater made out of a chemical oxygen demand (COD) of 10,000 mg/L. For more acclimation of microorganisms at the presence of atrazine in the environment, the reactor was fed with synthetic wastewater (15 L) containing 0.1 mg/L atrazine. The effluent was re-circulated to the influent while the concentration of atrazine and COD were evaluated in the re-circulated solution. At the first startup of the reactor, organic loading rate (OLR) in AMBBR was 0.5 g COD/L d. Due to methanogenic bacteria grow more slowly than acidogenic bacteria, OLR should be reduced at the reactor startup until organic acids produced by the fermentation bacteria that have rapid growth do not decrease the buffering system (Nasseri et al., 2014). Afterwards, OLR was gradually increased until it reached 2 g COD/L d. It was assumed that when the concentration of COD in the re-circulated solution increased to more than 95% degradation, acclimation is attained. To enrich the population of the microorganisms capable of

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