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Detrmination of Organophosphorus Pesticides (Diazinon and Chlorpyrifos) in Water Resources in Barzok, Kashan

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Abstract

***Background*:** The growing population and increasing needs to agricultural products increased use of pesticides resulting in contamination of the environment, including water. The purpose of this study was to determination of pesticide residues in agricultural water resources in Barzok city of Kashan in 2011.

***Materials and Methods*:** This study as a descriptive and cross-sectional study that was conducted at spring season. That totals of 135 samples of the agricultural water resources of Barzok were collected randomly and mixtures from different stations. After extraction and preparation of samples, the pesticide residues were determined by using the HPTLC (High Performance Thin Layer Chromatography) devices and CATS4 software. Finally, obtained data were analyzed by using ANOVA statistical methods.

***Results*:** The results of this study showed that maximum amount of diazinon and chlorpyrifos has been 22.43 and 11.79 ppm respectively, and the pesticide residues have declined gradually overtime of (p<0.001).Furthermore, after a month 95.9% of Chlorpyrifos and 88.8% of diazinon is degraded.

***Conclusion*:** Accordingly, the remaining of diazinon and chlorpyrifos after a month of spraying was higher than determined limited standards. Because Chlorpyrifos is broken down rapidly more than diazinon, it is recommended that besides educating farmers on the proper use of pesticides, unnecessary contact with the water resources of this region must stop at least one month after spraying.

Introduction

Water is the main component of life cycle. The importance of quality, maintenance, and development is increasing in the average. The origins of life are infected on the application of direct and indirect pollutant [1]. One of these pollutants is pesticides. Their using in the agriculture has increased because the growing population and the need to increase agricultural production and food [2, 3]. From these pesticides we can to cite: the organophosphorus, organochlorine, carbamate and pyrethroid pesticides. Organophosphorus is the largest and most diverse compounds of pesticides [4]. Organophosphorus pesticides, more than other compounds used by farmers due to the effect on a wider-angle of pests and also being more affordable than other pesticides [5].

These pesticides widely used to increase agricultural yields and also to control arthropod-borne diseases. However, because of lack of knowledge of most consumers with the harmful effects and correct using the principles of this pesticide, this work will be partitioned or irregular [6, 7]. Pesticides used in agriculture can enter in to surface water resources through irrigation and rainfall and diversion of streams and caused by pollution of these waters and resulting in the unpleasant effects indirectly on agricultural products also either directly affects on the health of people that living near these sources by entering the food chain [6-9]. Water pollution by pesticides is a global problem as well as environmental pollution [10]. Studies show that in the areas where pesticides used to control for agricultural pest and rodents also Birds and other animals are susceptible to contamination. Dead Bodies of Birds around the sprayed farms is witness to this claim [11]. Therefore, human exposure either in advertent, accidental is the inevitable result of the use of pesticides or pesticide residues in the environment such as air, water, soil, plants and animals and in animate objects occurs[12, 13].

Organophosphorus pesticides, due to the effect on the cholinesterase enzyme and abnormalities in the central nervous system (CNS) are considered as a serious threading factor of human health; also according to the universal statistic's most deaths from pesticides are related to these poisons [2]. Adverse short-term effects resulting from exposure to pesticides are such as abdominal cramps, vertigo, headaches, diplopia, nausea, ocular disturbances and dermatopathies, and Long term adverse

effects include increased likelihood of respiratory failures, recalling disturbance, depression, nervous defects, prostate cancer, leukemia and infertility. The long-term effects are as a global public health problem [14, 15]. World Health Organization estimates that every year 3 million worker of farms suffers from severe poisoning from pesticides in developing countries, which 18 thousand people die from them. In addition, 21% of the food products are contaminated with pesticides. Approximately, 2.5 million tons of pesticides are spent in agriculture annually, that 50% of these chemical pesticides in to the soil, water and other resources [16]. Therefore, water resources must be prevented from contaminating by pesticides. According to that measurement of pesticide residues in waters is important in the human health care and the environment controls. For this reason the first step in the control and management of pesticide residues in water resources is the determination of the Concentrations of the pesticides with acceptable accuracy and comparing obtained values with the universal standards [13]

A review of the Great Plains of North America by Donald and et al. shown that, there are 29 types of pesticides in water, which two types are of pesticides and 27 types of those are herbicides [17]. Furthermore, a study was conducted by Jaipieam et al. in Thailand shown that the average concentration of Organophosphorus pesticides in the farming communities, in the dry seasons of the year is 0.085 µg/l, and in the wet season is 0.418 µg/l. These concentrations in the communities without farming were less and about 0.004 µg/l [18]. In a study conducted on pesticide residues on the surface waters of Santana in the California by Kenet and et al. shown that in the 92% of the collected samples are found one or more pesticides, which some samples the Diazinon pesticide have been higher than the standard values [19].

In a study was conducted to determine pesticide residues in Raval and Simly lakes Pakistan by Shazia et al., the results shown that the pesticide organochlorine, organophosphorus and pyrethroid are in the studied samples and one of these was the diazinon pesticide [20]. In a measurement that was performed by shayeghi and et al. on the remaining of azynfosmethyl and diazinon in Qarasu river and Gorganrood, the results were shown that the highest amount of pesticide residues was in the summer and the rate of this for azynfosmethyl in Qarasu River was 14.65 ppm and in Gorganrood was 14.90 ppm and for diazinon in Qarasu river was 22.4 ppm and in Gorganrood was 6.74 ppm [5]. In a study that was performed by Hosseini on the organophosphorus pesticide residues in the river water of Kor and Sivand Rivers in the Fars province shown that most of Diazinon residue in the kor one day after spraying was 2.13 ppm, and the Sivand river was 2 ppm [21]. Barzok is a city in the southwest of Kashan with Average 250 to 400 mm annually rainfall, that it is one of the suitable areas for agriculture in Iran. Based on the Agriculture official's reports that the area under cultivation of crops and orchards in this city is 750 acres and annual average usages of variety pesticides are 800 liters, that diazinon and chlorpyrifos are used by most of the other pesticides in this area. Thus by measuring the pesticide residues in agricultural water resources and compares them with standard values and determining pollution of waters by these pesticides, the risks of pesticides are considered and it can be reduced costs of treatment and complications related and also can be caused changes in policymaking and management of health services.

Materials and Methods

This study as a descriptive and cross-sectional study that was conducted in three stages as follows: 1. Selection the location of sampling and samples 2. Extraction or sample preparation and 3. Detection and measurement.

For the sampling and starting of study five stations were selected in Zayandehroud spring and Saadabad river of Barzok: Station no.1: in the upstream of Zayandehroud, station no. 2: in the upstream of Saadabad river, Station no. 3: In the middle of Zayandehroud, Station No. 4: Middle of SaadAbad river and Station no. 5: At the end of each resources (Figure 1).

According to the Barzok agriculture organization reports, and farmers, spraying were performed May of each year. Due to the time of the spraying sampling time was selected as follows: Sampling times are as follows: 1. the first days of spraying. 2. A week after spraying 3. Two weeks after spraying 4. One month after spraying 5. Two months after spraying [22]. From each station 5 samples selected in different parts. In addition, a city as well as the control sample was taken at the end of each stage of sampling (drinking water Barzok city), and also to ensure the absence of pesticides before spraying on water a sample taken from each station. Totally of 135 samples were picked randomly and then collected samples from each station mixed together at the end of each stage of sampling two one-liter samples were tested. Thus in totally 60 samples were analyzed. According to the Sampling standards (that) were performed from 10 to 25 cm depth.

Samples address were recorded Station number of the container, the location, date of sampling, temperature and pH. Then each sample was added 50 cc Methylen Chloride to prevent the breakdown of pesticides, it's a little will shake and then the door of containers containing the sample closed and sealed. The samples were transferred to the laboratory at 4°C to the operation. Extraction samples of pesticides being deposited silt, sediment and was done. So that 1 liter sample was poured into the funnel dekantor and 50 cc saturated solution of sodium chloride was added to it. Then in two successive stages 50 and 100 cc of methylen chloride was added to dekantor. Organic phase were separated from each stage and then Anydride sodium sulfate were added to be free of water. The separated organic phase at a temperature of 30°C to evaporate in a vacuum rotary device (Evaporation rotary) and was concentrated to 2 cc. And then 10 cc of acetone was added to it and it has evaporated to 2-3 ml. and again sample was rinse with 2 cc of acetone. Then it

was evaporated to 1 cc. Then added 1 cc of benzene and volume it up to 0.8 cc was evaporated and again final volume was brought to 4 cc by benzene that was ready for measurement by HPTLC. To identify and determine the amount of pesticides, making the spot on the plate, aluminum-containing silicagel (Silicagel 60 F 254) and by Applicator and capillary tubes was performed. For making the spot needed to standard samples of each selective pesticide diazinon and chlorpyrifos. Using the micropipette, 10 µl of passive and standard samples that was prepared were Rate spot on the plate. Distance the first spot from the edge of the plates and other spots was considered 2 cm. Also the spot diameter was not more than 1 mm. After the rate spot, plates were kept at room temperature to dry. Then the plates were in the tank containing the solvent, hexane-acetone ratio 20-80 cc. After the spots rising related to pesticides and their related standards, plates were outside from the tank and were kept at room temperature to dry. To see the spots, plates were in to UV Cabinet with Fluorescence light. Then the spots were view in Wavelength 254 nm. Finally, the samples were transferred to the HPTLC and using the special software CATS4 Samples were to determine and analyzed by ANOVA Methods [22, 23].

Results

The results shown that the average diazionin remaining in the spraying station No. 1 on the first day was 10.85, in the first week 2.73, the second week 0.93, the first month 0.58 and at the second month reached to 0 ppm. In the station No. 2 on the first day of spraying were 9.18, in the first week 3.3, the second week 1.31, the first month 1.08 and at the second month reached to 0 ppm. Station No. 3 on the first day of spraying was 10.79, in the first week of 2.68, the second week of 1.2, the first month of 1.07 and at the second month reached to 0 ppm. Station No. 4 on the first day of spraying was3.33, in the first week of 2.91, the second week 1.73, the first month of 0.94 and at the second month reached to 0 ppm. Station No. 5 on the first day of spraying was 5.29, in the first week of 1.83, the second week of 1.47, the first month of 0.76 and at the second month reached to 0 ppm (Table 1).

Also the data shown that the average Chlorpyrifos remaining in the spraying station No. 1 on the first day was 15.18, in the first week 14.63, the second week 2.45, the first month of 1.88 and at the second month were reached to 0 ppm. Station No. 2 on the first day of spraying was 22.29, in the first week15.44, second week 1.83, the first month of 0.58 and at the second month were reached to 0 ppm. Station No. 3 on the first day of spraying was 22.43, in the first week10.61, second week 1.63, the first month of 0.36 and at the second month were reached to 0 ppm. Station No. 4 on the first day of spraying was 22.07, in the first week13.95, second week of 4.6, the first month of 1.04 and at the second month month were reached to 0 ppm. Station No. 5 on the first day of spraying was 13.98, in the first week10.78, second week 3.43 and at the first month and the second month was reached to 0 ppm (Table 1).

The data shown that the average remaining of chlorpyrifos and Diazinon regardless of the sampling stations, in the first day of spraying were 7.89 and 18.79 ppm; respectively. In the first week 2.69 and 13.08 ppm, after two weeks reached to 1.33 and 2.79 ppm, in the first month reached to 0.88 and 0.77 ppm and in the second month reached to 0 ppm Also, Statistical analysis shown that the pesticide residues decreased with the time that There was a significant relationship (p<0.001). Also between Two cases of pesticide there was Interaction. This means that the reduced of amount of Diazinon on time was not the same as with Chlorpyrifos and the reduction speed of Diazinon was less than Chlorpyrifos (p<0.001) (Figure 1).

Statistical analysis of data showed that there is a significant relationship between the reduction of pesticide residues with over time (p=0.001). Also there is interaction effect between the pesticides studied, in other word reduction rate of pesticides with over time is not the same and reducing rate diazinon was less than chlorpyrifos (Table 2). Percent reduction diazinon and chlorpyrifos relative to first day of spraying based on the sampling stations shown in the table below: The results shown that the reduction rate of diazinon and chlorpyrifos regardless of the sampling stations in the first week of spraying compared with the first day of spraying was 64.7 and 30.3%; respectively, in the second week of spraying compared with the first day was 83.1 and 85.1%, and at the first month to the first Day was 88.8 and 95.9% (Figure 2).

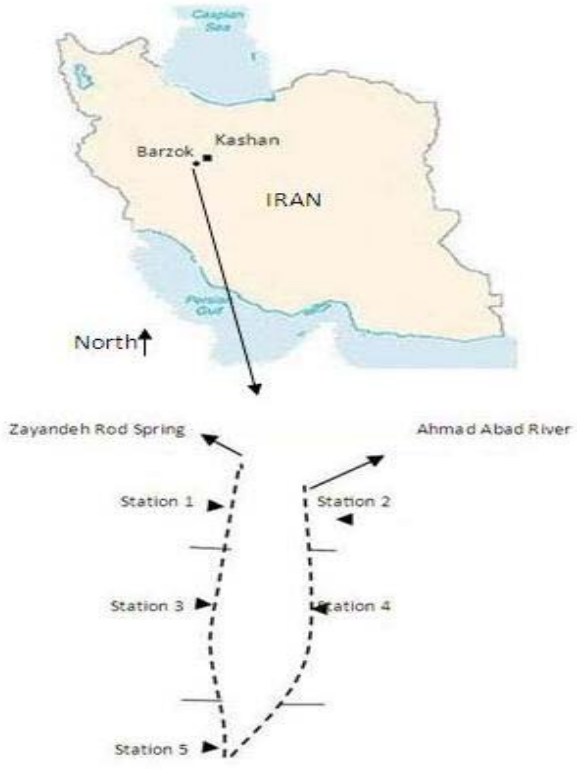

Figure 1. Locations of sampling stations in Barzok

Table 1. Diazinon and chlorpyrifos residues average (ppm) based on the sampling stations and sampling stages in agricultural water resources in Barzok of Kashan

Spraying time	Diazinon		Chlorpyrifos		(Green) p-Value
	Mean	SD	Mean	SD	
First day	7.89	3.34	18.79	4.38	
First week	2.69	0.69	13.08	2.43	
Second week	1.33	0.34	2.67	1.49	
First month	0.88	0.32	0.77	0.91	0.001>
Second month	0	0	0	0	
(Anova) *p*-Value	0.001>				

Table 2. The mean and standard deviation of diazinon and chlorpyrifos residues (ppm) in agricultural water resources in Barzok of Kashan

Station Number	Before spraying		The first day of spraying		First week		Second week		First month		Second month	
	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos
1	0	0	10.85	15.18	2.73	14.63	0.93	2.45	0.58	1.88	0	0
2	0	0	9.18	22.29	3.3	15.44	1.31	1.93	1.08	0.58	0	0
3	0	0	10.79	22.43	2.68	10.61	1.2	1.63	1.07	0.36	0	0
4	0	0	3.33	22.07	2.91	13.95	1.73	4.6	0.97	1.04	0	0
5	0	0	5.29	13.98	1.83	10.78	1.47	3.45	0.76	0	0	0
Control samples	0	0	0	0	0	0	0	0	0	0	0	0

Table 3. Percent reduction diazinon and chlorpyrifos relative to first day of spraying

StationNumber	First week		Second week		First month		Second month	
	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos	Diazinon	Chlorpyrifos
1	74.8	3.6	91.4	83.3	94.6	87.6	100	100
2	64	30.7	85.7	91.8	88.2	97.4	100	100
3	77.2	52.7	89.8	92.7	91	98.4	100	100
4	12.6	36.8	48	79.1	71.7	95.2	100	100
5	65.4	22.8	72.2	75.4	85.6	100	100	100

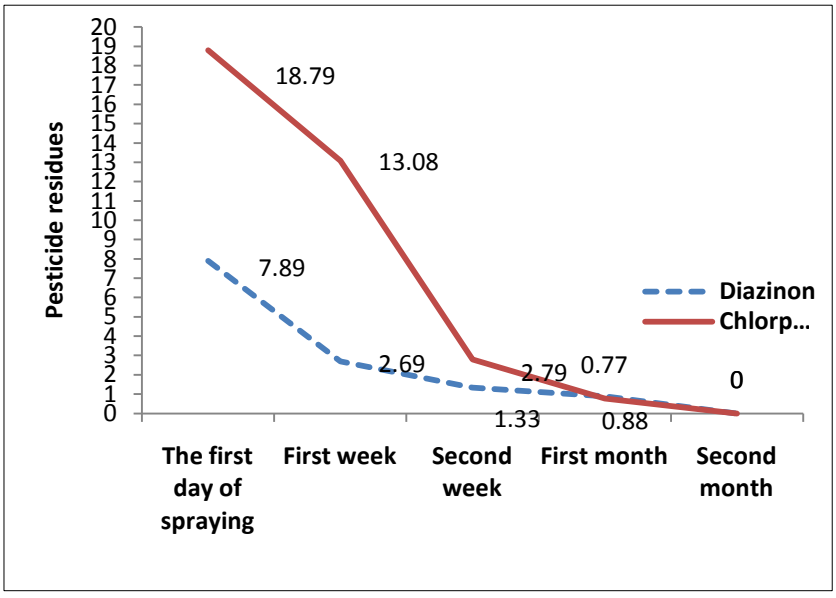

Figure 2. Average of Diazinon and Chlorpyrifos residues (ppm) in agricultural water resources in Barzok of Kashan

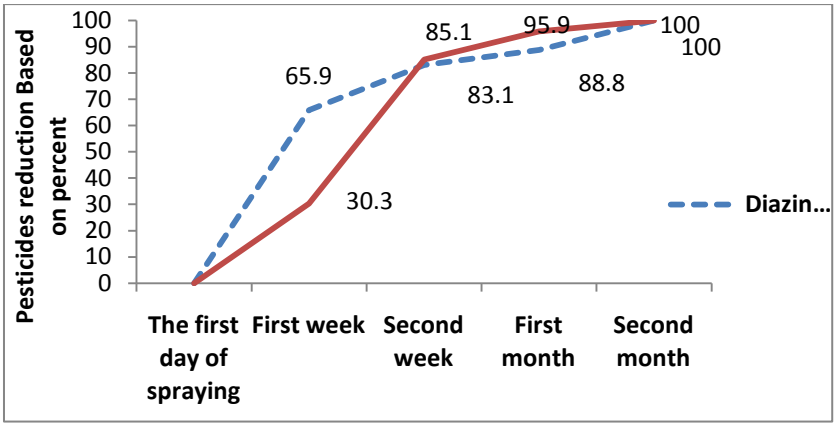

Figure 3. Diazinon and Chlorpyrifos reduction in agricultural water resources in Barzok of Kashan

Discussion

The results of this study shown that the maximum amount of chlorpyrifos was at station No. 3 (Middle of Zayandehroud) and the most amount of diazinon was at the stations No.1 (in the upstream spring Zayandehroud) on the first day of spraying. It seems that, there are more gardens and farms in this area and greater use of pesticides is sampled within the resource. In the interval, 1990-98 Water quality control board in SanJoaquin (California) conducted a study in relation to aquatic poisoning. Although they were unable to identify the cause of the poisoning but it is generally attributed to pesticides. Furthermore, the center conducted a research in the 91-92 and attributed the aquatic poisoning to the chlorpyrifos and diazinon in the water of this the area [23]. In a study that was done by Fianko et al. in Ghana in 2011 on pesticide residues in the fishes, and the risks associated with exposure to pesticides shown that more than 90% of the farmers do not the necessary precautions of application in the formulation of the pesticides and may be this is associated with disease about pesticides in agricultural areas [24]. In a study that has been done by Arjmandi et al. in the rice farms of the Mazandaran province shown that the organophosphorus pesticide residues in farm's fields after one day to two months of spraying was detected then the amount of pesticide residues are zero after this. These findings show that the lasting of organophosphorus pesticides in water, is an average two months. In this study, the highest level of diazinon was at station. no 1 which 1.14 ppm average amount [3]. In a study that was conducted by Shayeghi and et al. on the Karaj river and Amir-Kabir Dam, shown that the diazinon and malathion insecticide residues after one month spraying higher than authorized amounts and can create unpleasant effects on those people live that areas in which use of water and agricultural products of that region. However, remaining amount of insecticides is 0 after 2-3 months [25]. In a study that surveyed by Shayeghi et al. at the diazinon and malathion insecticide residues in the Shapor, Dalki and Mond water riveres in Booshehr province shown that the remains of these poisons after the first and second month of spraying were higher than standards levels, and Diazonin had the highest amount [26].

In a research was conducted by Honarpajouh on the two phosphorus pesticides in the Siminehrod and Mohabad rivers of the West Azerbaijan province show that these pesticides were shown in the growing seasons and the spraying and at other seasons, there was no or their amount was negligible. In this study, were observed that diazinon and azinphos-methyl insecticides samples, three months and two months after spraying, respectively [27]. In the survey that done by Selsele on the remains of three phosphorus insecticides in Tajan, Safa river, Tarak, and Babol rivers in the Mazandaran province shows that the amount of pesticides for malathion is 2.1 ppm, Diazinon 8.1 ppm and methyl azinofus 3 ppm [28]. According to similar conducted studies, in this study the remaining of diazinon and chlorpyrifos has declined gradually over time (p<0.001) and after two, months reached to zero. However, the amount of pesticide remains in agricultural water of Barzok higher than most other studies. This could be due to greater consumption and non-normative usage of pesticides and less knowledge of farmers with the lack of adequate training to correct use of pesticides. Note that, there are no certain standards for pesticide remaining in water on the Iran; thus the obtained results were compared with the German standard for diazinon (1 ppm) and Iran standard for chlorpyrifos (0.03 ppm), which the remnant of diazinon and chlorpyrifos pesticides is higher than some stations. Studies are shown that residual effects organophosphorous pesticides such as diazinon in water and environmental is higher than other pesticides [29]. Also the results of this study show that the residual rate of chlorpyrifos was more than diazinon, but after a month, more than 95% of the chlorpyrifos was removed, but this amounts for diazinon was 88.8%. Data shown that chlorpyrifos be eliminated more quickly than diazinon (p<0.001), that could be due to differences in

their chemical and physical structure. So with regarded to comparison of survival and price of these two pesticides, it is recommended that chlorpyrifos is used instead of similar cases, or instead of this pesticides could to use the other natural methods of pest control, that have fewer hazardous risks for living organisms and the environment or a combination of chemical and natural methods. Of course, the pH and temperature affected on the survival of pesticides in water so that at pH above 5 often organophosphorus is unstable, and the hydrolysis rate at pH above 8 with per each unit of pH increased into 10 times. Increasing pH increases the degradation of organophosphorus pesticides [23, 28]. According to that the pH of water in area samples in this study was above 7 and generally pH of this area were neutral toward alkaline, so organophosphorus pesticides have been low resistant in this waters. So it considered as an advantage. Also the temperature is increased the hydrolysis rate so that the hydrolysis rate is for increased per 10°C, 3.75 [22]. According to experiments carried out, the temperature of water at the sampling stations at least is 15°C and in the late-stage of sampling has been 20°C; this can be effective in reducing residue of pesticides in water. In a study was conducted by Petta et al. on water management in Fucino plain in Italy, the results show that the Rainfalls and irrigation are an important role in the transfer of pesticides to groundwater and surface water resources [30]. In this study, according to that the given average amount of rainfall in the region's is above average of Iran and do not using of modern methods of irrigation in this region, So rainfalls and poor irrigation can transfer pesticides to water sources in this region. Therefore to restrict the transfer of pesticides to water resources, using the appropriate irrigation methods and correct application of pesticides is proposed. Also, according to the spraying months (Ordibehesht and Khordad or May and June) and that many of the farmers are in the farm fields and gardens in this season and are in full contact to these waters and sometimes they used this water for drinking and other uses. It is recommended that at least a month after spraying avoid using and contact with these waters in this area or for drinking these waters, according to the pesticide's properties that decomposed these pesticides with rising temperatures, thus boiling the water before drinking is proposed.

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Authors' Contributions

All authors had equal role in design, work, statistical analysis and manuscript writing.

Conflict of Interest

The authors declare no conflict of interest.

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