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Treatment of Drainage Water Containing Pharmaceuticals Using Duckweed (*Lemna gibba*)

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Abstract

The potential use of duckweed (*Lemna gibba*) system to remove pharmaceuticals from drainage water (DW) was investigated. The system achieved removal of 66.12±1.4%, 47.50±2.0% and 66.50±1.7% for 1000 µg/L of acetaminophen (ACT), diclofenac (DFC), and progesterone (PRG), respectively. The uptake rate (k_{r1}) of ACT, DFC, and PRG was significantly decreased from 0.884±0.12 to 0.199±0.02, from 0.528±0.02 to 0.152±0.01 and from 0.719±0.03 to 0.264±0.01 at increasing the initial concentration from 1 to 1000 µg/L, respectively. Moreover, the duckweed uptake contributed the major removal pathway followed by duckweed sorption and microbial degradation for ACT, DFC and PRG.

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Keywords: Drainage water; Pharmaceuticals; Duckweed; Removal mechanism; Uptake

1. Introduction

Drainage water (DW) reuse is the most promising immediate and economically attractive option to secure more water for agriculture sector in Egypt. However, the reuse of DW as a reliable resource is limited due to its quality, which in turn determines the quantity that can be used for irrigation purposes [1]. Unfortunately, DW in Egypt suffers from different sources of pollution especially from emerging organic pollutants, including pharmaceuticals, personal care products, and pesticides. Pharmaceuticals are used extensively in human and veterinary medicine to

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prevent illness and also as growth promoters in livestock and fish farming as well as in agriculture[2]. The increasing worldwide consumption of medicines provides a continuous release of these substances or their metabolites to the environment. However, since wastewater treatment plants (WWTPs) are not designed for removing pharmaceuticals and their metabolites, many of them are released into surface waters[3]. These compounds are found in surface waters at low concentrations in the ng/L to µg/L range, despite their low concentrations, their eco-toxicological effects are unpredictable because of the large number of compounds possibly present[4]. Therefore, treatment of DW containing pharmaceuticals is necessary for producing an effluent quality complying for reuse in agricultural purposes.

Conventional chemical and biological treatment processes such as reverse-osmosis, membrane bioreactors, activated carbon, photocatalytic oxidation technology, can effectively remove pharmaceuticals at high concentrations [4, 5]. However, so far low-cost technology for removal of pharmaceuticals at low concentration does not exist. Development of a cost-effective, less complex and environmentally friendly method to eliminate pharmaceuticals at low concentration levels from the aquatic environment is imperative. Phytoremediation using submerged, floating, or emergent macrophytes is based on utilizing natural processes, and it represents an effective, low-cost technology, less energy consuming and preferred cleanup technology for the treatment of contaminated water[1]. From literature, one of the most promising aquatic families in the phytoremediation is duckweeds (*Lemnaceae*) which is widely distributed in the aquatic ecosystems in Egypt[6, 7]. The duckweeds are small floating aquatic macrophytes which grow on the nutrient rich surface and in fresh waters[7]. Phytoremediation of contaminated water using duckweed species is promising due to its ability to grow at wide ranges of temperature, pH, and nutrient (N&P) level in areas where land is available for its application[8-9]. The ability of duckweed to remediate organic chemicals, including pharmaceuticals was investigated by Richards[10] and Schroder et al.[11]. Uptake of organic chemicals by duckweed relies on a complex combination of abiotic and plant-driven processes[12]. Therefore, the objectives of the present research are to 1. Investigate the performance of duckweed (*Lemna gibba*) based treatment system for removal of different concentration of pharmaceuticals from drainage water and 2. Study the removal mechanism of pharmaceuticals from DW through the duckweed based treatment system.

2. Material and methods

2.1. Duckweed

The duckweed (*Lemna gibba*) used in the present study was harvested from pilot plant treating DW under local environmental conditions. The collected duckweed species was initially washed with tap water for 10 minutes to remove debris. The duckweed was cultured and used for batch experiments. The initial density of the duckweed was constant for all experiments at a level of 50 mg/cm^2 (wet weight).

2.2. Experimental set-up and calculations

Three pharmaceuticals namely acetaminophen (ACT), diclofenac (DFC) and progesterone (PRG) were examined at different initial concentrations (i.e., 1, 50, 500 and 1000 µg/L).Four clear glass reactors wrapped with opaque material with a capacity of 1 liter containing active duckweed (ADW), inactive duckweed (IDW), macerated duckweed (MDW) and no-duckweed (No-DW) were parallel utilized to assess the removal mechanism of pharmaceuticals by plant-associated processes (Table 1).

Sorption experiments were conducted using IDW where the populations were inactivated prior to experimentation through exposure to darkness in sealed reactors for a period exceeding 12h. Macerated duckweed (MDW) reactors were utilized to monitor the degradation of pharmaceuticals by microbial communities where the duckweed was macerated in a blender for 2 minutes prior starting the experiments. Reactors containing no-duckweed were utilized to document physicochemical removal via volatilization, photo-degradation, and presumably hydrolysis[12]. All batches were conducted in triplicate for a period of 10 days. All reactors placed beneath constant artificial light for 16 h at a photosynthetic photon density of 115±8 µmol/m^2.s and kept at a temperature of 23±2 ^{o}C.

The pharmaceuticals removal was calculated based on the mass fraction removed from the liquid phase, Eq.1.

$$R\% = \frac{(M_{Initial} - M_{Liquid}) * 100}{M_{Initial}} (1)$$

Where $R\%$ is pharmaceutical removal ratio; $M_{Initial}$ is pharmaceutical initial concentration (µg/L); and M_{Liquid} is the measured concentration of pharmaceutical in mixture (µg/L).

The uptake rate of pollutants by aquatic plants is described by the pseudo first-order reaction equation[12] as follows:

$$C_t = C_0 . e^{-kr.t} (2)$$

Where C_t is the effluent concentration of the pharmaceuticals at t time; C_0 is the initial concentration of pharmaceutical compounds; and k_r is the first-order removal rate constant (d^{-1}). First-order uptake rate coefficients are dependent on plant mass and initial concentration of the pharmaceutical compound[12-13].

Table 1. Experimental set-up for assessment the mechanism removal of pharmaceuticals from DW using duckweed plants

Reactor system no.	Experiment No.	Initial concentration (µg/L)	Conditions in reactor	Treatment Abbreviation	Main presumed removal mechanism
1	1.a 1.b 1.c 1.d	1 50 500 1000	400 ml DW+ active duckweed + light + mixture of pharmaceuticals*	ADW	Plant uptake
2	2.a 2.b 2.c 2.d	1 50 500 1000	400 ml DW+ inactive duckweed + dark + mixture of pharmaceuticals*	IDW	Sorption
3	3.a 3.b 3.c 3.d	1 50 500 1000	400 ml DW+ mactreated duckweed + light + mixture of pharmaceuticals*	MDW	Plant-associated microbial degradation
4	4.a 4.b 4.c 4.d	1 50 500 1000	400 ml DW+ no duckweed + light + mixture of pharmaceuticals*	No-DW	Volatilization + photodegradation

* Pharmaceutical mixtures (ACT, DFC and PRG)

2.3. Sampling and analytical measurements

Sample of 2 ml for each experiment were collected at contact time of 0, 0.25, 0.5, 1, 2, 4, 6, 8 and 10 d. The samples were filtered by micro syringe filters (0.2 µm) prior analysis using SHIMADZU HPLC (C-18 phenomenex direct phase column), degasser (20A5), pump (LC-20AT), and prominences Diode Array Detector (SPD-M20A). The mobile phase was 60% 0.025M KH_2PO_4 buffer solution in ultra-pure water and 40% acetonitrile at a flow rate of 0.50 ml/min and temperature of 60˚C.

3. Results and discussion

Assessment the efficiency of duckweed (*Lemna gibba*) plant for removal of pharmaceuticals (ACT, DFC and PRG) at different initial concentrations i.e. 1, 50, 500, 1000 µg/L from DW was extensively studied. The mechanism removal including plant uptake, sorption, plant-associated microbial degradation, volatilization and photo-degradation of the individual pharmaceuticals was investigated.

3.1. Acetaminophen (ACT)

The results showed that the ADW treatments contribute the highest aqueous depletion of ACT followed by the IDW, MDW and No-DW reactors. Greater depletion rates of ACT in ADW reactors indicated that duckweed either directly contributed to ACT removal[12]. Tront and Saunders[14] found that Duckweeds directly contribute to pollutant removal through active uptake, which is strongly correlated with plant activity. Fig. 1 shows the removal of different initial concentration of ACT from drainage water by duckweed uptake in ADW treatments. As depicted in Fig. 1, as contact time increased from 0.25 to 4d., the concentration of ACT in the treated water significantly reduced. The aqueous depletion of ACT was 84.51±3.0%, 80.25±1.5%, 73.80±2.0% and 60.25±1.0% after 4 d. of treatment for ACT of initial concentration of 1, 50, 500, 1000 µg/L, respectively. While, the removal ratios increased to 92.2±1.9%, 90.0±2.1%, 79.7±1.6%, and 66.12±1.4% after 10 d. of treatment for initial concentration of 1, 50, 500, 1000 µg/L, respectively. This affords a more intimate and longer contact time between the duckweed and the substrate enhanced the uptake of ACT. This indicates that contact time exceeded 4d. is essential for complete uptake of ACT by duckweed.The removal efficiency for ACT is substantially lower than that (99%) obtained by Farrel[15] in microcosm study using duckweed for treating waste water containing ACT after contact time of 4 d. The current duckweed system achieved removal of ACT (92.2-66.12%) which is comparable to removal (100%) by External Loop Airlift Membrane Bioreactor (ELAMBR)[17].

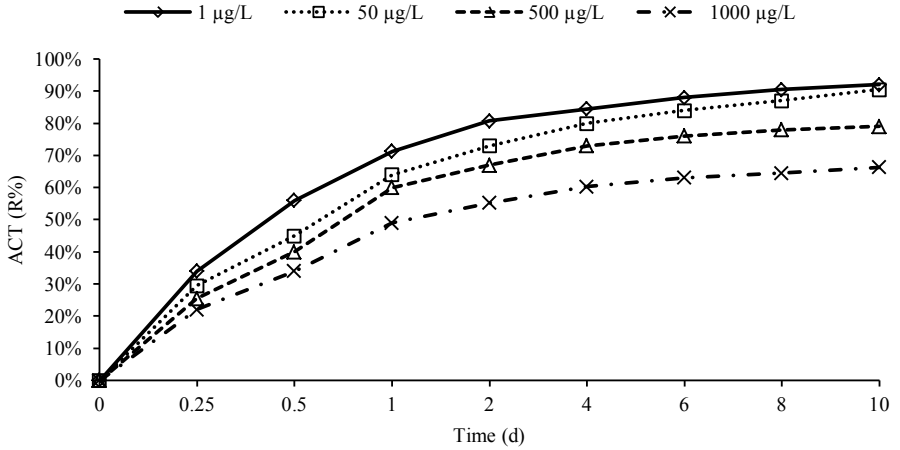

Fig.1 Removal of ACT of initial concentration of 1, 50, 500, and 1000 µg/L in ADW treatments.

The results presented in Table 2 show that the k_r (d^{-1}) values for ACT in the ADW and IDW reactors exceeded those operated with MDW and No-DW conditions. The ADW contributed the highest aqueous depletion of ACT followed by the IDW, MDW and No-DW reactors.

Table 2. Mechanism removal rate of ACT at different initial concentrations

	ACT				
	Treatment	ADW (k_{r1})	IDW (k_{r2})	MDW (k_{r3})	No-DW (k_{r4})
1µg/L	Measured k_r (d^{-1})	0.884±0.12	0.319±0.05	0.196±0.02	0.167±0.01
	Simulated k_r (d^{-1})	1.218±0.16	0.458±0.04	0.252±0.01	0.207±0.01
	R^2	0.98	0.91	0.84	0.82
50 µg/L	Measured k_r (d^{-1})	0.555±0.09	0.221±0.03	0.155±0.01	0.141±0.01
	Simulated k_r (d^{-1})	0.941±0.07	0.288±0.02	0.185±0.01	0.164±0.01
	R^2	0.94	0.86	0.84	0.84
500 µg/L	Measured k_r (d^{-1})	0.297±0.01	0.105±0.01	0.096±0.01	0.084±0.01
	Simulated k_r (d^{-1})	0.547±0.02	0.121±0.01	0.252±0.03	0.089±0.01
	R^2	0.78	0.756	0.811	0.911
1000 µg/L	Measured k_r (d^{-1})	0.199±0.02	0.137±0.01	0.087±0.01	0.054±0.01
	Simulated k_r (d^{-1})	0.282±0.01	0.166±0.01	0.103±0.01	0.057±0.005
	R^2	0.76	0.75	0.58	0.81

The removal rate of 0.884±0.12, 0.319±0.05, 0.196±0.02 and 0.167±0.01 d^{-1} were found for ADW, IDW, MDW, and No-DW treatments, respectively. Based on these results, uptake followed by sorption process is the main removal mechanism of ACT using duckweed plants. Nevertheless, the uptake and sorption process are strongly initial ACT concentration dependant. The removal rates ($k_{r1}$and k_{r2}) were significantly decreased from 0.884±0.12 to 0.199±0.02 d^{-1} and from 0.319±0.05 to 0.137±0.01 d^{-1} at increasing the initial concentration of ACT from 1 to 1000 µg/L, respectively. Duckweed uptake capacity for pharmaceuticals decreases with the increase in the initial concentrations, which can be attributed to the increase of the ACT concentration than the uptake capacity of the duckweed. Similar trends were observed by Reinhold and Saunders[12]who found that increasing the initial concentration of pharmaceuticals reduced the duckweed uptake efficiency.

3.2. Diclofenac (DFC)

The results showed that the ADW treatment contribute the highest aqueous depletion of DFC followed by the IDW, MDW and No-DW reactors. The aqueous depletion of 1µg/L initial concentration of DFC was 88.75±3.0%, 66.25±1.5%, 47.80±2.0%, and 30.5 ± 1.0% for ADW, IDW, MDW, and No-DW reactors, respectively. The removal ratios dropped to 49.2±1.9%, 40.0±2.1%, 34.7±1.6%, and 26±1.4% for ADW, IDW, MDW, and NO-DWreactors, at 1000 µg/L initial concentration of DFC, respectively. This excellent performance towards the removal of DFC in ADW reactors can be attributed to the duckweed uptake process for DFC. Fig.2 shows the removal of DFC with different initial concentration from drainage water by duckweed uptake through the ADW reactors. The results presented in Fig.2 showed that the aqueous depletion of DFC dropped from 83.51±3.0% to 65.25±1.5%, from 65.25±1.5% to 58.80±2.0% and from 58.80±2.0% to 47.50±2.0% after 10 d. of treatment at increasing the initial concentration from 1 to 50, from 50 to 500, and from 500 to 1000 µg/L, respectively. The duckweed system removal efficiency for DFC are comparable to the results of DFC degradation by using Glass photo-reactor[18]. The results demonstrate that duckweed systems provided less energy consuming treatment technology for drainage water containing DFC.

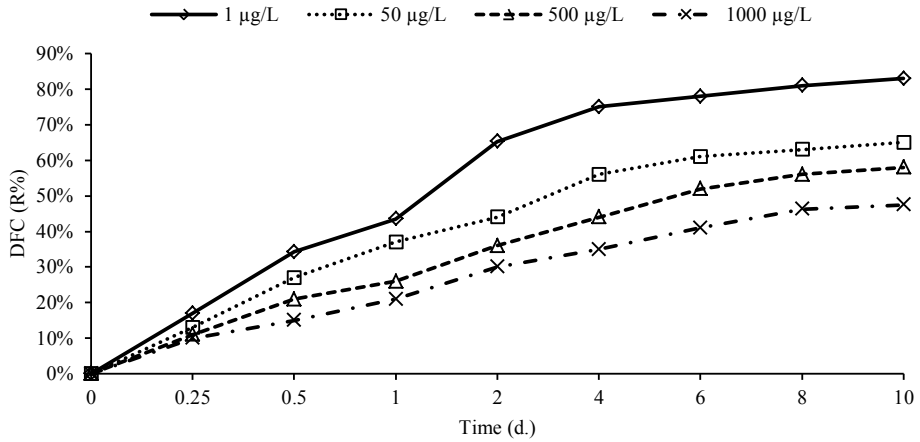

Fig.2 Removal of DFC of initial concentration of 1, 50, 500, and 1000 µg/L in ADW treatments.

Table 3 summarized the mechanism removal rate (k_r) values for DFC from drainage water. The results showed that the uptake rate (k_{r1}) was significantly affected by increasing the initial concentration from 1 to 1000 µg/L. The uptake rate (k_{r1}) was decreased from 0.528±0.02to 0.152±0.01 d^{-1} at increasing the initial concentration from 1 to 1000 µg/L, respectively. While, sorption (k_{r2}) and microbial degradation mechanism removal rate (k_{r3}) was decreased from 0.263±0.01 to 0.116±0.01 and from 0.154±0.01 to 0.094±0.01 at increasing the initial concentration from 1 to 1000 µg/L, respectively. The results revealed that duckweed uptake was the major aqueous-depletion process, followed by duckweed sorption and microbial degradation. However, volatilization and photo-degradation, showed a minor role in the removal of the DFC from drainage water in duckweed systems.

Table 3 Mechanism removal rate of DFC at different initial concentrations

		DFC			
	Treatment	ADW (k_{r1})	IDW (k_{r2})	MDW (k_{r3})	No-DW (k_{r4})
1µg/L	Measured k_r (d^{-1})	0.528±0.02	0.263±0.01	0.154±0.01	0.088±0.01
	Simulated k_r (d^{-1})	0.549±0.03	0.286±0.01	0.171±0.01	0.095±0.01
	R^2	0.99	0.98	0.93	0.87
50 µg/L	Measured k_r (d^{-1})	0.275±0.01	0.095±0.01	0.076±0.01	0.056±0.01
	Simulated k_r (d^{-1})	0.346±0.01	0.101±0.01	0.082±0.01	0.061±0.01
	R^2	0.93	0.91	0.86	0.76
500 µg/L	Simulated k_r (d^{-1})	0.224±0.01	0.129±0.01	0.097±0.01	0.068±0.01
	Measured k_r (d^{-1})	0.186±0.01	0.114±0.01	0.088±0.01	0.065±0.01
	R^2	0.88	0.831	0.789	0.87
1000 µg/L	Measured k_r (d^{-1})	0.152±0.01	0.116±0.01	0.094±0.01	0.075±0.01
	Simulated k_r (d^{-1})	0.167±0.01	0.129±0.01	0.103±0.01	0.083±0.01
	R^2	0.934	0.868	0.868	0.77

3.3. Progesterone (PRG)

The results revealed a significantly depletion of PRG at increasing the contact time from 0.25 to 10 d, for all treatment processes. At initial concentration of 1µg/L, the aqueous depletion of PRG was 95.25±3.0%, 93±3.4%, 64.5±3.2% and 43.5±2.4% for ADW, IDW, MDW and No-DW reactors, respectively. This was not the case at initial concentration of 1000 µg/L, where the removal ratios dropped to 66.50±1.7%, 63.2±1.9%, 35.3±1.5%, and 18.8±1.4% in ADW, IDW, MDW, and No-DW reactors, respectively.The results presented in Fig.3 showed that the aqueous depletion of PRG dropped from 98.35±3.0% to 95.25±3.0%, from 95.25±3.0% to 75.80±2.0% and from 75.80±2.0% to 66.50±1.7% after 10 d. of treatment at increasing the initial concentration from 1 to 50, from 50 to 500, and from 500 to 1000 µg/L, respectively.This study attributed approximately (66.50 to 95.25%) progesterone removal to uptake by duckweed plants, which is lower than the fraction (99%) that is reported by Farrel[14]. Hörsing et al.[19] attributed approximately 80% progesterone removal to duckweed plants.

Table 4summarizes the removal rate coefficient values of the PRG at different initial concentrations. The results showed that the uptake process is the main removal mechanism of PRG followed by sorption processes. The uptake rate (k_{r1}) was 0.719±0.03 and 0.563±0.01 d^{-1} in ADW and IDW at initial concentration of 1µg/L and decreased to 0.264±0.01 and 0.222±0.01 d^{-1} at initial concentration of 1000µg/L, respectively. This indicates that the duckweed uptake capacity for PRG decreased with the increase in the initial concentrations in the range of 1 to 1000µg/L.

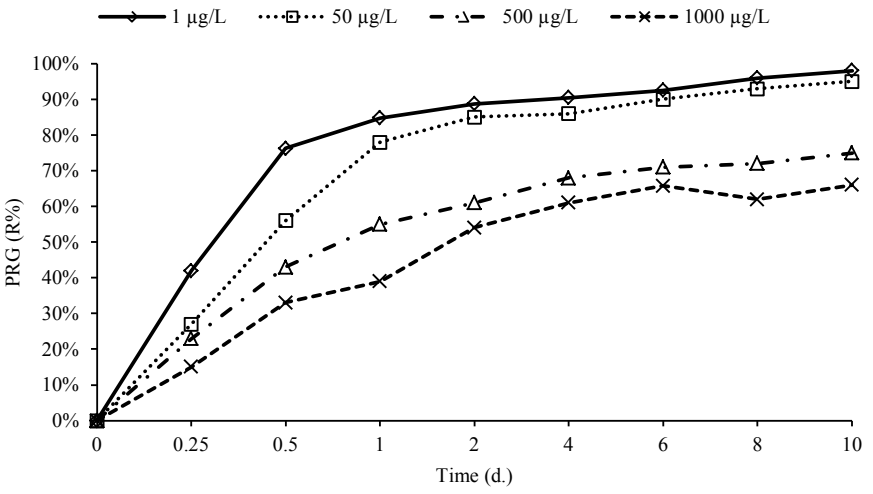

Fig.3Removal of PRG of initial concentration of 1, 50, 500, and 1000 µg/L in ADW treatments.

Table 4 Mechanism removal rate of PRG at different initial concentrations

		PRG			
	Treatment	ADW (k_{r1})	IDW (k_{r2})	MDW (k_{r3})	No-DW (k_{r4})
1µg/L	Measured k_r (d^{-1})	0.719±0.03	0.563±0.01	0.356±0.01	0.180±0.01
	Simulated k_r (d^{-1})	0.693±0.11	0.491±0.54	0.342±0.08	0.174±0.01
	R^2	0.960	0.930	0.850	0.830
50 µg/L	Measured k_r (d^{-1})	0.394±0.02	0.281±0.02	0.224±0.01	0.134±0.002
	Simulated k_r (d^{-1})	0.356±0.13	0.274±0.21	0.275±0.01	0.128±0.003
	R^2	0.861	0.650	0.904	0.860
500 µg/L	Measured k_r (d^{-1})	0.292±0.01	0.255±0.01	0.197±0.01	0.106±0.003
	Simulated k_r (d^{-1})	0.289±0.02	0.267±0.02	0.212±0.01	0.112±0.005
	R^2	0.848	0.868	0.935	0.933
1000 µg/L	Measured k_r (d^{-1})	0.264±0.01	0.222±0.01	0.091±0.002	0.044±0.002
	Simulated k_r (d^{-1})	0.257±0.01	0.218±0.02	0.101±0.004	0.045±0.002
	R^2	0.752	0.782	0.782	0.804

4. Conclusions

The current study demonstrated the performance of duckweed treatment systems in removing pharmaceuticals from drainage water. The results revealed that uptake rate of the examined pharmaceuticals are initial concentration dependant. Duckweed uptake capacity for pharmaceuticals decreases with the increase in the initial concentrations, which can be attributed to the increase of the pharmaceuticals concentrations than the direct uptake capacity of the duckweed that relies on duckweed activity. Moreover, the duckweed uptake contributed the major removal pathway followed by duckweed sorption and microbial degradation for ACT, DFC and PRG. Volatilization and photodegradation showed a minor role in removal of the pharmaceuticals in duckweed systems. The kinetics of the pharmaceuticals removal by using duckweed systems indicated that the removal of ACT, DFC and PRG typically followed first-order exponential decay relationships. The results presented herein demonstrates that the duckweed treatment systems represent a cost-effective, less energy consuming and environmentally friendly technology to eliminate pharmaceuticals at low concentration levels from drainage water.

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