



## Removal efficiency of penicillin G in horizontal subsurface flow wetlands

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**ABSTRACT:** Antibiotics are potential pollutants that represent an important environmental problem because of their toxic effects on the food chain and aqueous streams. The goal of this study was to determine the efficiency of a horizontal subsurface flow constructed wetland for a pharmaceutical pollutants antibiotic penicillin G. This study used constructed wetland pilot system for removal of penicillin G in artificial wastewater. In this study, the effects of initial concentration of wastewater, hydraulic retention time, and reed on the pollutant removal efficiency were investigated. The data was analyzed using the central composite design which is the most commonly used response surface methodology design. 30 Samples of wastewater were taken from the output of constructed wetlands subsurface and tested in the laboratory-based on the standard reference method for experiments in water and wastewater. The results showed that reed, and retention time, have a direct relationship, and enhance them to increase efficiency. The initial concentration of wastewater is inversely related to removal efficiency. In the constructed wetland, the removal efficiency for 72 hours and different input concentrations was between 94.17% and 73.61%. Based on the study results, it can be stated that subsurface constructed wetland can remove the maximum concentration of hospital wastewater and even double this concentration with efficiency up to 90 percent, and it can be used as a proper treatment system for removal of penicillin G.

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## 1. INTRODUCTION

Water resources are contaminated with dangerous levels of antibiotics which have an adverse impact on the environment and human health [1]. The most appropriate wastewater treatment system is a system capable of operating with local workers, with economic benefits, ease of construction and management, and low environmental impact [2]. Constructed wetlands (CWs) are used as an environmentally friendly wastewater treatment technology that provides multiple economic, ecological, technical, and societal benefits. CWs are very effective in removing organic matter, toxic metals, and nutrients from wastewaters. Among the CWs, the subsurface flow CW significantly removed the pollutants more efficiently than the surface flow CW [3, 4]. In this study horizontal subsurface flow constructed wetland (HSSF-CW) was employed to remove penicillin G antibiotic from wastewater, due to the high efficiency of HSSF-CW in the removal of hard degradable materials and high hydraulic loading rate. Response surface methodology (RSM) was used to investigate the effects of different operating conditions on the removal of penicillin G by HSSF-CW.

## 2. METHODOLOGY

In this study, two different HSSF-CW were constructed

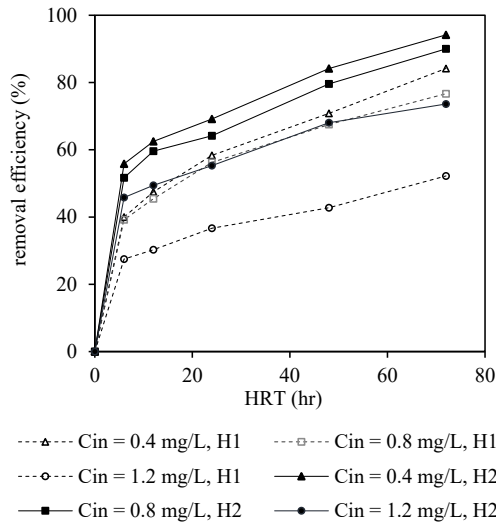
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at the University of Birjand, one planted with phragmites australis (H2) and the other one without plant (H1) as a control. The dimensions of HSSF-CWs were 400 cm in length, 150 cm in width, and 55 cm in depth, with a surface area of 60000 cm<sup>2</sup> and a slight slope of 1.5%. The sewage level was 5 cm below the surface of the media. The coarse-grained aggregates with the size of 5 cm and perforated PVC pipes were put into the inlet and outlet zones in each cell in order to produce a uniform distributed flow. The remaining 360 cm length of two cells was filled with fine sand. CW was fed with well water for 16 weeks in order to grow the root and rhizome of the reed and adapt to the conditions.

In this study, penicillin G concentration in effluent from both CWs for wastewater with concentrations of 0.4, 0.8, and 1.2 mg penicillin G/L at hydraulic retention time (HRT) of 6, 12, 24, 48, and 72 h was determined. Concentrations of penicillin G were measured by high-performance liquid chromatography (HPLC) series 1200 system from Agilent Technologies.

In this study, five levels of central composite design (CCD) with three factors (Initial concentration of penicillin G, HRT, reed) were selected as an experimental design to estimate the main effects and interactions on the concentration of penicillin G in CWs. The effect of independent variables on the response was assessed using analysis of variance (ANOVA) and a p-value of <0.05 was used as the results significance





**Fig. 1. The effect of initial concentration of penicillin G, HRT and reed on the penicillin G removal efficiency**

level [5].

### 3. RESULTS AND DISCUSSION

The results of the study showing the effect of HRT and initial concentration of penicillin G ( $C_{in}$ ) on penicillin G removal efficiencies in two types CWs (H1, H2) were presented in Fig. 1. According to Fig. 1, the positive effect of reed on penicillin G removal in CW increased with increasing inlet pollution concentration. Pollutant removal efficiency at HRT of 72 h in the planted constructed wetland (H2) compared to non-planted constructed wetland (H1) for initial concentrations of 0.4 mg/L, 0.8 mg/L, and 1.2 mg/L increased 11.88%, 17.39%, and 41%, respectively.

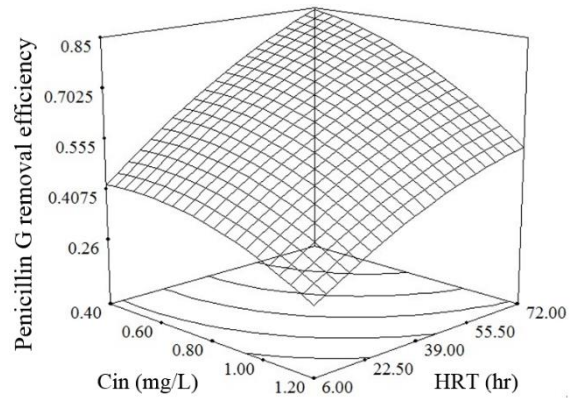
The models for penicillin G removal in planted CW ( $Y_1$ ) and non-planted CW ( $Y_2$ ) were significant by the F-test at the 95% confidence level. The following fitted regression models (equations in terms of actual values) were used to investigate the effects of initial concentration of penicillin G ( $C_{in}$ ) and HRT on pollutant removal efficiency in planted CW (Equation 1) and non-planted CW (Equation 2).

$$Y_1 = 0.172 + 0.016 \times C_{in} - 3.512E-3 \times HRT - 2.88E-3 \times C_{in} \times HRT + 0.506 \times C_{in}^2 + 2.585E-5 \times HRT^2 \quad (1)$$

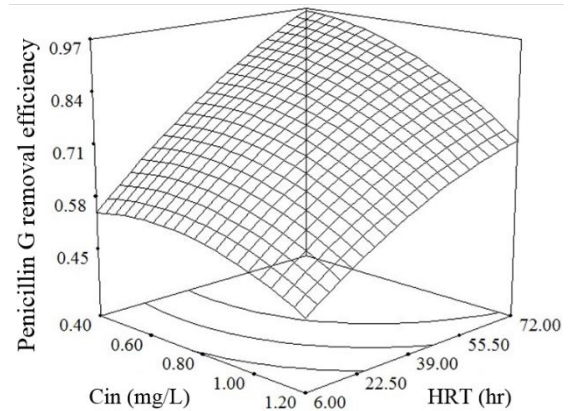
$$Y_2 = 0.239 - 0.224 \times C_{in} - 3.735E-3 \times HRT - 3.52E-3 \times C_{in} \times HRT + 0.506 \times C_{in}^2 + 2.585E-5 \times HRT^2 \quad (2)$$

Comparisons of Equations 1 and 2 showed that the Initial concentration of penicillin G decreased from 1.2 mg/L to 0.6 mg/L for the effluent concentration from H1 and H2 at HRT of 59 h and 14h, respectively.

Three-dimensional plots were employed to graphically show the effects of the independent variables and their interactions on the penicillin G removal efficiency in H1 (Fig.



(a)



(b)

**Fig. 2. Response surface plots of penicillin G removal efficiency as a function of HRT and initial concentration of penicillin G ( $C_{in}$ ) in (a) non-planted and (b) planted CW**

1.a) and H2 (Fig. 2.a).

Comparisons of Figs. 1.a and 1.b showed that the effect of HRT on the reduction of pollutant concentration in the planted CW (H2) is greater than that in non-planted CW (H1). These results indicate that the plant in CW increased the efficiency of adsorption and biological treatment of penicillin G. These results have been confirmed in other studies [6, 7].

The desired goal of the model is to maximize penicillin G removal efficiency. To confirm the accuracy of the optimization, duplicate verification experiments for each condition was carried out. The slight difference between the predicted and actual values of the response successfully verified the optimum points determined by RSM. It implies that the strategy to optimize the constructed wetland conditions and to obtain the maximal penicillin G removal efficiency by RSM in this study is successful.

### 4. CONCLUSIONS

The results indicate that penicillin G concentration was effectively removed from synthetic wastewater by horizontal subsurface-flow constructed wetland. The *Phragmites australis* could enhance the penicillin G removal performance

of HSSF-CW. The *Phragmites australis* adapted extremely well to wetland conditions. The result shows that the shoots reached their maximum height within 4 months after planting. The maximum heights of *Phragmites australis* during spring and summer was  $100 \pm 5$  cm. Determination of the optimum conditions for the treatment of penicillin G in HSSF-CW was another objective of this study. The desirability function was used to find the optimum conditions. To validate the optimization of conditions, four experiments with optimized conditions were conducted in duplicate and their mean values were used to confirm the results from the analysis of the RSM. The result shows that the validity of the RSM optimization.

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