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Pesticides Residue in Drinking Groundwater Resources of Rural Areas in the Northwest of Iran

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ABSTRACT

Background: The majority of rural population in Iran depends on groundwater resources for drinking purposes. In recent years, pesticide contamination of limited water resources has become a serious challenge worldwide. This study quantified the pesticides residue in rural groundwater resources in the northwest of Iran.

Methods: A total of 78 groundwater samples were collected in June and September 2011 from all 39 drinking water wells. Liquid-liquid extraction (LLE) followed by Gas Chroma9tography/Mass Spectrometry (GC/MS) was used to determine the selected pesticides.

Results: Detection frequencies of profenofos, malathion, diazinon, endosulfan, trifluralin, deltamethrin, methyl parathion, and fenitrothion were determined with the concentrations exceeded 0.1 µg/L in 2.6, 17.9, 15.4, 10.3, 2.6, 2.6, 7.7, and 44.9% of the samples, respectively. Total pesticides residue was also observed in 26.9% of the samples with concentrations exceeded 0.5 µg/L. Among them, profenofos, malathion and diazinon were detected as the most frequently observed pesticides with the maximum concentrations of 0.542, 0.456 and 0.614 µg/L, respectively.

Conclusion: Higher pesticides residue than European Economic Commission (EEC) guidelines occurred in a number of monitored resources.

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Introduction

Based on the available information, about 96% of the world's total freshwater resources are in underground forms. Therefore, a main part of worldwide population in both urban and rural areas depends on groundwater resources as the sole drinking water source. As an example, groundwater resources are used for drinking purposes by about 50% of total population and 95% of the residences in agricultural areas in the United States.[1] In Europe, 70% of drinking water supply is based on groundwater resources as well as drinking water is almost entirely provided through groundwater in Austria and Denmark.[2] In Iran, more than 87% and 56% of rural and urban areas utilize groundwater resources, respectively. More or less, a similar condition is prevailing in the rural areas of East Azerbaijan Province, Iran.[3]

Restricted water resources protection, its importance and its role in various aspects of development are undeniable. Nowadays, contamination of limited water resources has become one of the most serious environmental, social, economical, and political challenges, especially in developing countries. During the last two decades, use of pesticides for agricultural and non-agricultural purposes has significantly increased worldwide, which is in coincidence with changes in farming practices and increasingly intensive agriculture. According to the United States Environmental Protection Agency (USEPA), the amount of pesticides used in the world and the United States of America (USA) was 2.4 and over 0.5 million ton, respectively, in 2007.[4] As stated by Statistical Centre of Iran, the total amount of pesticides distributed all over the country was 2291 ton in 2011. Accordingly, the average amount of pesticides used in Iran was estimated to be 0.7 kg/ha.[5]

Use of pesticides in agriculture may lead to the contamination of groundwater resources.[6-8] In fact, it has been estimated that less than 0.1% of the pesticide applied to crops actually reaches the target pest and the remaining enters the environment unreasonably and thus contaminates soil, water, and air, in which it can poison or adversely affect nontarget organisms.[9] The World Health Organization (WHO) recommended classification of pesticides by hazardto organisms is presented in Table 1. Application of banned and expired pesticides, unsustainable and uncontrolled consumption of pesticides, the lack of farmers' knowledge about the safe use of pesticides, and the lack of proper supervision by relevant organizations can cause more contamination of water resources in developing countries. The problem has become more prominent where groundwater aquifers are used as the main source for drinking purposes.[10]

Table 1: WHO classification of pesticides by hazard[11]

WHO class		**LD_{50} for the rat (mg/kg body weight)**	
		Oral	**Dermal**
Ia	Extremely hazardous	<5	<50
Ib	Highly hazardous	5-50	50-200
II	Moderately hazardous	50-2000	200-2000
III	Slightly hazardous	Over 2000	Over 2000
U	Unlikely to present acute hazard	5000 or higher	

LD_{50}: Lethal dose, 50%.

Pesticide contamination of groundwater has a stronger persistence than surface water, which may cause continuous toxicological effects for human health if used for public consumption.[10] Based on the research conducted in recent years, pesticide contamination of water resources has become a serious concern in both developed and developing countries. Existence of low to high concentrations of pesticides in both surface and groundwater resources has been reported in the agricultural areas from different countries.[2,6,8,12-19]

There are several studies in the literature confirmed the pesticide contamination of water resources in Iran. The findings of the study on organochlorine pesticides (OCPs) residue in the Karun River revealed the highest OCPs in the range of 71.43 to 89.34 µg/L.[20] In addition, surveys have shown the maximum level of diazinon and malathion of 768.9 and 506.6 µg/L, respectively, for Babolrood River.[21] In the southern coast watershed of Caspian Sea, the organophosphorous pesticides residue in surface and ground water resources could be a threat to the aquatic life.[7]

So far, there have been no discussions about pesticides residue in drinking water resources in the rural areas of East Azerbaijan

Province in Iran. Therefore, this study aimed to monitor the pesticides residue in all drinking groundwater resources in the rural areas of Malekan County, as one of the important agricultural districts of this province.

Materials and Methods

Studied area

Malekan, an area of intensive agricultural use, is located in the northwest of Iran (45˚55'- 46˚26'E, 36˚57'-37˚17'N) with an altitude of 1300 m above the sea level (Fig. 1). It has a semi-arid climate with annual precipitation ranging from 250 to 361 mm. Its 1006 km^2 area is composed of two urban and 81 rural districts. According to the census conducted in 2011, the population of urban and rural areas was 31487 and 74631 people, respectively.[22] In excess of 91% of rural regions were utilizing groundwater resources (39 water wells) as the sole drinking water source. The sampling area was mainly under the cultivation of crops such as grapes, wheat, and barley. As stated by Agriculture Jihad Ministry of Iran, about 21 ton of pesticides were consumed for agricultural purposes in the country in 2012.[23]

Fig. 1: Location of Malekan County in Iran

Considered pesticides and their characteristics

Among the commonly used pesticides in the county, ten compounds that were widely used in agriculture were considered in this study include trifluralin, chlorpyrifos, deltamethrin, malathion, endosulfan, methyl parathion, diazinon, fenitrothion, profenofos, and larvin. The selected pesticides characteristics, their toxicity data and human health effects are presented in Table 2.

Table 2: Pesticide characteristics, ecotoxicity data and human health effects.[11]

Common name	CAS No.	Chemical type	WHO class	LD_{50} mg/kg	Human health effects
Trifluralin	1582-09-8		U	>10000	Acute oral toxicity and dermal irritation; acute inhalation toxicity and eye irritation potential
Chlorpyrifos	5598-13-0	OP	III	>3000	Cholinesterase inhibition; overstimulation of the nervous system; respiratory paralysis and death
Deltamethrin	52918-63-5	PY	II	135	Gastrointestinal; neurological; respiratory; dermal and ocular effects
Malathion	121-75-5	OP	III	2100	Cholinesterase inhibition
Endosulfan	115-29-7	OC	II	80	Effects on nervous system; high acute oral and inhalation toxicity; eyes irritant
Methyl parathion	298-00-0	OP	Ia	14	Cholinesterase inhibition
Diazinon	333-41-5	OP	II	300	Cholinesterase inhibition
Fenitrothion	122-14-5	OP	II	503	Neurotoxic to adults; potential hazards to fetuses, infants and children
Profenofos	41198-08-7	OP	II	358	Cholinesterase inhibition
Larvin	59669-26-0	C	II	66	Probable human carcinogen

OP: Organophosphorus compound; PY: Pyrethroid; OC: Organochlorine compound; C: Carbamate.

Sample collection

A total of 78 groundwater samples were collected in June and September, 2011 from all 39 drinking water wells according to the standard methods of the examination of water and wastewater.[24] Characteristics of the monitored water wells are shown in Table 3. All samples were collected in duplicate from each location in sterilized 1 L polythene containers and immediately transported to the laboratory. Before the sample collection, 30-40 L of water was flushed out.

To avoid the passing and trapping of air bobbles, the containers were carefully filled and allowed to overflow. The samples were stored at 4°C for further extraction and analysis within 24 h.

Table 3: Characteristics of the monitored water wells

Code of water well	Depth of water table (m)	Discharge rate (L/s)	No. of covered villages	Distance from agri.[a] site (m)	Remarks
1	18	9	3	250, Located at the village	Potential cont. by agri. drainage, manure, and domestic wastewater
2	42	14	3	Located at agri. site	---
3	42	20	4	Located at agri. site	Potential cont.[b] by agri. runoff
4	15	10	3	Located at agri. site	Potential cont. by agri. and surface runoff
5	15	2.6	2	Located at agri. site	Potential cont. by agri. and surface runoff
6	42	10	2	50, Located at the village	potential cont. by agri. drainage, domestic wastewater, and leachate from solid waste
7	42	15	2	10, Near Mardaq River	Potential cont. by agri. and surface runoff
8	150	21	22	70	Potential cont. by surface runoff
9	9	3.1	2	Located at agri. site, near Mardaq River	Potential cont. by agri. and surface runoff
10	27	20	3	Located at agri. site, near Mardaq River	Potential cont. by agri. drainage and manure
11	42	12.5	2	10, Near Mardaq River	Potential cont. by agri. and surface runoff
12	15	13	1	Located at agri. site	Potential cont. by agri. drainage
13	13	8	1	Located at agri. site	Potential cont. by agri. drainage, manure and surface runoff
14	42	10.5	1	Located at the village, at the agri. site	Potential cont. by agri. drainage, manure, domestic, and oily wastewater and surface runoff
15	---	---	1	Located at agri. site	Potential cont. by surface runoff and manure
16	42	15	1	Located at agri. site	Potential cont. by agri. and surface runoff
17	42	20	1	Located at agri. site, near the dairy industry	Potential cont. by agri. drainage
18	42	5	1	Located at agri. site	Potential cont. by agri. drainage
19	42	16.6	1	Located at agri. site	Potential cont. by agri. drainage
20	42	5	1	Located at agri. site	Potential cont. by agri. drainage
21	42	10	1	Located at agri. site	Potential cont. by agri. drainage
22	15	10	1	Located at agri. site, near Leilan Chai River	Potential cont. by agri. drainage, manure, and surface runoff
23	6	1.4	1	Located in the vicinity of the village	Potential cont. by agri. drainage, manure, domestic wastewater, and surface runoff
24	48	15	1	100, Located in the vicinity of the village	Potential cont. by agri. drainage, manure, and domestic wastewater
25	42	20	1	Located at agri. site	Potential cont. by agri. drainage, manure, and surface runoff
26	12	---	1	Located at agri. site	Potential cont. by agri. drainage
27	45	2.5	1	Located at agri. site	---
28	32	5	1	Located at agri. site, near Mardaq River	Potential cont. by agri. drainage, manure, and surface runoff
29	36	20	1	Located at agri. site	Potential cont. by agri. drainage, manure and domestic wastewater
30	32	12	1	Located at agri. site	---
31	32	2.5	1	Located at agri. site, near Mardaq River	Potential cont. by agri. drainage, manure, and surface runoff
32	18	1.5	1	Located at agri. site, near Mardaq River	Potential cont. by agri. Drainage, and surface runoff
33	22	6	1	---	Potential cont. by manure
34	42	7.8	1	Located at agri. site, near Mardaq River	---
35	42	11	1	Located at agri. site	Potential cont. by agri. drainage and surface runoff
36	10	8	1	Located at agri. site, near Leilan Chai River	Potential cont. by agri. drainage and surface runoff
37	48	12	1	Located at agri. site	Potential cont. by manure
38	---	---	1	Located at agri. site, near the village	Potential cont. by agri. drainage, manure, domestic wastewater, and solid waste leachate
39	42	11	1	Located at agri. site	Potential cont. by agri. drainage

[a] Agricultural [b] Contamination

Chemicals, instrumentation, and analytical procedure

The collected samples were filtered using Whatman's filter paper with the pore sizes of 0.45 μm and 0.5 L of which was used for liquid-liquid extraction (LLE) using methylene chloride and hexane obtained from Merck. LLE was applied in triplicate for each sample with 10 mL of the solvents in each run. The pesticides residue was analyzed by GC/MS (Agilent-7890A/5975C), the conditions of which are presented in Table 4.

Table 4: GC/MS conditions

Condition	Values
Carrier gas	Helium
Carrier gas flow	1 mL/min
Temperature program	90°C (2 min)
	To 100°C at 25°C/min
	To 290°C (5 min) at 5°C/min
Injector temperature	250°C

Ethical considerations

There are no ethical considerations in this study.

Statistical analysis

Data were presented using mean (SD). Normality of the data distribution was assessed and confirmed using one sample Kolmogorov–Smirnov Test. The correlation between well depth and concentrations of pesticides residue and correlation between discharge rate and concentrations of pesticides residue were tested using Pearson correlation. *P*-values of <0.05 were considered as significant. All the analyses were performed by SPSS 13 (SPSS Inc. IL., Chicago, USA) software.

Results

The average plus SE values of pesticides residue in all drinking water wells in Malekan rural areas are depicted in Fig. 2. It is noteworthy that larvin was not detected in any of the samples in both June and September, as well; chlorpyrifos was rarely detected in water samples in September. Other eighth monitored pesticides in this study were detected with values less or greater than 0.1 μg/L in both samples collected in June and September.

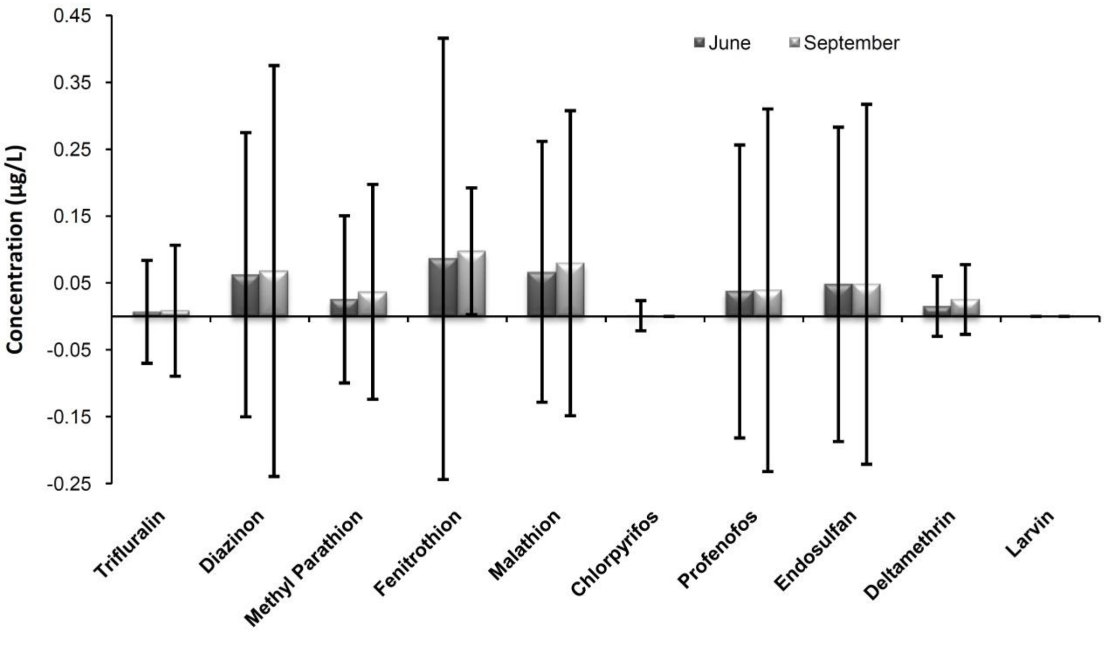

Fig. 2: The average plus SE values of pesticides residue in water samples

Detection frequency, median and maximum concentrations, and percentage of the monitored pesticides with concentrations exceeding 0.1 and 0.5 μg/L for single and total pesticides, respectively, are presented in Table 5. As can be seen from this table, profenofos, malathion, and diazinon were quantified in a majority of samples (87.2, 83.3, and 83.3%) with median concentrations of 0.021, 0.044, and 0.039 μg/L, respectively.

Table 5: Detection frequency, median and maximum concentrations, and percentage of the monitored pesticides with concentrations exceeding 0.1 and 0.5 μg/L for single and total pesticides, respectively

Pesticide	Detection frequency (%)	Median conc. (μg/L)	Max. conc. (μg/L)	Samples above 0.1 μg/L (%)	Samples above 0.5 μg/L (%)
Trifluralin	17.9	0	0.196	2.6	
Diazinon	83.3	0.039	0.614	15.4	
Methyl Parathion	62.8	0.014	0.321	7.7	
Fenitrothion	75.6	0.098	0.66	44.9	
Malathion	83.3	0.044	0.456	17.9	
Chlorpyrifos	2.6	0	0.045	0	
Profenofos	87.2	0.021	0.542	2.6	
Endosulfan	78.2	0.009	0.538	10.3	
Deltamethrin	52.6	0.003	0.104	2.6	
Total pesticides	89.7	0.317	1.88		26.9

Median conc.: Median concentrations; Max. conc.: Maximum concentrations.

Values of single pesticides residue in the drinking water wells (Codes 1-39) are shown in Fig. 3. As can be observed, the concentration of trifluralin in water well with code of 14 exceeded the maximum contaminant level (MCL) of European Economic Commission (EEC) (0.1 μg/L) in both sampling periods. Accordingly, concentrations of diazinon in the most of water wells were below the MCL for individual pesticides; however, the residues were higher than MCL in resources with codes of 14, 20, 24, 35, and 36. In the case of methyl parathion, higher values were observed in samples 14, 24, and 35 in both sampling periods. In addition, the concentrations of fenitrothion residues were greater than the MCL in the majority of samples (codes 2, 3, 9, 10, 12, 14, 17, 18, 20, 21, 23, 24, 25, 28, 30, 34, 35, 36, and 37). In addition, malathion residues were quantified in higher values than the MCL in water wells with codes of 2, 14, 20, 24, 28, 35, and 36. In the case of profenofos, the exceeded value was only observed in the sample with code of 5. Finding also revealed that the samples with codes of 2, 8, 12, and 31 are faced the exceeded values of endosulfan residue at both sampling times. Accordingly, deltamethrin residues were slightly higher than the MCL in water wells with codes of 2 and 3.

Concentrations of total pesticides in water resources as an important indicator for assessing water contamination with a group of pesticides are shown in Fig. 4. It can be seen from this figure that the total pesticides residue were quantified in higher values than the MCL in water resources with codes of 2, 5, 8, 12, 14, 20, 24, 31, 35, and 36.

It is apparent from Fig. 3 and 4 that the water well with code of 14 was the most frequently contaminated groundwater resource in terms of single and total pesticides residue. Total pesticides residue of 1.88 μg/L as the highest value occurred in water well with code of 14 in September. Median and maximum concentrations of the monitored total pesticides were 0.317 and 1.88 μg/L, respectively. Additionally, total pesticides residue was detected in 26.92% of the collected groundwater samples with concentrations exceeding 0.5 μg/L.

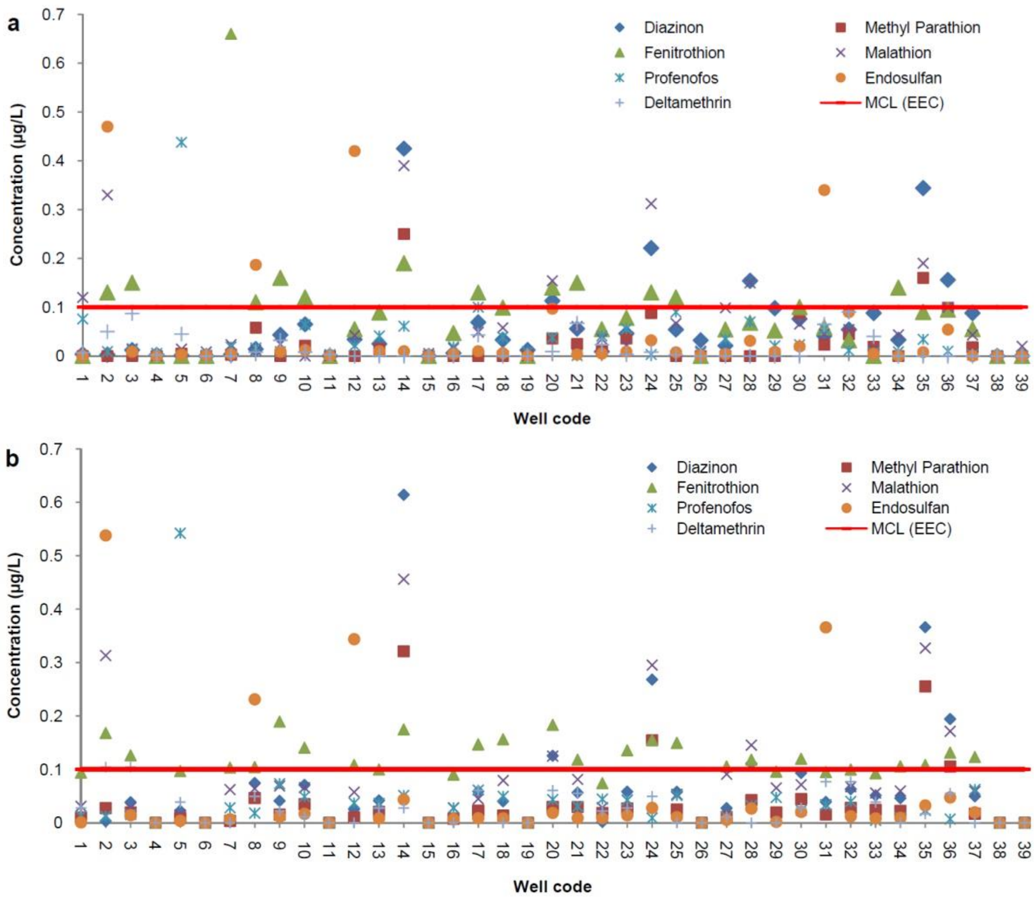

Fig. 3: Single pesticides concentrations in drinking water wells. (a) June and (b) September.

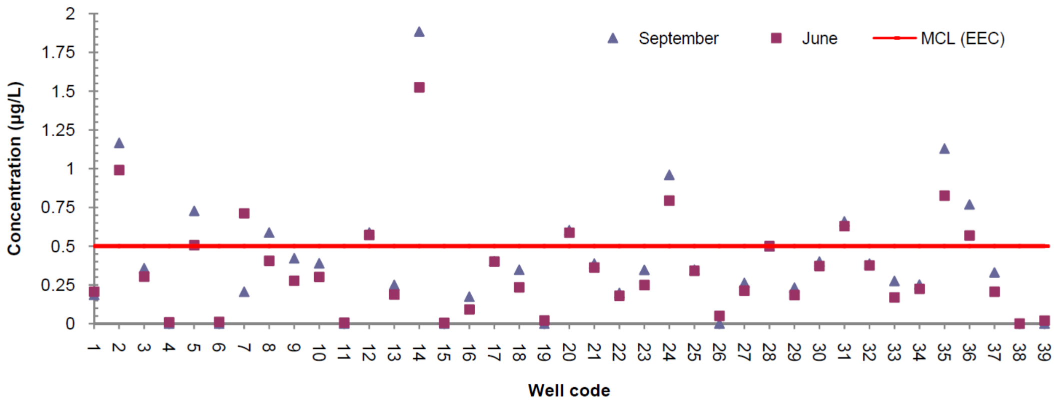

Fig. 4: Total pesticides in drinking water wells.

Discussion

According to the national standards[25] and those suggested by WHO,[26] among the pesticides monitored in this study, the MCL values was defined only for trifluralin and chlorpyrifos as 20 and 30 µg/L, respectively. However, in accordance with EEC[6] for drinking water, the total and individual pesticide levels should not exceed 0.5µg/L and 0.1 µg/L, respectively.

As shown in Fig. 2, larvin and chlorpyrifos were not detected in the water samples monitored in this study. According to

WHO's report, chlorpyrifos has low solubility in water, is strongly absorbed by soil, and does not readily leach from it. However, it has been detected in surface and ground waters in the USA, usually at concentrations below 0.1 and 0.01 µg/L, respectively.[26] A mean concentration of chlorpyrifos as 0.016 µg/L has been identified in the groundwater samples collected from the southern coast watershed of Caspian Sea in Iran.[7]

As mentioned in the table 5, the maximum concentrations of profenofos (0.542 µg/L), malathion (0.456 µg/L), and diazinon (0.614 µg/L) were observed in the water samples collected in September. However, the concentrations of profenofos, malathion, and diazinon in 2.6, 17.9, and 15.4% of the collected samples exceeded 0.1 µg/L, respectively. In a study conducted in Iran, mean concentration of diazinon equal to 0.019 µg/L has been detected in the groundwater samples collected from the southern coast watershed of Caspian Sea.[7] In another study, mean and maximum concentrations of diazinon in surface water samples in Iran have been reported in the range of 77.6 to 101.6 µg/L and 768.9 µg/L, respectively. Accordingly, the mean residues of 55.7 – 75.9 µg/L and maximum value of 506.6 µg/L for malathion have been reported in the surface water samples in Iran.[21] In addition, maximum concentrations of diazinon and malathion as 30.8 and 26 ng/L have been quantified in groundwater resources in Spain[27] and maximum residue of malathion as 29.84 µg/L has been reported for groundwater samples in India.[8] Malathion is moderately to highly mobile in soil and has been detected in some groundwater samples in the USA.[28] Under least favorable conditions (i.e. low pH and little organic content), malathion may persist in water with the half-life of months or even years. On the other hand, diazinon is moderately mobile in some soils, particularly those with organic matter content of $<3\%$ and can leach from soil into groundwater.[26] According to the USEPA, profenofos is not highly mobile and is not expected to leach into groundwater under normal use.[4] However, profenofos residues have been detected in 4.8% of drinking water samples in Mekong Delta, Vietnam, with the median and maximum concentrations of 0.04 µg/L.[19]

Endosulfan contamination has been detected in soils, water, air, and food products because of its widespread use and potential for environmental transport.[10] Because of its threats to human health and the environment, a global ban on the manufacture and use of endosulfan was negotiated under the Stockholm Convention in April 2011. Although the use of endosulfan is globally prohibited for agricultural purposes, it was detected in 10.3% of the collected samples in this study with concentrations exceeding 0.1 µg/L. As can be seen from the table 5, endosulfan was detected in 78.2% of groundwater samples with the median and maximum concentrations of 0.009 and 0.538 µg/L, respectively. Previously, the detection percentage of 8% for endosulfan has been reported from monitored groundwater resources in Pakistan.[18] Maximum malathion residue of 164.2 ng/L has also been observed in drinking water samples in India.[15]

According to the results, trifluralin was detected in 17.9% of water samples with the median and maximum concentrations of 0 and 0.196 µg/L, respectively. It was quantified in 87.4% of the collected samples with concentrations of lower than 0.1 µg/L (table 5). Trifluralin has low water solubility and high affinity to soil. So, it can persist in soils for months (DT_{50} 57-126 d) and is extremely resistant to leaching with a little lateral movement in soil.[29] It has been detected in surface water at concentrations above 0.5 µg/L and rarely in groundwater.[26] However, it has been observed with the maximum concentration of 0.182 µg/L in drinking water wells in Brazil.[13]

Deltamethrin has a little potential to leach into groundwater due to its strong tendency to bind to soil organic matter. In addition, according to WHO's report, no methyl parathion has been detected in any of the groundwater samples analyzed in the USA due to its no movement through soil.[30] As can be seen from the table 5, deltamethrin and methyl parathion were detected with concentrations of lower than 0.1 μg/L in 87.4% of the water samples. Totally, detection frequencies for deltamethrin and methyl parathion were 52.6 and 62.8% with the median concentrations of 0.003 and 0.014 μg/L and maximum values of 0.104 and 0.321 μg/L, respectively. However, up to 0.46 μg/L concentrations were detected in surface water samples in the agricultural areas in the USA.[30] Methyl parathion contamination in 5.4% of the monitored groundwater resources has been reported in Pakistan.[18] However, EPA has accepted voluntary cancellation of many of the most significant food crop uses of methyl parathion, one of the most toxic and widely used organophosphate pesticides.

Among the monitored pesticides in this study, fenitrothion was the most frequently found pesticide in groundwater samples with 44.9% of the samples containing > 0.1 μg/L. The detection frequency of fenitrothion was 75.6% with the median and maximum values of 0.098 and 0.66 μg/L, respectively. Fenitrothion is stable in water only in the absence of sunlight or microbial contamination.[26] These conditions usually exist in groundwater resources. However, according to the results of the study in Spain, maximum concentration of fenitrothion was 24.8 ng/L in the monitored groundwater resources.[27]

Results obtained from the statistical analysis of pesticides residue with well depth and discharge rate have shown no significant correlations between well depth and discharge rate with pesticides residue.

Conclusion

Standard values in Iranian environmental guidelines exist for only two pesticides monitored in this study: 20 and 30 μg/L for trifluralin and chlorpyrifos, respectively. Concentrations of these two pesticides in all groundwater samples do not exceed Iranian and even WHO's current drinking water criteria. Due to the lack of guideline values for other 8 pesticides monitored in this study, the MCL values which were established by EEC for discussing the contamination of water resources were used.

- Pesticides residue higher than EEC guideline was observed in a number of water resources in the case of single and total pesticides.
- Fenitrothion was the most frequently detected pesticide in terms of the samples containing residue concentrations above 0.1 μg/L.
- Water well with code 14 was the most contaminated groundwater resource because of the detection of nearly all individual pesticides with the amounts of higher than 0.1 μg/L and total pesticides with the concentration of 1.88 μg/L.
- Although the use of endosulfan is globally prohibited for agricultural purposes, it was detected in 10.3% of the collected samples in this study, with concentrations exceeding 0.1 μg/L.
- There were no significant correlations between well depth and concentrations of pesticides residue and between discharge rate and pesticides residue of pesticides residue.

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Competing interests

The authors declare that there is no conflict of interest.

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