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Phytoremediation of Organophosphorus Pesticides from Aqueous Media Using Azolla Filiculoides (Case study: Anzali Wetland)

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ABSTRACT

Recently, there has been an increased attempt to enhance the effectiveness of phytoremediation of polluted Environments such as aquatic media. The main goal of this research was elimination of diazinon and fenitrothion from aqueous media using Azolla filiculoides. In addition efficacy of effective parameters such as contact time, pesticides concentrations and different mass of A. filiculoides was evaluated. The results demonstrated that the proposed method had the significant advantages of simplicity and efficiency for diazinon and fenitrothion determination from real water samples. Based on results, the pesticides removal efficiency of A.filiculoides was more than 95%. On the other hand, findings of the experiments revealed that the proposed method was very efficient and useful for removing diazinon and fenitrothion from real water samples.

Keywords: Phytoremediation; Azolla Filiculoides; Pesticide; Anzali Wetland; Pollution

Introduction

Organic compounds released in the environment by anthropogenic activities. This has led to serious consequences on the environment due to their toxicity, hydrophobic nature and persistence in the environment for a longer period of time. Furthermore, organic compounds can enter the food chain, and due to their toxicity they can cause mutagenicity and carcinogenicity in animals and humans [1]. These days, different kinds of pesticides containing various groups of insecticides, herbicides, fungicides, nematocysts, acaricides, etc. are applied in agricultural activities. Due to pesticides complex chemical structures they are extremely stable and resistant to decomposition. Toxic pesticides have potential hazards for their consumers, environment, and chemosphere at a larger scale and can have negative impacts on the society and economics [2]. Insecticides are one of the most frequent water contaminants that can be found in different water resources. Therefore, the application of organochlorine insecticides is prohibited. Recently, organophosphorus pesticides are widely utilized all over the world [3]. Diazinon [O,O-diethyl O-(2-isopropyl-6-methylpyrimidin-4-yl) thiophosphate] is one of the organophosphorus insecticide that is applied in considerable quantities in agricultural and nonagricultural industries. Toxicity of diazinon is caused by its inhibition

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of acetyl cholinesterase. The studies on animal metabolism have demonstrated that diazinon affects their bodies by restricting acetyl cholinesterase metabolism [4]. Diazinon has been classified by the World Health Organization (WHO) as class II having a moderate hazard. This insecticide has log kow, vapor pressure, and Henry's law constant of 3.3, 1.4*10−4 mm Hg at 20°C, and 1.4*10−6 mm3 mol−1, respectively.

It is non-polar and moderately mobile, which raise a concern when involving groundwater and surface-derived drinking water [5]. Fenitrothion [O,O-dimethyl O-(4-nitro-m-tolyl) phosphorothioate], is mainly used in agriculture, to control chewing and sucking insects on rice, cereals, fruits, vegetables, and etc. In addition, due to photolysis and hydrolysis it undergoes degradation. This insecticide can be stable in water only when sunlight and microbial contamination do not exist. The major reason for degradation in soil can be attributed to biodegradation, although photolysis can also be another determinant. In World Health Organization (WHO), this insecticide is classified as moderate hazardous of class II. Fenitrothion has vapor pressure of 18mPa at 20°C and 14mg·L−1 solubility in water at 30°C [6,7]. Some common and efficient treatment methods to remove diazinon and fenitrothion from polluted water resources include chemical adsorption, coagulation, membrane process, bioremediation, and Advanced Oxidation Process (AOP). In addition, physical methods, such as activated carbon adsorption, nano-filtering, and Ultra-Low Pressure Reverse Osmosis (ULPRO) can be mentioned [8-10]. Phytoremediation mention to innovations including the application of living plants to elimination of pollution from contaminated media such as water [11]. Due to A. filiculoides rapid growth rate, ability to live in contaminated waters, and the high potential in absorbing various pollutants, it was proposed as a suitable plant with high ability to use in the phytoremediation processes [8]. The main goal of this study was to estimate the efficiency of A. filiculoides to eliminate diazinon and fenitrothion concentration in water. In addition, we attempted to intensify the phytoremediation potential of this plant by changes in some factors including initial concentration of pesticides, treatment time and plant fresh weight.

Materials and Methods

Chemicals and Reagents

Azolla filiculoides and water that we need for the experiments were collected from the Anzali wetland, Gilan, Iran. Diazinon and fenitrothion (95% technical grade) were purchased from Caron co., China.

Experimental Procedures and Methods

Preparation of A. Filiculoides to Remove Diazinon and Fenitrothion: Healthy and matured A. filiculoides, used here were collected from the Anzali wetland, Gilan, Iran. According to the purpose and time of sampling, the smaller plants with light green in color were selected. In addition plants rinsed with distilled water and sterilized with Mercuric chloride (0.1%) for 30s, then washed with deionized water several times. Experiments for diazinon and fenitrothion elimination were carried out separately in the laboratory at ambient temperature of 25-28°C. To examine the ability of proposed method 4.5, 9.5 and 13.5 g of the water fern were laid on the surface of 12 glass aquariums with 1000 cm^3 volumes which filled with real water. Then the various concentrations of pesticides (30, 50, 70 and 100 mg L^{-1}) were prepared from the pesticides stock solution with initial concentration of 1000 mg L^{-1} and add them in to the glass containers. All the experiments were run for 15 day period under fluorescent lamp as a light source and greenhouse conditions. During the incubation the evaporated water was compensated by adding deionized water when needed. Sampling was done every five days (0, 5, 10 and 15 days) until the 15th day of the experiment. All experiments were conducted triplicates and the data were statistically analyzed by Microsoft Excel 2010 and SPSS [12].

Chromatographic Conditions

To assess the efficiency of proposed method in the elimination of diazinon and fenitrothion from real water samples, the water samples were collected from three different parts of International Anzali Wetland. Before evaluating the plants efficiency, all the mentioned water samples were analyzed to examine the available pesticides concentrations in the water. Based on the results there were no concentrations of diazinon and fenitrothion in the samples. At the beginning of the procedure, the water samples were passed through a membrane filter with a pore size of 0.45 μm to separate all the suspended solid particles. Real samples analysis was carried out on an Agilent 8790 system through Gas Chromatography-Mass Spectrometry equipped with nitrogen-phosphorus detection (NPD) and a 30 m* .53 mm i.d., 1.5 μm film thickness, HP1 (polydimethylsiloxane) capillary column. Nitrogen was used as carrier gas (linear velocity 34 cm s^{-1}). The detector gas (hydrogen) flow was 4 mL min^{-1}, the air flow was 93.8 mL min^{-1} and nitrogen was used as detector make-up at 32.3 mL min^{-1}. The injector temperature was 180°C. The column temperature was programmed from 155°C to 250°C at 3° min^{-1}. The detector temperature was 320°C and the injection volume was 1 μL [7].

Evaluation of the Removal Ability of A. Filiculoides

To evaluate the elimination potential of A. filiculoides, the effective parameters in the process, including initial concentration, contact time and the adsorbent dosage were examined. To investigate the adsorption potential of A. filiculoides to eliminate diazinon and fenitrothion the removal percentage (%R) were determined based on the following equation [13,14]:

$$\%Removal\ efficiency = \left(C_0 - C_e / C_0\right) * 100 \quad (1)$$

where C_0 and C_e are the initial and final concentrations of diazinon and fenitrothion in the solution, respectively. To analyze of numerical results statistically, ANOVA factorial experiments were conducted. In most environmental experiments, the researcher seeks to compare and examine two or more factors, and this issue can be realized in factorial experiments, because in this method, the effect of one or more independent variables on dependent variables can be measured and examined separately or simultaneously. Also, among the advantages of this method, we can mention economic and time saving, more information generation and more accuracy. In this method, the effect of each of the investigated factors (concentration of pollutant, time and amount of adsorbent) on the removal of diazinon and fenitrothion by Azolla has been studied.

Results and Discussion

Phytoremediation of Diazinon and Fenitrothion by Azolla Filiculoides

Table 1: Evaluation of the diazinon and fenitrothion removal ability of A. filiculoides.

Evaluation of effective parameters on Diazinon removal process				Evaluation of effective parameters on Fenitrothion removal process			
Amount of A.filiculoides	Optimum contact time	Optimum concentration	Maximum removal percentage	Mass of A.filiculoides	Optimum contact time	Optimum concentration	Maximum removal percentage
4.5 g	15 days	70 mg L-1	95%	4.5 g	15 days	100 mg L-1	95%
9.5 g	15 days	100 mg L-1	97%	9.5 g	10 days	70 mg L-1	99%
13.5 g	15 days	100 mg L-1	97%	13.5 g	10 days	70 mg L-1	99%

To evaluate the accuracy of the proposed method, replicate analysis of real water samples was performed in different days of retention time. As displayed in Table 1, a good compliance with the added and found diazinon and fenitrothion concentrations has been accrued by the proposed procedure. Concentrations of residual diazinon and fenitrothion in the water samples were measured through gas chromatography with nitrogen phosphorus detection (NPD) (an Agilent 8790A GC system). As a consequence, high removal efficiencies were observed in all treatments with more than 95% the remaining concentrations of all the samples were less than 20 µg L^{-1}. Thus, the suggested method was suitable to remove noticed pesticides from the real water samples.

Statistical Analysis of the Removal Efficiencies of the Azolla Filiculoides

Due to the multifactorial nature of the experiment, to analyze the information obtained from the evaluation of the residual concentration of diazinon and fenitrothion the factorial ANOVA test has been used. In addition the effect of contact time, initial concentration of pesticides, the mass of A. filiculoides and the interaction of these parameters on the removal process was investigated. As demonstrated in Tables 2 & 3 the parameters of contact time, concentration of pesticides and amount of plants have a significant interaction with the elimination process. Based on the results the influence factor of the parameters was 0- 0.05. The interaction of the effective parameters showed a significant relationship between these variables with the removal process of diazinon and. However as demonstrated in the results just the contact time has a significant relationship with the removal process of fenitrothion, which means that the only controlling factor of the removal process of fenitrothion by Azolla is the contact time.

Table 2: The interaction of effective parameters with diazinon removal process.

Source	Type Ill Sum of Squares	df	Mean Square	F	Sig
Corrected Model	201.607a	23	8.766	5.926	-001
Intercept	644.144	1	644.144	435.469	-000
density	48.801	3	16.267	10.997	-001
time	80.909	2	40.454	27.349	-000
weight	13.015	2	6.508	4.399	-037
density * time	28.61	6	4.768	3.224	-040
time weight	2.35	4	0.587	397	-807
density weight	27.923	6	4.654	3.146	-043
Error	17.75	12	1.479		
Total	863.501	36			
Corrected Total	219.357	35			

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Table 3: The interaction of effective parameters with fenitrothion removal process.

Source	Type Ill Sum of Squares	df	Mean Square	F	Sig
Corrected Model	68.673a	23	2.986	1.789	-148
Intercept	31.828	1	31.828	19.068	-001
density	2.759	3	920	-551	-657
time	55.057	2	27.528	16.492	-000
weight	8	2	-004	-003	-997
density * time	2.33	6	-388	-233	-958
time weight	697	4	-174	-104	-979
density weight	7.822	6	1.304	-781	-601
Error	20.031	12	1.669		
Total	120.532	36			
Corrected Total	88.704	35			

Conclusion

We conclude by nominating Azolla filiculoides as a phytoremediation agent that can be used for the organophosphorus pesticides such as diazinon and fenitrothion. This technology offers efficient, easy, cost-effective and environmentally-sustainable solution to the concern of pesticides accumulation in public reservoirs and waterways. A. filiculoides which is abundantly used as a nitrogen-infuser for rice cultivation provides a vehicle for purifying water bodies infested with diazinon and fenitrothion. Based on pervious papers and this research it is also recommended that artificial pond-based systems cultivated with A. filiculoides can easily be employed as a biological filter to eliminate pesticides, heavy metals, dyes and etc. from agricultural and industrial effluents and to let the resultant flow-through exit from the drainage system.

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