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The impact of two insecticides on the pollutant cycle and quality of surface and groundwater resources in the irrigated lands of Yasuj, Iran

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A R T I C L E I N F O

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A B S T R A C T

The increase in the need for food and agricultural development has led to an increase in the use of insecticides. The use of insecticides leads to air, soil and water pollution. This study investigated the pollutant concentration cycle in the environment by analyzing diazinon and deltamethrin in a river and groundwater sources affected by an agricultural area. The samples were analyzed based on the standard method for insecticides in water using a GC-MS. The results showed that the quality of the surface water affected by the agricultural effluents decreased so that the changes of dissolved oxygen, nitrate, turbidity, TOC, BOD, and COD were 15.2%, 189.6%, 00%, 53%, 176%, and 57.5%, respectively. The concentration of diazinon and deltamethrin in agricultural wastewater was 86 μg/L and 11.62 μg/L. The self-treatment capacity of the river reduced the concentration of diazinon in the distance of 2 km and 15 km by 80.8% and 90.3%, respectively. These conditions were observed for deltamethrin in 74.8% and 96.2%, respectively. Also, the concentration of the two insecticides in water resources has temporal and spatial variation. The difference between the maximum and minimum concentration of diazinon and deltamethrin at different times was 183.5 and 1.73, respectively. The concentration of diazinon and deltamethrin in the downstream groundwater of the studied irrigated area was 0.3–0.7 μg/L, respectively. Although the soil structure and the self-purification capacity of the river caused a significant reduction of insecticides, the remained concentration of these pollutants in underground and surface water resources can still be a health and environmental concern.

1. Introduction

One third of water consumption in various sectors such as drinking, industry, and agriculture is supplied from surface water sources such as rivers, lakes, dams, and canals, which shows their importance [1]. In this situation, any change in the quality of surface water resources can be a serious risk for sustainable water supply, especially in developing countries [1,2]. Surface water pollution is one of the serious concerns around the world, which has intensified under the influence of industrial activities as well as the expansion of

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agriculture [3]. Also, groundwater is an important source of water supply, especially in arid and semi-arid regions, which face limited surface water resources [4]. However, urbanization, agricultural and industrial activities, and even climate change are a threat to the quality of underground water resources, which can cause water contamination with heavy metals, hydrocarbons, pesticides, microplastics, and trace organic contaminants [5,6]. Agriculture is known as one of the important sources of surface water and ground water sources contaminants that pesticides and insecticides are the most important of them [7–9].

Population growth and increasing urbanization have increased the need for food in the world, and meeting this need through agricultural development has caused an important challenge in the use of land and water resources [10,11]. Food supply with the development of irrigated agriculture has caused an increase in the ratio of irrigated land and leads to greater use of water resources [11,12]. Economic competition for more production can increase the use of water resources for agriculture, which is largely dependent on climatic conditions and water availability [13,14]. Therefore, there is a concern that the development of agriculture and the conversion of dry land to irrigated land to produce the required food will intensify the stress on the water bodies [15,16]. One of the ways to increase agricultural products in a situation where the need for food has increased and land resources are limited is to expand cultivation twice or more per year, which leads to an increase in water consumption as well as an increase in the use of fertilizers and insecticides [17].

These conditions have caused agriculture to be recognized as one of the important sources of water pollution. In fact, although cities and industries are the main source of water pollution due to wastewater generation [18], the use of chemical fertilizers and insecticides in agriculture is an important source of water pollution [7,8]. Water pollution caused by agriculture can be evaluated by its effect on the quantity and quality of water. In this cycle, primary factors including population growth, changing consumption patterns, and climate change have led to an increase in the need for food and energy. Also, secondary factors including the development of irrigated lands, rainfed agriculture, and inland aquaculture have led to an increase in water consumption [19]. The effect caused by primary factors and secondary factors has caused a decrease in the quantity of water, an increase in various pollutants such as insecticides, nitrates, sulfates and emerging pollutants in water resources [19–21]. Therefore, traditional agriculture in developing countries, in addition to consuming a high volume of water, is the source of the entry of pollutants such as insecticides into river and groundwater, which can exacerbate the water crisis in these areas [22].

Today, the use of insecticides to increase the quantity and quality of agricultural products is widely used in the world, but their consumption can have serious environmental and health consequences [23]. Insecticide residue in surface and groundwater is one of the serious problems caused by agriculture [24,25], which can vary according to various factors such as the type of agriculture, the type of insecticide, the insecticide quantity, and the water condition in the region. Organophosphate and Pyrethroid insecticides are one of the most widely used insecticides for more than 60 years, but they have health and environmental consequences for humans and organisms exposed to them [23,26]. Therefore, continuous monitoring of rivers and underground water sources affected by agriculture in terms of the concentration of this group of insecticides used in agriculture is necessary for health and environmental risk control [27]. This study analyzed the concentration of diazinon and deltamethrin, the two widely used insecticides in Iran in the agricultural

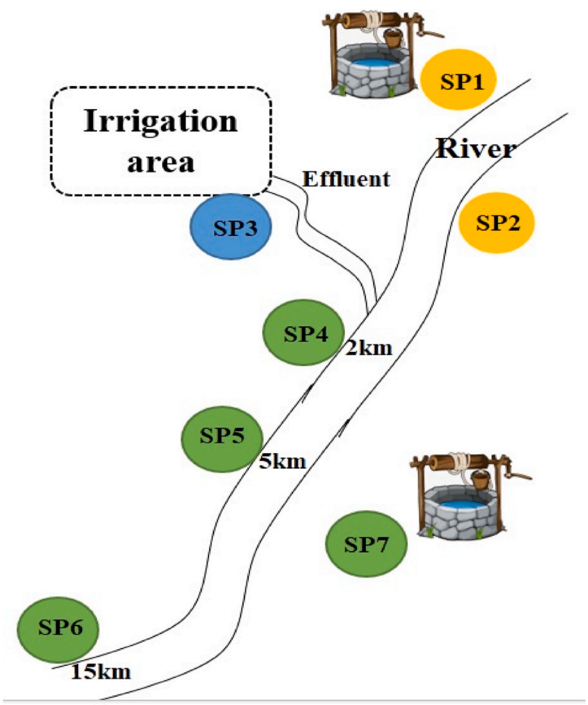

Fig. 1. Sampling points in this study.

effluent, downstream river, and downstream groundwater wells for 6 months with the aim of evaluating the cycle of pollutants concentration in the water resources.

2. Method

This study was conducted in the irrigated lands of Yasuj, Iran. For this purpose, the impact of an irrigated area (Tolkhosro farmlands) on Beshar River and groundwater was investigated. Two insecticides widely used in agriculture in the region, namely diazinon and deltamethrin, were selected to investigate the areas affected by agriculture and determine the concentration cycle of toxins. More sampling locations were determined as controls in the upstream area of agricultural lands that were not affected by irrigation. Sampling was in January, March, May, July 2022. As shown in Fig. 1, seven locations were selected for sampling, which included two samples from wells (groundwater), four samples from rivers (surface water) and one sample from agricultural effluent. One sample from the wells and one sample from the river were the control samples. In addition to two sampling points in upstream, the sampling points included one point in groundwater and three points in the river at a distance of two, five, and 15 km from the entering point of effluent into the river. The aim of choosing these points for sampling was to investigate the effect of self-treatment capacity of the river on changes in the concentration of insecticides. Also, after the distance of 15 km from the point of entry of irrigated area effluent, due to the entry of new polluting sources including residential wastewater and agricultural effluent from other lands, no further sampling points were defined. In order to evaluate the concentration of insecticides in the river and groundwater before entering the effluent from the studied irrigated area, two sampling points were selected (SP1 and SP2). A sampling point was selected to determine the concentration of insecticides in the effluent of the studied irrigated area (SP3). As explained above, three sampling points were chosen to determine the changes in the concentration of insecticides in the river (SP4-SP6). And finally, a sampling point was selected to determine the effect of agricultural effluent on the quality of groundwater (SP7). In this way, this study included seven sampling points. The samples were analyzed based on the standard method for insecticides in water using a GC-MS. The GC-MS was calibrated with 20, 50, 100, and 200 ppb standards. Five liters of water was taken in each samples, which were kept at $4^{^\circ C}$ and transported to the laboratory. 10 μl of internal standard of triphenylphosphine (TPP) with a concentration of 10 ppm and sodium chloride were added to 90 ml of sample. The magnet was thrown into the container, then 1 ml of hexane was added to it. After 20 min, the prepared sample was injected into the GC-MS.

3. Results and discussion

Table 1 shows the quality of river and groundwater upstream of irrigated lands. The results showed that the water quality of the river before entering the agricultural effluent was very favorable and the amount of dissolved oxygen was 7.2 mg/L. The BOD and COD of the river water were 1.2 and 3.3 mg/L, respectively, which indicated a negligible organic load in the river. Another sign of good surface water quality in the studied area was low turbidity (8 NTU) and neutral pH. Also, the quality of groundwater in the studied area before the entrance of agricultural effluent showed that turbidity, BOD, and COD were 6 NTU, 0.6, and 1 mg/L, respectively. Comparison of surface and groundwater quality parameters with the water consumption standards in Iran showed that the quality of both resources for agriculture was completely in accordance with the standard and was very close to the drinking standard. Based on this, all parameters of surface water quality were in accordance with drinking standards as well as agricultural standards, except for turbidity, which was slightly out of drinking standards. Considering that compliance with consumption standards is one of the desirable characteristics of water in terms of health and economy, which can be effective in reducing water management costs [28], the studied water resources before entrance of agricultural wastewater in terms of economic and health were suitable.

However, the entrance of agricultural effluent into the river caused a significant change in its quality parameters. As shown in Fig. 2, dissolved oxygen decreased from 7.2 mg/L to 6.1 mg/L. Also, BOD and COD of the affected river water by the agricultural effluent were 3.45 and 5.2 mg/L, respectively. The biggest change caused by the agricultural effluent into the river water was seen in turbidity, which was 8 NTU higher than the turbidity of the river before the entrance of agricultural effluent. However, as shown in Fig. 2, the highest ratio of changes in the quality parameters of the river after the entrance of agricultural effluent included changes in TOC, nitrate, and BOD, respectively. The TOC of the river increased by 640% from 0.3 to 1.96 mg/L affected by agricultural effluent. Also, Nitrate and BOD of the river water increased by 190% and 176%, respectively. The decrease of dissolved oxygen and the increase of chloride in the river water affected by agricultural effluent were 15% and 42%, respectively. Considering that the agricultural wastewater leads to reduction in surface water quality, it is necessary to use different treatment methods for this type of wastewater [29]. Natural methods such as constracted wetlands have a good ability to treat agricultural wastewater, Rozema et al., 2016 reported

Table 1
Surface and groundwater quality before entrance of agricultural wastewater.

	DO	Nitrate	Turbidity*	Sulfate	Chloride	TOC	COD	BOD	Hardness	Alkalinity	TDS	pH
SW	7.2	2.9	8	15	9.8	0.3	3.33	1.25	NA	NA	NA	7.2
GW	3.7	NA	6	31	6.8	0.65	1	0.6	175	186	165	6.5
DWS	NR	50	5	250	250	NR	NR	NR	200	NR	1000	6.5–8.5
AWS	NR	30	NR	NR	NR	NR	NR	NR	NR	NR	NR	6.5–8.4

SW= Surface water (SP2), GW = Ground Water (SP1), DWS = Drinking Water Standard (Iran), AWS= Agricultural Water Standard (Iran), NA= Not Analyzed, NR= Not Reported, *NTU.

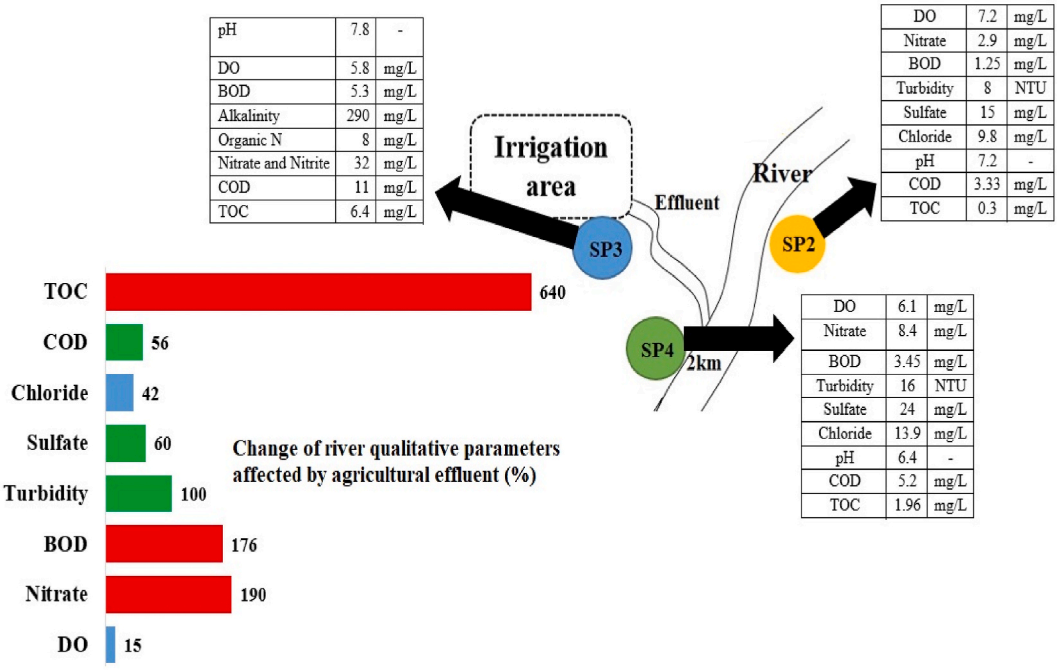

Fig. 2. River water quality changes affected by agricultural wastewater.

81%, 83%, 75%, and 42% reduction efficiency of BOD, TSS, TKN, and nitrate in agricultural wastewater, respectively [30]. Even high technologies can be used to recover useful compounds from agricultural wastewater. The use of membrane technology to recover carboxylic acid from digested agricultural wastewater was reported by Zacharof et al., 2016. with good efficiency [31]. Considering the economic limitations and problems of using hi-tech wastewater treatment technologies in rural areas [32], natural methods such as constructed wetlands and lagoons can be useful in reducing the negative effects of agricultural wastewater on the studied surface water resources.

The concentrations of diazinon and deltamethrin at seven sampling points at four different times are shown in Table 2. The results showed that the concentration of diazinon and deltamethrin in the sampling points before the entrance of agricultural wastewater was very low. However, the effect of insecticide concentration on agricultural wastewater (SP3), significant concentrations of insecticide were seen at all sampling points after the entrance of agricultural wastewater. The lowest concentration was associated with SP7, which represented the insecticide concentration in groundwater. Also, according to the insecticide concentration in the agricultural effluent, the concentration of diazinon and deltamethrin in the studied locations had temporal changes. The concentration of the studied insecticides was higher in May and July than in January and March. The significant decrease in the concentration of the studied insecticides in the groundwater compared to the surface water, which was 98.5% and 76% in the maximum concentration of diazinon and deltamethrin, respectively, can be caused by the treatment capacity of the soil bed. There are several studies that prove that the soil media has the capacity to reduce the concentration of insecticides from water with physicochemical and even microbial mechanisms [33,34]. Recent studies reported that several mechanisms in soil can help reduce insecticide concentrations. For example, Cycon et al., 2017 investigated the effect of Bioaugmentation mechanism in remediation of insecticide-polluted soil and stated that environmental factors and insecticide concentration have an important effect on the efficiency of this process [35]. Also, the soil media have the ability to absorb various pollutants, including insecticides, which depends on the characteristics and composition of the soil structure

Table 2
Detected concentrations at sampling points at different times (μg/L).

	Diazinon				Deltamethrin			
	Jan	Mar	May	Jul	Jan	Mar	May	Jul
SP1	ND	ND	ND	ND	ND	0.1	ND	ND
SP2	ND	ND	0.11	ND	ND	0.15	ND	0.13
SP3	**7**	**27**	**86**	**66.3**	**2.13**	**4.71**	**11.62**	**9.38**
SP4	0.17	1.36	16.5	31.2	1.68	2.18	2.92	2.08
SP5	0.1	1.1	9.4	18.6	1.05	0.95	1.37	1.11
SP6	0.1	0.8	8.3	13.8	0.42	0.38	0.44	0.34
SP7	0.1	0.32	0.3	0.45	0.65	0.58	0.7	0.76

ND= Not detected.

[36].

The statistical analysis shown in Fig. 3 showed the correlation of the temporal variation of diazinon concentration in agricultural effluent and the temporal variation of diazinon concentration in the river and also in the groundwater. Correlation of temporal variation of insecticide concentration in agricultural effluent and groundwater was 0.67. While the correlation of temporal variation of insecticide concentration in agricultural effluent and surface water at distances of two, five, and 15 km was 0.78, 0.77, and 0.82, respectively. Correlation of temporal variation of deltamethrin concentration in agricultural effluent with deltamethrin concentration in the river as well as deltamethrin concentration in groundwater is shown in Fig. 4. Correlation of temporal variation of deltamethrin concentration in agricultural effluent and groundwater was 0.66. While the correlation of temporal variation of insecticide concentration in agricultural effluent and surface water at distances of two, five, and 15 km was 0.84, 0.80, and −0.02, respectively. Therefore, the variation of studied insecticides in water sources was completely affected by its concentration in the effluent of irrigated lands.

Investigating the pollutant cycle in the downstream of agricultural areas showed that the concentration of diazinon and deltamethrin has spatial variation. The data in Table 2 shows that the concentration of diazinon and deltamethrin decreased gradually in different parts of the river. As shown in Fig. 5, the concentration reduction ratio was different for each of the investigated insecticides, and even the change ratio was different at different times. For example, the change ratio of diazinon and deltamethrin concentration at 5 km after the agricultural area in July was 71.9% and 88.2%, respectively. In the same distance, the reduction of diazinon concentration in Jan, Mar, and May samples was 98.5%, 95.9%, and 93.6%, respectively. However, the decrease in deltamethrin concentration at a distance of 5 km from the agricultural area in Jan, Mar, and May samples was 50.7%, 79.8%, and 88.2%, respectively. Temporal variation in reduction of the concentration of the studied insecticides can be caused by changes in the concentration in the emission source. Also, the changes in the flow of agricultural effluents and the increase in the amount of insecticides during irrigation can be the cause of the observed variations. Also, the seasonal change of the river flow is effective in the capacity of pollutant dilution, and it is expected that the capacity of pollutant dilution will decrease in the warm months when there is a decrease in river flow due to decrease rainfall and also increase water consumption [37]. In the rainy season and the increase in the flow of the river, which coincides with the decrease in irrigation and the decrease in the use of insecticides, the river exceeds its self-purification capacity [38], which was significantly seen in the Jan and Mar samples in this study. Finally, although the self-purification capacity of the river had a good ability to dilution the insecticide and also the soil media was effective in reducing the insecticide in the groundwater resources, but a significant amount of diazinon and deltamethrin, especially in the May and July samples were found to be a serious health threat.

Direct exposure to diazinon can have health effects such as headaches, nausea, skin irritation, runny nose, and vomiting, and even long-term exposure can cause neurological disorders, including memory loss [39]. Also, due to the widespread use of diazinon in agriculture, various side effects such as damage to target organs and tissues, reproductive damage, cytotoxic and genotoxic effects have been reported in different animals [40]. Serious concerns about the exposure of birds to diazinon, its effects on various types of aquatic animals, including salmon, and the possibility of carcinogenicity in humans have caused the need for more research on this insecticide [40]. One of the obvious effects of diazinon includes male fertility through reduce sperm production, decreased sperm quality, and

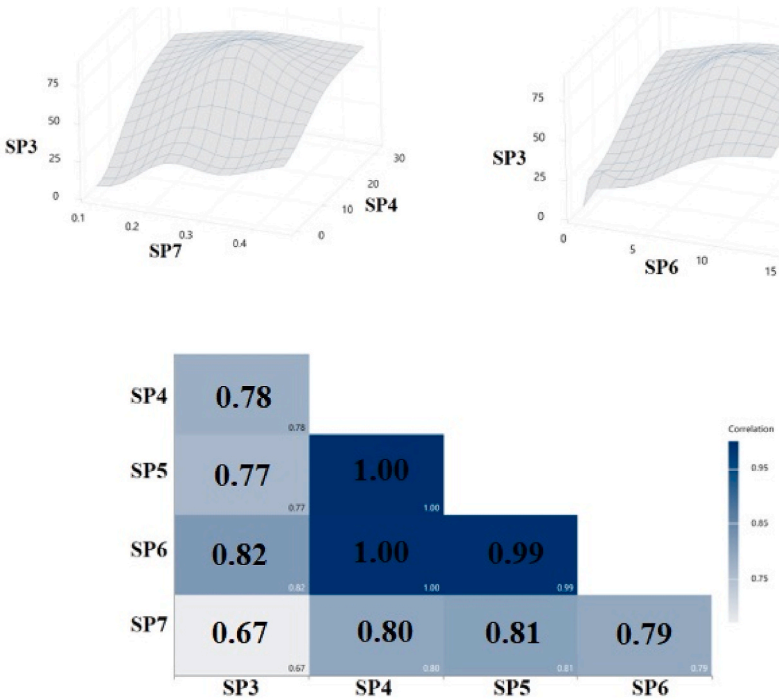

Fig. 3. Correlation of diazinon concentration in agricultural effluent and water resources.

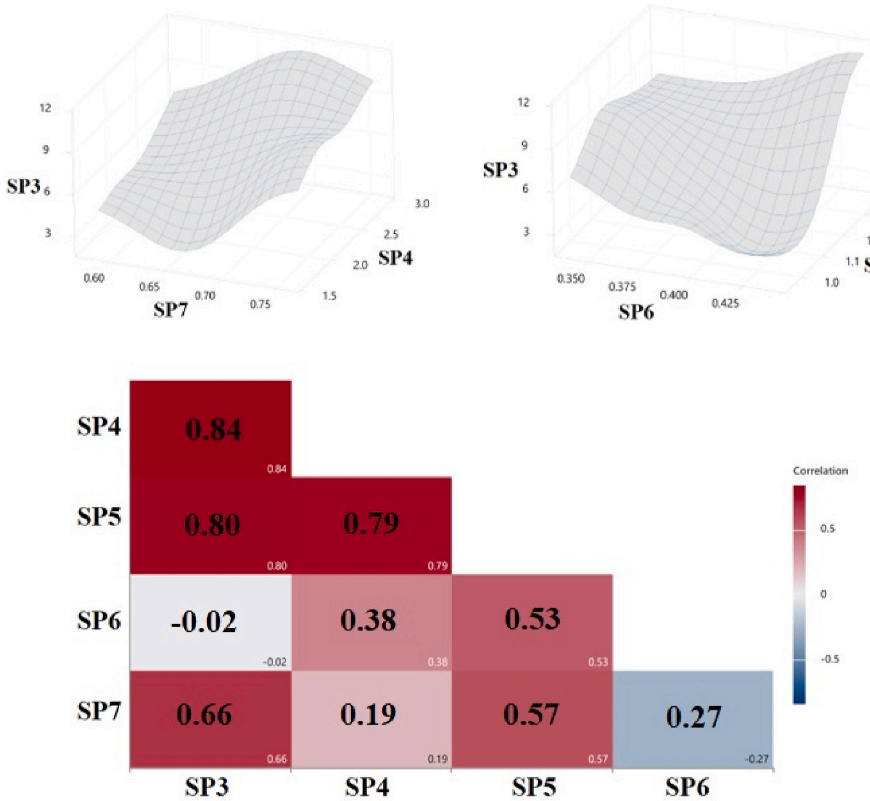

Fig. 4. Correlation of deltamethrin concentration in agricultural effluent and water resources.

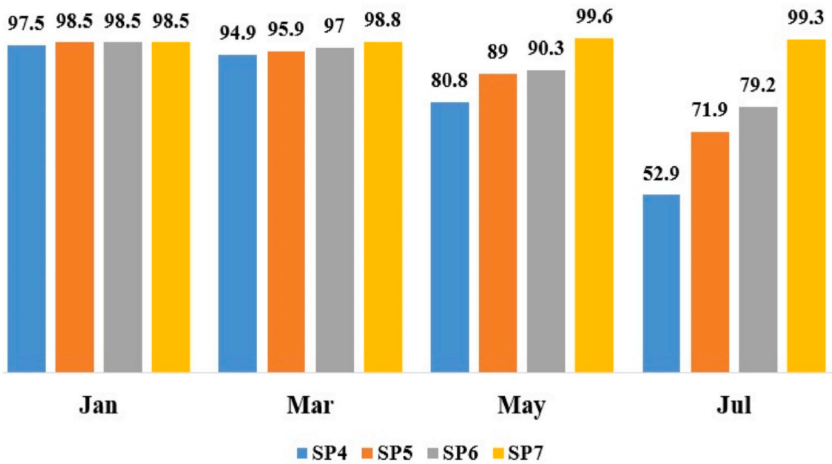

Fig. 5. Temporal and spatial variation in the reduction of diazinon concentration (%).

impaired sexual hormones production [41]. Therefore, the concentrations of diazinon observed in this study can be a threat to the local environment and the citizens exposed to it, especially the users of contaminated water resources. In addition, deltamethrin has known effects on the nervous system and also causes behavioral disorders [42]. The entrance of deltamethrin into the food chain and into the body through food and water can cause toxicity in humans, and this was contrary to the initial view about this insecticide, which was thought to have low toxicity for mammals [43,44]. One of the most important observed effects by deltamethrin is the effect on the liver, which confirms the concern of this insecticide entering the food chain [45]. Therefore, for the studied areas, it is necessary to carry out risk assessment studies based on the level of exposure to the observed concentrations in groundwater and surface water resources. For example, the study of the risk of organophosphates in surface water resources in southern Iran showed that although the treatment processes had the ability to significantly reduce organophosphates, the health risk of remained concentrations of insecticides is still a concern [28]. Therefore, considering the health importance of deltamethrin and diazinon on the health of citizens, the management of insecticide use in agricultural lands should be considered as the first step in risk reduction in the studied areas.

In this study, there were strengths and limitations that can be considered in similar studies in the future. The strengths of this study included the investigation of the effect of self-treatment capacity of the river on the changes in the concentration of insecticides, the variety of sampling points, the simultaneous attention to the pollution of surface and groundwater sources, and the selection of a developing area to conduct the study. However, there were some limitations that should be noted. Consideration of other relevant aspects including health effects and epidemiology, toxicity of insecticides and burden of disease, and water treatment needs could have been included to the study. The need to study other insecticides and the need to study the cumulative effect of various polluting sources on river and groundwater quality can be considered as other limitations of the study. We were aware of this limitation and tried to propose the future direction of studies to overcome this limitation in the future.

4. Conclusion

The concentration cycle of diazinon and deltamethrin in water resources affected by irrigated lands in Iran was studied. The results showed that the entrance of agricultural effluent into the river caused a decrease in water quality, so that TOC, BOD, nitrate, and turbidity increased by 640%, 176%, 190%, and 100%, respectively, and the amount of dissolved oxygen decreased from 7.2 mg/L to 6.1 mg/L. The concentration of diazinon and deltamethrin in agricultural wastewater during land irrigation was 86 μg/L and 11.62 μg/L, respectively. The entrance of this concentration of insecticide into the river in the warm season, when there is a decrease in the water flow, caused the concentration of diazinon to be 16.5 μg/L, 9.4 μg/L, and 8.3 μg/L at a distance of 2, 5, and 15 km from the entry of agricultural wastewater, respectively, which was caused by The pollution dilution in different distances of the river. This quantity for deltamethrin was 2.9 μg/L, 1.3 μg/L, and 0.4 μg/L, respectively. The highest concentration of diazinon and deltamethrin in groundwater resources was 0.45 μg/L and 0.76 μg/L, which was 99.4% and 93.45% lower than the maximum concentration in agricultural wastewater, respectively. Although the concentration of studied insecticides in surface water resources gradually decreased due to dilution and the soil structure as a natural media has been able to insecticide reduction before entering to the groundwater, however, the observed concentrations should still be considered as a health and environmental risk.

Author contribution statement

Zahra Zardosht: Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Farhad Khosravani: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Soheila Rezaei: Sajad Ghaderi: Analyzed and interpreted the data; Wrote the paper.

Ghasem Hassani: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

Data availability statement

Data will be made available on request.

Additional information

No additional information is available for this paper.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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