



Research article

Biodegradation of atrazine from wastewater using moving bed biofilm reactor under nitrate-reducing conditions: A kinetic study



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ABSTRACT

In this study employed an anoxic moving bed biofilm reactor (AnMBBR) to evaluate the effects of hydraulic and toxic shocks on performance reactor. The results indicated a relatively good resistance of system against exercised shocks and its ability to return to steady-state conditions. In optimal conditions when there was the maximum rate of atrazine and soluble chemical oxygen demand (COD) removal were 74.82% and 99.29% respectively. Also, atrazine biodegradation rapidly declines in AnMBBR from $74\% \pm 0.05$ in the presence of nitrate to 9.12% only 3 days after the nitrate was eliding from the influent. Coefficients kinetics was studied and the maximum atrazine removal rate was determined by modified Stover & Kincannon model ($U_{\max} = 9.87 \text{ gATZ/m}^3\text{d}$). Results showed that AnMBBR is feasible, easy, affordable, so suitable process for efficiently biodegrading toxic chlorinated organic compounds such as atrazine. Also, its removal mechanism in this system is co-metabolism.

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1. Introduction

Large amounts of wastewater are produced due to human activities. All effluent discharging into the environment requires treatment to meet effluent disposal standards (Qasim, 2017). So far, various biological and physicochemical methods of wastewater treatment have been designed (Boujelben et al., 2017). To control waste streams of organic contaminants, biological treatments are

generally more acceptable than the physicochemical ones, as they are more reliable and environmentally friendly, with high compatibility and removal efficiency. Other advantages of these treatments include simple design, operation, and construction, as well as relatively limited use of energy and it is a cost-effective method (Ahmed et al., 2017; An et al., 2018). Several studies have assessed the feasibility of biodegradation of organic compounds under anoxic conditions (Akmirza et al., 2017; Kobayashi et al., 2017). In biodegradation of organic pollutants, anoxic bioreactors can overcome the shortcomings of aerobic (mainly economic) and anaerobic (low removal) processes (Dorival-García et al., 2013; Moussavi and Ghorbanian, 2015). In respiratory metabolism, more energy is released in comparison to anaerobic fermentation. Consequently, respiratory metabolism is favored for biodegradation

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of organic contaminants (Gupta et al., 2016; Moussavi et al., 2016; Mulla et al., 2018).

With population growth in recent years and consequently the shortage of farmland and the destruction of crops by pests, using pesticides such as atrazine (ATZ) increased significantly (Derakhshan et al., 2018b, 2018c). Atrazine (2-chloro-4-ethylamino-6-isopropylamino-s-triazine) as a kind of s-triazine herbicides is one of the most common herbicides in the world. Atrazine as a poison is known as possible human carcinogens (Group 2B) (Baghapour et al., 2013; Derakhshan et al., 2016). Atrazine is regarded as an environmental pollutant due to its slow biodegradation and high potential to cause surface water and groundwater. Typically, atrazine half-life is variable but can last up to 360 days and even more (Nasseri et al., 2014).

Nitrogen compounds are nutrients limiting water systems and entering aquatic ecosystems through point and non-point sources. They can cause a variety of problems, including a reduction in concentrations of dissolved oxygen in water sources which can lead to the death of fish (Derakhshan et al., 2018a; Parvizishad et al., 2017). Moreover, it can bring about eutrophication and other problems such as increased costs of treatment and algae toxins production with potentially fatal effects for the human (Boeykens et al., 2017; Wu et al., 2016). There are contradictory results about the effect of nitrogen presence compounds on the process of removing atrazine in soil and aquatic environments. Some studies have revealed that nitrogen can alert the microbial process and subsequently can reduce atrazine degradation (García-González et al., 2003; Laursen and Carlton, 1999; Pathak and Dikshit, 2012). On the other hand, some investigators have held that nitrogen compounds have no role in atrazine removal process indicating the interference role of nitrogen compounds for the removal of atrazine by microorganisms (Ghosh and Philip, 2004; Wen et al., 2016).

There are combinations of suspended and fixed-film process in aerobic and anaerobic bioreactors in atrazine removal from waste streams (Baghapour et al., 2013; Nasseri et al., 2014). However, no information is available on atrazine biodegradation (removal rate and efficiency) in hybrid bioreactors under anoxic conditions. Overall, the biological treatment changes dangerous toxins into nontoxic compounds (Qasim, 2017). However, for using the advantages of anoxic atrazine biotreatment, further research should be performed to determine the best configuration and operation protocols of reactors and design novel high-rate bioreactors with improved efficacy and treatment of atrazine streams. The main objective of this study was primarily to determine the efficiency of anoxic moving bed biofilm reactor (AnMBBR) under nitrate-reducing conditions for removing considered pollutants from wastewater. Secondly, this study evaluated the interference effects of nitrogen sources on atrazine biodegradation as chlorinated herbicides 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 the absence or presence of nitrates were evaluated in an AnMBBR reactor. As indicated in Fig. 1S, the AnMBBR reactor is used that is made of Plexiglas (diameter = 20 cm, height = 50 cm, freeboard = 2 cm) with a working volume of 15 L. 60% 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 m²/m³ (Derakhshan et al., 2018c). 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 experimental. The duration of mixing lasted for 1.25 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 sewage, and the system best, synthetic wastewater was used. pH level of raw sewage was set about 7.5 ± 0.1 using sodium bicarbonate (0.5 mol/L). The water needed for synthetic wastewater was provided from water supply network. The synthetic wastewater had the following composition: NaHCO₃ = 20 mg/L, MgSO₄·7H₂O = 5 mg/L, KH₂PO₄ = 5 mg/L, CaCl₂·2H₂O = 5 mg/L, FeSO₄·7H₂O = 0.2 mg/L, CuSO₄·5H₂O = 0.001 mg/L, H₃BO₃ = 0.2 mg/L, MnSO₄ = 0.5 mg/L, (NH₄)₂HP₂O₄ = 50 mg/L, Sucrose = 300 mg/L, atrazine variable = 0.01, 0.1, 1 and 10 mg/L (Ghosh and Philip, 2004; Nasseri et al., 2014).

Based on the previous studies, the maximum removal efficiency of atrazine biodegradation occurs at 32 °C (Baghapour et al., 2013; Nasseri et al., 2014). Accordingly, 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.

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 aerobic bacteria collected from Shiraz urban wastewater treatment plant which had no operational problem including bulking, rising, and pinpoint phenomena, 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. Activated sludge was used as seed because it poses heterotrophic microbial populations. 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. It was assumed that when the concentration of COD in the re-circulated solution increased to more than 90% degradation, acclimation is attained.

To enrich the population of the microorganisms capable of degrading atrazine, the recycle solution was replaced with a fresh one containing 0.1 mg/L atrazine, and as mentioned before the effluent was re-circulated to the influent. This procedure was repeated for 5 consecutive cycles to ensure atrazine-degrading and microbial population increase. To ensure the microbial activity in this phase, surface cultivation of mixed liquor suspended solids (MLSS) in the bioreactor was frequently performed in a mineral salt medium (MSM) solution containing atrazine. The MSM preparation method was conducted according to the previous study (Derakhshan et al., 2018c). Also, to evaluate the growth of biofilm on media in this phase, Scanning Electron Microscope (SEM) was applied. This stage lasted for 67 days. The external wall of the reactor was covered with the aluminum foil in order to avoid the confounding effect of light (photo-catalyst) and algae growth. In addition, a control pilot with the same physical characteristics of the main pilot was utilized to increase the accuracy and omit the effect of the confounding factors. A brief explanation is brought in the Supplementary data.

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