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Spatial Distribution and Health Risk Assessment of Pesticide Residues in Surface Water and Tap Water in the Northwest of Iran

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Abstract

Background: This study aimed to determine the spatial assessment and risk assessment of three widely used pesticides, chlorpyrifos (CPF), malathion, and diazinon, in the tap water distribution network and surface water of Ardabil Province, northwest Iran.

Methods: Sampling was conducted at 41 points in the water distribution network and 30 surface water points. Pesticide values were detected by gas chromatograph-flame ionization detector (GC-FID) after solid-phase extraction.

Results: The ranking of pesticide compounds in tap water was CPF > malathion > diazinon. The mean concentrations of CPF, malathion, and diazinon in surface water were found to be 0.006, 0.013, and 0.003 mg/L, respectively. The highest pesticide concentrations were detected in the northeast region. CPF concentrations exceeded the maximum concentration level (MCL) in 7% of tap water samples.

Conclusion: Based on USEPA standards, the average non-carcinogenic health risks from exposure to pesticides through water consumption remained within the acceptable limits, with a hazard index (HI) below 1 (HI < 1 indicates "acceptable risk" per USEPA). Sensitivity analysis indicated that CPF concentration and the exposure duration to malathion had the most significant influence on chronic daily intake (CDI) and potential noncancer health effects. Although pesticide levels in drinking water mostly stayed within safe limits, concentrations in the northern region exceeded permissible levels. Therefore, further efforts are essential to regulate pesticide contamination and reduce related health risks in the future.

Keywords: Drinking water, Pesticides, Risk assessment, Water supply, Iran

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Introduction

The human population is projected to reach nearly 9 billion by 2050, and to meet the increasing food demand, agricultural production must rise significantly (1,2). In recent years, pest and disease control have greatly enhanced agricultural output (3-5). The State of Food and Agriculture 2023 report highlights a notable rise in pesticide usage, with global consumption reaching 3.54 million tons of active ingredients in 2021, i.e., an 11% rise over the past decade and double the amount since 1990 (6). While pesticides benefit human life by improving crop yields and controlling diseases, they also pose health risks through occupational or environmental exposures. Pesticides are among the most significant toxic environmental contaminants worldwide. The agriculture sector has repeatedly warned about the toxic effects of excessive pesticide use on public health and the environment (7,8). In developing countries, pesticide poisoning incidents are frequent due to rising consumption. Chronic exposure to pesticides can impair organ function and harm human health (9,10). Pesticide residues enter aquatic environments through drift, drainage, equipment washing, improper disposal of empty containers, leaching, and surface runoff. As a result, soil and water sources become major sinks for environmental pesticide residues (11,12).

Organochlorine pesticides (OCPs) and organophosphorus pesticides (OPPs) are the two main groups of pesticides used globally. OPPs are primarily used as insecticides and consist of organophosphate compounds. After the ban on persistent OCPs in the late 1990s, OPPs became a popular alternative. However, their high neurotoxicity and carcinogenicity, attributed to their role as acetylcholinesterase inhibitors, have been widely reported (13-15). Studies have linked OPP exposure to various diseases, including Parkinson's (16), diabetes (17), and prediabetes-like metabolic disorders due to alterations in hepatic cell signaling pathways (18). Adverse effects on metabolites such as citrate (decrease), glycerophosphocholine, threonine, and glycine (increase) have also been documented (19,20). OPP toxicity can lead to blood and biochemical changes, oxidative stress, and lipid peroxidation (21). Clinical symptoms of acute and chronic exposure include miosis, salivation, lacrimation, gastrointestinal distress, nausea, vomiting, diaphoresis, anxiety, muscle weakness, seizures, and respiratory failure (22). Late complications may include fatigue, paresthesia, and headache (23).

Even low concentrations of OPPs resist degradation, and minor chemical changes in their molecular structure can significantly alter their toxicity. OPPs bioaccumulate in soil, water, air, and agricultural resources, contaminating non-target organisms such as humans, fish, and birds. Minor chemical changes in the OPPs cause a noticeable shift in their toxicity from one species to another (24-26). Thus, the adverse health effects of pesticides cannot be ignored. Numerous studies worldwide have examined OPP residues in water resources (27-32). While some have assessed the health risks of pesticides in treated water, evidence of their presence in drinking water resources is abundant (33). Iran imports or manufactures approximately 14,000 tons of agricultural pesticides annually (34). Ardabil Province, northwest Iran, has 232,000 hectares under irrigated cultivation. To the authors' knowledge, no study has investigated pesticide residues in Ardabil water resources or their associated health and environmental risks. This study focuses on three widely used OPPs, CPF, malathion, and diazinon, to (1) monitor their concentrations in tap and surface water and (2) assess their health risk using the Monte Carlo approach. The findings aim to address environmental concerns regarding the use of OPPs and serve as a foundational reference for future research.

Materials and methods

Study area

The study was conducted in northwest Iran between latitudes 38°15′13.45″N and longitudes 48°17′59.96″E, in a region located between Gilan and Azerbaijan provinces, with a distance of approximately 70 km from the Caspian Sea. The region has a cold semi-arid climate and an annual mean temperature ranging from −25 °C to 35 °C. The yearly temperature of the district is 10.01 ºC (50.02 ºF), which is 8.42% lower than the average levels in Iran (35). The water resources include several aquifers, rivers, dams, and reservoirs, the most significant being the Aras border river.

Sampling

The location of the study area, situated in the north of Iran, is illustrated in Figure S1. The study area has been divided into four sub-areas based on ecological characteristics and types of cultivation, as depicted in the same figure. The Moghan region is one of the most important intensive agricultural areas of Ardabil Province with developed water resources, including agricultural irrigation networks. The water ecosystem of the Moghan region includes the Aras River and its marginal wetlands, the Dareh Rood River, water supply channels, and drains. All kinds of grains, industrial products, fiber products, and fodder are among the products of this region. Ardabil Plain, located in the central part of Ardabil Province, has fertile agricultural lands, and potato is its most important agricultural product. In Meshgin Shahr city, horticulture is the most important agricultural activity, including the production of apples and grapes. Finally, in Khalkhal and Kowsar, located in the south of the province, the cultivated crop species include barley, alfalfa, legumes, onions, leeks, etc., cultivated in a semi-mechanized manner. Therefore, pesticides including triazophos, malathion, phorate, dichlorvos, dicofol, endosulfan, CPF, and diazinon are used in the cultivation of these crops.

Forty-one drinking water samples (29 from surface resources and 12 from deep wells) and 30 surface water samples were collected in 2020 (Figure S1, S2).

The prevalence of three OPPs (CPF, malathion, and diazinon) was measured in these samples. These pesticides were the most commonly used, according to the report of the agricultural research center in Ardabil Province.

The bottles were rinsed with distilled water in the lab and with the sampling water on-site before sample collection. Following standard methods for water and wastewater examination, dechlorination of the samples was carried out at the time of collection. To protect water samples, ascorbic acid was added. The samples were then transferred to a coolbox, ensuring they were shielded from light and maintained at a temperature of 4 °C (36).

Extraction and chemical analysis

The solvents used for the extraction, namely acetone, methanol, and MTBE (methyl tert-butyl ether), were obtained from Merck Germany (gas chromatography grade). The standards for the pesticides chosen for the study were obtained from Merck Germany (99.6% purity level). HCl (1 N) and NaOH (0.1 N) were used to adjust the pH, and deionized water (Milli Q Millipore 18.2 MΩ cm−1 conductivity) was used for the preparation of all solutions.

The pH of the samples was adjusted in the laboratory. Solid phase extraction (SPE) of pesticide residuals was conducted using the Environmental Protection Agency (EPA) standard method (3535a). Styrene divinyl benzene-reverses-phase sulfonate (SDB-RPS) was used as the cartridge for SPE, and each cartridge was washed in two stages before use (by 5 mL acetone, then let to dry, and by 5 mL methanol). In the next step, the cartridge was conditioned with 5 mL of methanol and 20 mL of reagent water without drying. The disk was dried by maintaining a vacuum for about 3 minutes after the sample was passed through the solid-phase media. For elution or separating the analytes from the cartridge or disk, 0.6 mL of acetone was added to the cartridge, and it was soaked for 1 minute. The bottle was rinsed twice with methyl tert-butyl ether (MTBE). The evaporation method was used to bring the volume to exactly 5 mL. Finally, a volume of 1 μL was injected into the instrument (36).

The gas chromatograph (VARIAN Model CP-3800) equipped with a flame ionization detector (FID) was used for determining the OPPs. The gas chromatograph conditions for analyzing pesticides in water samples are shown in Table S1 (36).

Quality control and quality assurance

By preparing different solutions, the limit of quantification (LOQ) and limit of detection (LOD) of the pesticides were calculated. The signal-to-noise ratio (S/N) was recorded after injecting the solutions. In this study, the LOD was defined as three times the background noise of the chromatographic instrument (S/N ratio of 3:1), and the LOQ was defined as an S/N ratio of 9:1. The LOD and LOQ of the studied pesticides are shown in Table S2. Samples with values below the LOD were not detectable (ND).

Hazard quotients (HQs) for non-carcinogens risk assessment

Non-carcinogenic hazards are quantified as hazard quotient (HQ), a unitless value representing the probability-specific adverse effect. HQ is defined as the ratio of chronic daily intake (CDI) to the toxicity threshold value, known as the chronic reference dose (RfD) (Eq. 1). Here, CDIi represents the average daily intake of a pesticide from water, in mg/kg/day; Ci is the concentration of the pesticide (mg/L) in water. IR is the ingestion rate in L/day; EF is the exposure frequency in days/year; ED is the exposure duration in years; and BW is the body weight of the exposed individual in kg. For noncarcinogens: AT = ED * 365 days per year, and CF is the conversion factor (kg/mg). The requirement factors for computing exposure, risk assessment, and RfD values for CPF, malathion, and diazinon, according to the Integrated Risk Information System (IRIS: https://www.epa.gov/iris), are summarized in Table S3 (37).

$$CDI = \frac{C_i \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{CF}}{\mathrm{BW} \times \mathrm{AT}} \quad (1)$$

The noncancer risk was estimated using the hazard index (HI) from Equations 2 and 3, as follows:

$$HQ = CDIi \,/\, RfDi \quad (2)$$

$$HI = \sum HQ \quad (3)$$

In this study, HI values were calculated from the results of drinking water samples for the adult group. In the second step, sensitivity analysis was performed, and the uncertainty risk assessment model was incorporated using the Monte Carlo simulation approach with Crystal Ball software (Version 11.1.2.3, Decisioneering, Inc., Denver, CO, USA). Moreover, a representative risk distribution was produced by 10,000 trials, which is enough to confirm the result's stability (38).

Statistical analysis

Descriptive and inferential analyses were conducted using SPSS software (version 22.0). Calibration curves for OPP standards were generated using linear regression analysis. Data are represented as mean ± standard deviation (SD). Statistical significance was set at $P < 0.05$ for all tests. Pesticide distribution maps were created using ArcGIS 10.1, with independent raster layers for OPPs interpolated and visualized using the Kriging pattern.

Results

The analysis of drinking water samples from Ardabil Province in northwestern Iran is summarized in Table 1. The results for the tap water distribution system samples revealed elevated levels of CPF at 0.05 mg/L and malathion at 0.01 mg/L, while diazinon was not detected. Based on average concentrations across sampling locations, the pesticides were ranked in the following order: CPF (0.007 mg/L) > malathion (0.004 mg/L) > diazinon. However, in surface water samples, the mean surface water concentrations were 0.006 mg/L (CPF), 0.013 mg/L (malathion), and 0.003 mg/L (diazinon).

Figures 1-5 illustrate the spatial distribution of pesticide concentrations, highlighting that approximately 7% of tap water samples exceeded Iran's maximum concentration level (MCL) for CPF. The highest levels were found in the northern and central regions, primarily linked to

Table 1. Statistical description of the pesticides in water samples (mg/L)

Pesticide	Distribution (tap) water network (*n*=41)			Surface water (*n*=30)		
	Chlorpyrifos	Malathion	Diazinon	Chlorpyrifos	Malathion	Diazinon
Mean	0.007	0.004	ND	0.006	0.013	0.003
Std. Deviation	0.01	0.003	ND	0.004	0.02	0.007
Median	0.003	0.005	ND	0.005	0.01	ND
Minimum	ND	ND	ND	ND	ND	ND
Maximum	0.05	0.01	ND	0.01	0.08	0.03

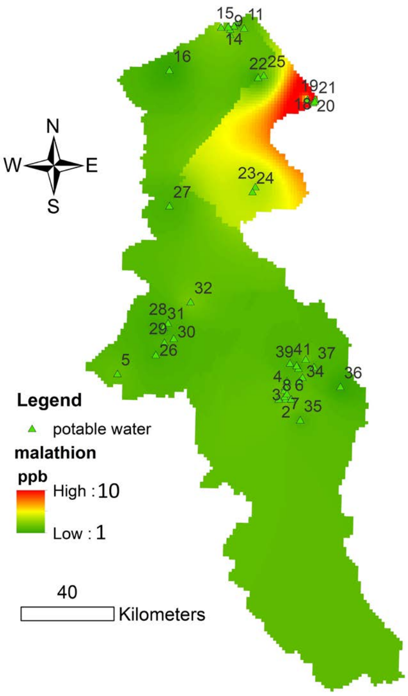

Figure 1.. Spatial trend of malathion in potable water

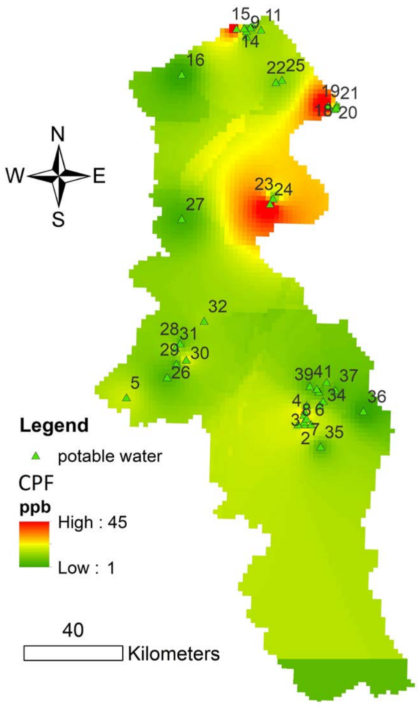

Figure 3. Spatial trend of malathion values in surface water

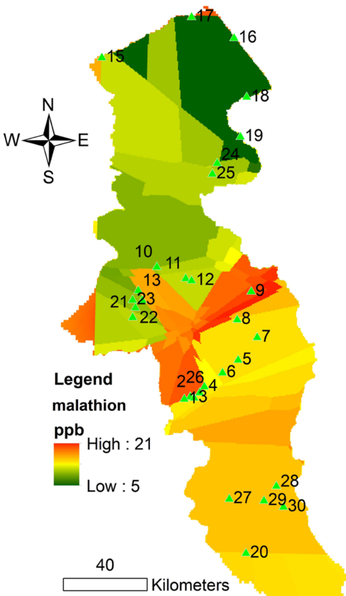

Figure 2. Spatial trend of CPF in potable water

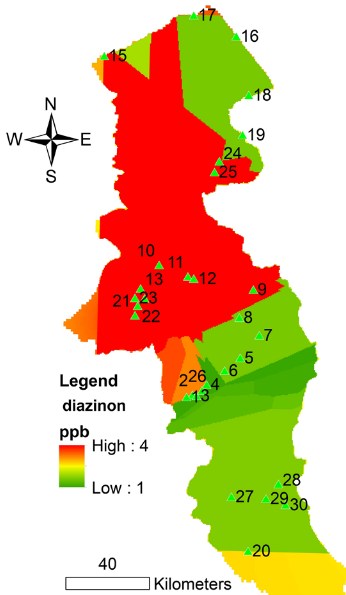

Figure 4. Spatial trend of diazinon values in surface water

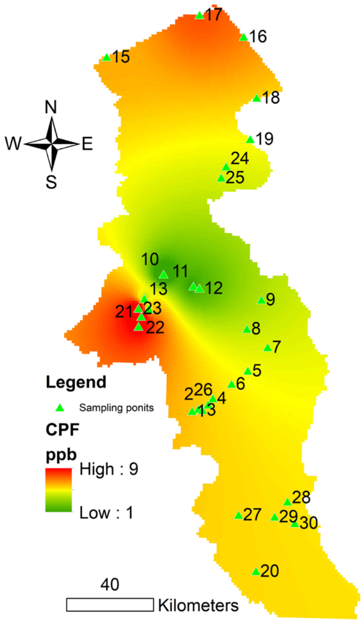

Figure 5. Spatial trend of CPF values in surface water

intensive agricultural activities and pesticide application. Furthermore, surface water contamination in central areas may also stem from domestic or livestock pesticide use. The non-carcinogenic risk assessment results, shown in Table 2 and Figures 6 and 7, indicate that all pesticide exposure levels remained below the risk threshold, with HI values under 1.

Figures S4 and S5 present the sensitivity analysis, which revealed that CPF concentration and the exposure duration for malathion were the most influential parameters affecting CDI and non-carcinogenic health risks. Additionally, body weight showed an inverse correlation with risk levels, and environmental variability contributed significantly to the uncertainty in exposure assessments.

Discussion

A high amount of pollution was detected in the drinking water network of northeast Ardabil Province. Significant pesticide contamination was detected in the drinking water network of northeast Ardabil Province. Some organizations have established maximum residue limits. The European Union (EU) has recommended 0.1 µg/L as the maximum allowed concentration for individual pesticides and related products, and 0.5 µg/L for total pesticides in drinking water (39). The USEPA reported an acceptable concentration of 20 µg/L for diazinon in drinking water and 1 µg/L for short and long-term exposure (40). The Institute of Standards and Industrial Research of Iran has established a maximum permissible concentration of CPF at < 0.03 mg/L (Standard No. 1053) (41). According to the pesticide distribution map, 1–2% of tap water samples exceeded Iran's MCL for CPF in this region. Numerous studies have documented the potential health risks associated with OPP exposure, including cancer, neurological disorders (particularly Parkinson's disease and depression), diabetes, and respiratory conditions (42-45). Consequently, the detection of these three pesticides in our study raises significant public health concerns. The study conducted by Shayeghi et al on the Tehran drinking water supply showed that the residues of malathion and diazinon were 4.1 and 3 mg/L, respectively, which were higher than the allowed limits (46). Karyab et al reported diazinon (0.0194 mg/L) and malathion (0.0181 mg/L) in Qazvin Province, which is in line with our findings (47). In the study of Ebrahimzadeh et al in Sistan Plain, south-east Iran, the value of diazinon was detected at 0.013 to 0.084 mg/L in drinking water wells. The mean diazinon concentration in all water resources was below the maximum permissible level established by the Canadian Guidelines for Drinking Water Quality (48). The study by Derbalah et al was carried out to monitor the presence of organophosphorus in drinking water plants in Egypt and showed the presence of several OPPs in the range of 0.070 to 2.95 mg/L. CPF has high frequency relative to other compounds in drinking water and malathion values were less than the detectable concentration (49). Diazinon is a frequently used insecticide because of its low cost and high efficacy as an acetylcholinesterase inhibitor (50). However, diazinon was detected in the surface water samples and was not observed in any of the purified water samples. Positive processes in the environment, such as UV photolysis, microbial degradation, hydrolysis, or chemical oxidation, degrade a part of pesticide concentrations. Drinking water treatment facilities such as activated carbon absorption, chlorine, UV photolysis, and ozone treatment can reduce pesticide concentrations before human consumption (51,52).

The study by Elfikrie et al in Malaysia showed the efficiency of pesticide removal in the conventional water treatment method (filtration-coagulation-flocculation-sedimentation). Before the water treatment process, nine pesticides with a maximum detection level of 0.3928 µg/L were detected in water samples. After the combined water treatment process, all the targeted pesticide compounds were found to be less than 0.1 µg/L (53). According to similarly conducted studies, in the present study, diazinon was not detected in any drinking water samples.

Table 2. Non-carcinogenic results of pesticides (HI)

Hazard Index (HI)	Distribution water network		
	Chlorpyrifos	Malathion	Diazinon
Mean	0.01	0.001	ND
Std. deviation	0.01	0.001	ND
Maximum	0.07	0.001	ND

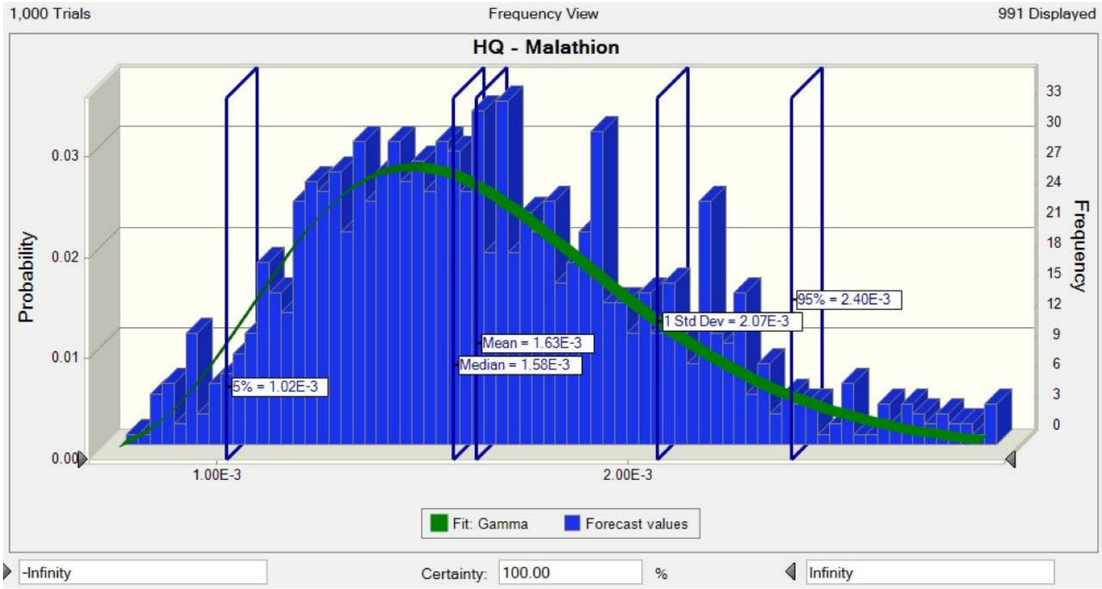

Figure 6. Simulation histogram for HQ caused by malathion exposure in drinking water

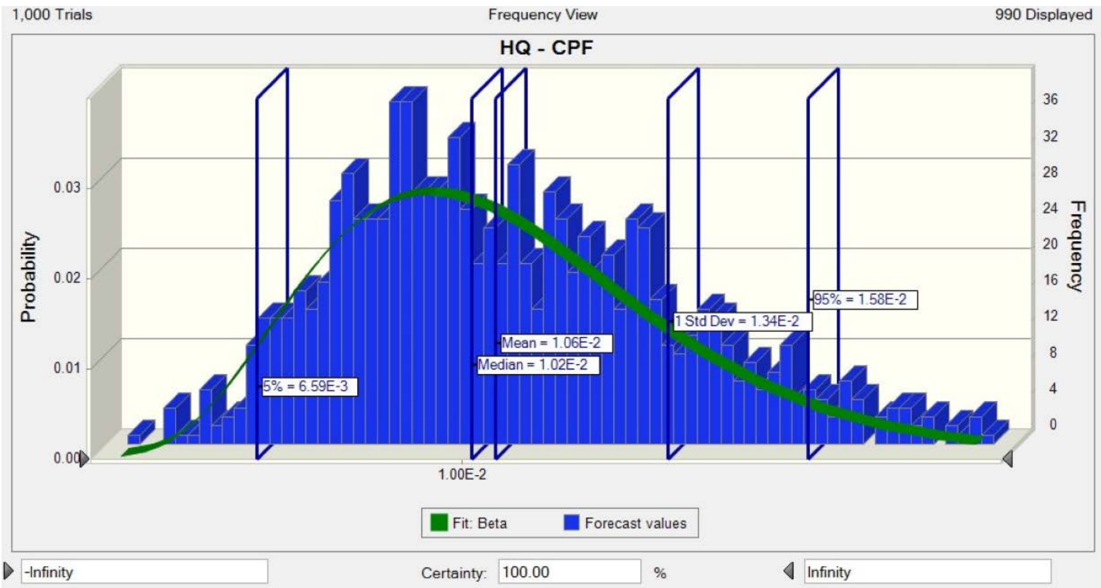

Figure 7. Simulation histogram for HQ caused by CPF exposure in drinking water

On the other hand, in the northern regions where potable water is sourced from surface water sources, the intensity of contamination with CPF and malathion is in the high range (Figure S6 and Figures 1, 2). It can be concluded that in these regions, which have agricultural uses, pesticide residues in water sources are more intense.

Most farmers use more OPPs than others because of their low cost and wide range of uses. Malathion, CPF, and diazinon have been used all over Iran (54,55). According to the results, the mean value of malathion was 0.013 mg/L in surface water samples. The study of Safari et al on surface waters in Iran, which is consistent with the results of the present study, showed high levels of diazinon and malathion (55). They found agricultural and horticultural activities in to be the reason for the high concentration of these OPP compounds, which is similar to our findings for our study area. The study by Fadaei et al on the surface water of Mazandaran, in the north of Iran, showed that the amounts of malathion and diazinon pesticides were higher than the allowed limits. European Union (EU) standards set a maximum allowable concentration of 1–3 µg/L for total pesticides in surface waters. In Mazandaran (northern Iran), the measured concentrations significantly exceeded these limits, with diazinon ranging from 77.6 to 101.6 µg/L and malathion from 55.7 to 75.9 µg/L. This substantial contamination primarily results from intensive pesticide use in rice cultivation throughout the region. (56). The study of Zarei-Choghan et al reported that the mean concentration of some OPPs (CPF, malathion, ethion, dichlorvos, trifluralin, and diazinon) in the Naseri wetland in Iran ranged from 0.14 to 0.35 and 0.054 to 0.2 µg/L in summer and autumn, respectively (57). In Chilika Lake, India, CPF was found in the range of 0.019–2.73 µg/L, which is consistent with the results of the present study (58).

In contrast, Parana, Brazil, documented widespread organochlorine pesticide contamination, including lindane (2.17 ppb) and chlordane (0.181 ppb), with 97% of municipalities exceeding EU limits (59). Similarly, North India demonstrated elevated OCPs (particularly α-HCH, p,p'-DDE) in both groundwater and tap water, with α-HCH concentrations slightly exceeding the WHO guideline of 0.001 µg/L (60). Polish research identified seasonal variations and detected 22 pesticides in river water (maximum concentration: 0.472 µg/L), including persistent compounds like isoproturon in distant filtration wells (61). Most notably, monitoring in Mexico and Iran's Gorganrood River revealed malathion concentrations dramatically surpassing WHO standards, reaching 863.49 µg/L in Mexico and 88.11 µg/L in Gorganrood during spring (62).

As can be seen in the land use map of Ardabil Province (Figure S6), agriculture has been widely developed and mechanized in the northern regions of Ardabil Province, mainly due to its suitable climate and its proximity to the Caspian Sea. Wheat and summer crops are cultivated three times a year in this area. Therefore, a high level of pesticide residues in water resources was expected, considering the many fruit orchards in the region and the use of pesticides for pests.

The highest points of pesticides in surface water located in the north and central regions are shown in Figures4 and 5. In the central areas, unlike the northern areas, there is no intensive agriculture, but it seems that domestic uses and the use of pesticides for pest control in livestock are the main reasons for pollution.

Non-carcinogenic risk assessment

Recently the HQ, which measures the non-carcinogenic risk of specific contaminants, has been used to assess the risks caused by environmental contamination such as assessment of the risk caused by metals and metalloids in marketed infant formulas (63), heavy metal contamination in soil, water, and vegetables (64), BTEX compounds (65), trihalomethanes through successive showering events (66), and $PM_{2.5}$ emissions from various sources (67).

According to the USEPA, HI values more than 1 could be over the potential toxicity. The non-carcinogenic risk results of pesticides (HI) are shown in Table 2 and Figures 6,7. The highest mean score of HI was reported for CPF with a value of 0.07. None of the samples had a noncancer risk, considering HI < 1. This is consistent with the results of some studies. In Egypt, the target HQ (THQ) values for non-carcinogenic pesticide hazards observed in surface and drinking water were less than 1 (68). In Kashmir, India, CPF was detected in surface, ground, and tap water samples with a THQ assessment for CPF greater than 1 (69). In another study in Iran, HQ > 1 for CPF exposure was obtained in 20% of pregnant women based on the USEPA guideline values (70).

Sensitivity analysis

Using a probabilistic approach, the variation in the HQ was estimated by applying the Monte Carlo technique with 1000 repetitions. HQ values, including standard deviation, mean, median, 5th percentile, and 95th percentile, were predicted using uncertainties. The main variables related to cancer risk were identified by sensitivity analysis.

The effective factors in the HQ due to exposure to high values of toxic elements were highlighted by sensitivity analysis (Figures 4 and 5). The study results indicated that the pesticide concentration for CPF and the ED of water for malathion had the most effect on noncancer risk. Also, BW, CPF, and malathion showed an inverse correlation. While pesticide exposure appears to be a satisfactory way to assess the risks pesticide exposure poses to human health, it has certain limitattions. These include changes in risk factors from day-to-day or area-to-area, differences in human exposure to pesticides, and unreliable detection tools (65,71).

Conclusion

This study investigated the concentrations of three pesticides, namely CPF, malathion, and diazinon, in both tap water and surface water in Ardabil Province. High concentrations of CPF (0.05 mg/L), malathion (0.01 mg/L), and diazinon (not detected) were detected in the distribution (tap) water network. The mean pesticide levels in tap water samples across the study area were found to be below the established standards in Iran. However, it is noteworthy that in the northeastern part of the study area, CPF levels exceeded Iran's MCL in 7 percent of the samples. The mean values of CPF, malathion, and diazinon in surface water were measured at 0.006, 0.013, and 0.003 mg/L, respectively. The highest concentrations of pesticides in surface water were observed in the northern and central regions, which had high agricultural activities. Considering noncancer risks, the HI values for drinking water samples were negligible. Sensitivity analysis highlighted that pesticide concentration, particularly for CPF, and exposure duration for malathion had the most significant impact on average daily dose (ADD) and HQ.

These findings underscore the importance of monitoring and modeling pesticide levels in public water systems to evaluate associated health risks. Furthermore, the study underscores the necessity of limiting pesticide usage in agricultural activities and promoting the adoption of environmentally friendly alternatives.

This study has several limitations. The small sample size restricted our statistical power. While the estimated risks for the selected pesticides were lower than the EPA RfD (HQ < 1.0), this does not imply that there are no health risks. Other exposure pathways, such as inhalation and dietary intake, should also be taken into account.

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Authors' contributions

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Investigation: Anoshirvan Sedigh.
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Software: Amir Mohammadi, Zahra Atafar.
Supervision: Mehdi Vosoughi, Tayebe Sadeghi, Abdollah Dargahi.
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Visualization: Amir Mohammadi, Zahra Atafar.
Writing – original draft: Tayebe Sadeghi, Amir Mohammadi.
Writing – review & editing: Tayebe Sadeghi, Amir Mohammadi, Zahra Atafar.

Competing interests

The authors declare that there is no conflict of interest regarding the publication of this work.

Ethical issues

The study was approved by the Ethics Committee of Ardabil University of Medical Sciences, Iran (Ethical code: IR.ARUMS.REC.1398.622). The authors confirm that all data were collected during the study. No data from this study have been or will be published separately elsewhere.

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Supplementary files

Supplementary file 1 contains Tables S1-S3 and Figures S1-S6.

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