

## Evaluation of phosphate insecticides and common herbicides: monitoring and risk assessment in water treatment plant, distribution networks, and underground water wells

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### ABSTRACT

Despite the negative effects that the use of pesticides (such as herbicides and insecticides) have on human health and water resources, a significant portion of the world's agricultural production depends on them. The purpose of this study was to determine selected residual concentrations of pesticides (diazinon, ethion, malathion, alachlor, methyl-parathion, trifluralin, atrazine, chlorpyrifos, and azinphos-methyl) in samples from Shiraz potable water sources. For this purpose, water treatment plant, groundwater wells, treated surface water, and a mixture of groundwater and treated surface water were taken. In addition, statistical and risk analyses (carcinogenic and non-carcinogenic) were used. According to the results, chlorpyrifos with 84.4% had the highest removal efficiency and methyl-parathion with 10% had the lowest removal rate in the Shiraz water treatment plant process. The highest mean concentration was related to azinphos-methyl (1.5 µg/L) and chlorpyrifos (0.59 µg/L) in the groundwater samples. All measured compounds in water source samples were below standard levels, except for chlorpyrifos and azinphos-methyl, which were reported in groundwater above the limit recommended by the Environmental Protection Agency (EPA). The results showed that while the selected pesticides measured had a low non-carcinogenic risk for both adults and children, malathion and trifluralin posed a high carcinogenic risk for adults.

**Key words:** herbicides, insecticides, risk assessment, treatment, water

### HIGHLIGHTS

- The pesticides were monitored in surface water, various stages of water treatment, groundwater, and the distribution network.
- Methyl-parathion with 10% had the lowest removal rate in the water treatment plant.
- Trifluralin showed the highest carcinogenic risk in the consumers of treated surface water.
- Results show that immediate steps must be taken to ensure the drinking water's quality.

## 1. INTRODUCTION

In many countries around the world, ensuring a sustainable water supply has become a challenge. The world has experienced an increase in droughts, hurricanes, and floods due to population growth, industrialization, and global warming (Kalaimurugan *et al.* 2022; Khan *et al.* 2022; Wang *et al.* 2022). These factors, along with the accumulation of sewage and pollutants in water sources, have also contributed to water scarcity problems and the water crisis (Wang *et al.* 2022).

Pesticides (including herbicides, insecticides, fungicides, disinfectants, and compounds used to control rodents) are pollutants and today, with the increase in population and the increase in demand for food and agricultural products, their use in agriculture has increased (Rezania *et al.* 2016; Wang *et al.* 2022). Although there are over 800 different types of pesticides, the most important are organochlorines, organophosphates, carbamates, and pyrethroids, as they are the most common and diverse (Wang *et al.* 2022).

Organophosphate pesticides (OPPs), which are categorized as non-persistent pesticides, typically are the most widely used in conventional farming areas, because they are effective against a wide range of pests and are cheaper than other pesticides (Wang *et al.* 2022). Through spraying, irrigation, and rainfall, agricultural pesticides can contaminate surface water resources. Therefore, indirect impacts on water sources could be harmful. Indirect impacts include the health of local residents who

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depend on these resources for their water, agricultural produce, entry into the food chain, etc. (Gissawong *et al.* 2020; Chen *et al.* 2021; Wang *et al.* 2022). OPPs are easily broken down by the environment and rapidly broken down by microbes. However, long-term exposure to OPPs in humans, even at low levels, can cause excess OPPs to bioaccumulate in the body. Long-term exposure to OPPs can lead to disease and cholinesterase inhibition, which can cause paralysis or muscle weakness (Jaipeam *et al.* 2009).

A number of authors have written about how pesticides can contaminate both surface and groundwater. As an illustration, 2,4-D, malathion, deltamethrin, atrazine, chlorothalonil, and endosulfan were found in surface water samples from Ethiopia (Teklu *et al.* 2015). An investigation into the levels of different pesticides in surface and groundwater resources was conducted in Hungary. They discovered that the samples frequently contained concentrations of atrazine (44%), acetochlor (31%), prometryn (18%), and diazinon (3%), all of which were frequently higher than the recommended levels (Suresh *et al.* 2021). Nine OPPs were discovered in the Sarno River in Italy as part of a study by Montuori *et al.* These substances included dichlorvos, diazinon, fenitrothion, dimethion, malathion, chlorpyrifos, dimethoate, and tolcumylphosphorus. Total OPP concentrations in water samples in an Italian study ranged from 5.5 to 39.2 ng/L. According to the findings, this river is most likely one of the major rivers that discharges OPPs into the Tyrrhenian Sea (Montuori *et al.* 2015). Studies have revealed that the concentrations of the pesticides under investigation, including malathion and diazinon, were higher in Argentina's Uruguay River than what the EU and US Environmental Protection Agency (EPA) have advised (Williman *et al.* 2017; Silva-Madera *et al.* 2021). In Cienega, Mexico, researchers looked at the levels of various pesticides in drinking water and surface resources. They came to the conclusion that the concentration of malathion exceeded the EPA's permissible limits in drinking water resources (100 µg/L for children and 200 µg/L for adults) (Silva-Madera *et al.* 2021).

Since pesticides are present in water sources, they need to be purified. As a result, monitoring is crucial to ensure the removal of these compounds at the treatment plant's outlet. However, studies have not focused on this issue up to this point. As a result, in Shiraz city, this study measured the levels of nine different types of pesticide residues in treated surface water (their sources were the dam), groundwater well, and a mixture of groundwater and surface water. Additionally, it has been estimated how consumers perceive their own deterministic health risks. As a result, this study adds new knowledge to the field of drinking water quality monitoring and evaluation.

## 2. MATERIALS AND METHODS

### 2.1. Study area

Shiraz, the capital of the Fars province, has a population of more than 1.8 million people, making it the fifth most populous city in Iran with a 223.4 km<sup>2</sup> area, which is located at 29°37'8" N latitude and 52°31'14" E longitude. The height of this city above sea level is approximately 1,500 m. The Shiraz aquifer flows mainly from northwest to southeast. The Iran Meteorological Organization (IRIMO) reports that the country's average annual temperature and precipitation are 18 °C and 324 mm, respectively. Deep and semi-deep wells scattered throughout the area provide roughly half of the water used for domestic and industrial purposes. Nearly 80–85% of the city's municipal water is consumed as a result of inadequate sewage systems. Among the most significant economic activities are the electronics and petrochemical industries. Although agriculture and tourism are also significant contributors to Shiraz's economy, they are not the only ones (Alamdar *et al.* 2019; Tabatabaei *et al.* 2021).

### 2.2. Sampling

Considering the effect of variations on the residues of selected pesticides (diazinon, ethion, malathion, alachlor, methyl-parathion, trifluralin, atrazine, chlorpyrifos, and azinphos-methyl), we collected samples from the Shiraz water treatment facility (the raw water entering the facility, the ozonation outlet, and the facility's final water output (after coagulation, flocculation, and disinfection units). Since there are three categories of drinking water consumers in Shiraz city, water samples also were taken from the consumption sources of the consumer groups, including: (1) treated surface water (which is supplied from dam water), (2) water from underground wells, and (3) mixed surface and groundwater. According to the WHO standards, sampling was done between April and July 2022 from a depth of 10 to 25 cm (Noble *et al.* 2020). Pre-cleaned 1.5 L polyethylene bottles were used to store the samples. Prior to being transported to the laboratory, 50 mL of methylene dichloride methane was added to each sample and it was lightly shaken to prevent the decomposition of some pesticides. The container's lid was then securely fastened and kept at 4 °C (Wang *et al.* 2022).

### 2.3. Sample extraction and analysis

Extraction and analysis of selected pesticides from water samples was done based on the study of Gang Wang *et al.* (2022). To extract the pesticides, a liquid–liquid extraction method was used. A solvent was used to wash the separation cone. In order to transfer the sample to the cone, 10 mL of it was taken. Ten millilitres of ethyl acetate was added to this volume of sample and stirred using a magnetic stirrer for 15 min. The phases were then separated. The ethyl acetate solvent phase and the lower phase, which contained water and a higher density through the decanter cone and accumulated in another container, were separated. Adding 10 mL of dichloromethane solvent to the remaining aqueous phase allowed for a second iteration of this process. The aqueous phase was then removed from the cone and thrown away after shaking the vessel to separate the phases. The solvent phase from the previous step, ethyl acetate, was combined with the solvent phase of dichloromethane. The sample containing the mixture of these two solvents was rotated at 30 °C to evaporate the solvents with mild and efficient drying. Then 200–500 µL of the ethyl acetate solvent that had been twisted into the flask was added. For the purpose of identifying selected pesticides, a vial sample was injected into a Hewlett Packard (HP) 6890 series GC fitted with an ECD (Electron Capture Detector). Helium, a carrier gas with a purity of 99.99%, was used throughout the run at a constant flow rate of 9.2 mL/min. At a combined flow rate of 60 mL/min, the gas composition was argon:methane (95:5). In the unsplit state, there was a 2 µL injection volume. The detector temperature was 250 °C, and the injector temperature was 220 °C. On a constant current mode, the ECD was run. The oven was preheated to 110 °C for 1 min, then increased to 200 °C withholding for 2 min, and then raised to 260 °C at 15 °C/min while remaining at 269 °C for 6 min. The adaptation of the sample peak retention times with calibration standards was used to determine the residue concentration. By adding 50 mg/L of the researched pesticides to water samples, a recovery test was conducted to ascertain the effectiveness of the extraction method. The recovery rates for malathion and diazinon were 94 and 97%, respectively, demonstrating the effectiveness of the extraction technique.

### 2.4. Risk assessment and uncertainty

Using Equation (1) (Wang *et al.* 2022), the average daily dose (ADD) method was employed to calculate the human exposure to selected pesticides through the direct ingestion pathway. Calculations were conducted for two groups: adults (representing the general population) and children (considered as a sensitive group).

$$\text{ADD} = \frac{C \times \text{IR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \quad (1)$$

where ADD is expressed as the average daily dose of substances taken orally, C represents the pollutant's concentration in drinking water, IR represents the rate at which water is consumed, EF represents exposure frequency, ED represents exposure duration, BW represents body weight, and AT represents the average lifespan (non-cancer:  $\text{ED} \times 365$  and cancer:  $70 \times 365$ ). The RF is equivalent to 365 days years<sup>-1</sup>.

According to the EPA, carcinogenic or cancer risks (CR) refer to 'the incremental likelihood that a person will develop cancer over the course of their lifetime as a result of exposure to a potential carcinogen'. Equation (2) is used to calculate the likelihood of cancer. A risk level of  $1 \times 10^{-6}$  has been considered the point of excess cancer risk, indicating a 1 in 1,000,000 chance of developing cancer through consumption of drinking water containing arsenic and toxic metals, estimated in mg/L for 70 years. The slope factor (SF) value (per mg/kg day) is only available for trifluralin and malathion, which were adopted from US EPA screening levels. This level must be below the safe level for carcinogenic risks. The EPA considers risks between  $1 \times 10^{-4}$  and  $1 \times 10^{-6}$  to be borderline and risks above  $1 \times 10^{-4}$  to be unacceptable. Health risks are present at a carcinogenic risk of  $1 \times 10^{-4}$ ; as such, it is sufficiently high to warrant remediation and some form of intervention.

$$\text{CR} = \text{ADD} \times \text{SF} \quad (2)$$

Equation (3) is used to calculate the risk of non-carcinogenesis:

$$\text{HQ} = \frac{\text{ADD}}{\text{RfD}} \quad (3)$$

where RfD represents the reference dose for selected pesticides and HQ represents the hazard quotient. An HQ greater than 1 is thought to indicate a significant risk. The values from Tables 1 and 2 were used to calculate the CR and HQ indexes (obtained from US EPA). The precise determination of these is.

$$HI = \sum HQs \quad (4)$$

where HI is the sum of the HQs of the selected pesticides in a mixture.

## 2.5. Statistical analysis

All analyses were carried out using SPSS 21 software. In order to verify the data distribution, the Shapiro–Wilk normality analysis was used. The values of the measured variables between water sources were compared using the Mann–Whitney *U* analysis. The figures were created using GraphPad Prism 10.2.1.395.

## 3. RESULTS AND DISCUSSION

### 3.1. Concentration of selected pesticides measured in drinking water sources

In order to determine the levels of selected pesticides in the drinking water and the risks associated with them for children and adults, we analyzed samples from different sites. The main statistics for selected pesticides are shown in Figures 1–4.

#### 3.1.1. Water treatment plant

At first, the amount of selected pesticides in Shiraz water treatment plant was checked in order to check the removal percentage of these pesticides in the treatment plant. Selected pesticide levels have been assessed in the raw water, ozonation outlet, and output of the treatment plant (after coagulation, flocculation, and disinfection units). The results presented in this study show that the treatment process was effective in removing pesticides from the water (Figure 1): Chlorpyrifos had the highest removal rate at 84.4%, followed by alachlor at 80%, azinphos-methyl = 77%, ethion = 66.6%, diazinon = 65.4%, malathion = 63%, and atrazine = 50%. In comparison to other pesticides, trifluralin and methyl-parathion were removed from water at low rates. According to studies, the removal rates for these pesticides are only 16.2 and 10%, respectively, compared to up to 50% for other pesticides. It is challenging to remove these pesticides from water sources due to their chemical makeup. Since methyl-parathion is highly soluble in water, conventional water treatment techniques struggle to get rid of it (Keith *et al.* 2017). Trifluralin and methyl-parathion also had lower removal rates in this study when compared to other pesticides, indicating that modifications to the water treatment procedure may be required to effectively remove these pesticides.

**Table 1** | Values for the parameters used for risk assessment (Wang *et al.* 2022)

	IR (l/day)	ED (years)	EF (day/year)	BW (kg)	AT (days)
Children	1	6	350	32	Non-cancer: 2,190 Cancer: –
Adults	2	70	350	70	Non-cancer: 25,550 Cancer: 25,550

**Table 2** | RfD (mg/kg day) and SF (per mg/kg day) for selected pesticides

	Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	Azinphos-methyl
Assessment type	Non-cancer	Non-cancer	Cancer/Non-cancer	Non-cancer	Non-cancer	Cancer	Non-cancer	–	–
RfD (mg/kg day)	$2 \times 10^{-4}$	$5 \times 10^{-4}$	$2 \times 10^{-2}$	$1 \times 10^{-2}$	$2.5 \times 10^{-4}$	$7.5 \times 10^{-3}$	$3.5 \times 10^{-2}$	$5 \times 10^{-2}$	–
SF (per mg/kg day)	–	–	0.26	–	–	$7.7 \times 10^{-3}$	–	–	–

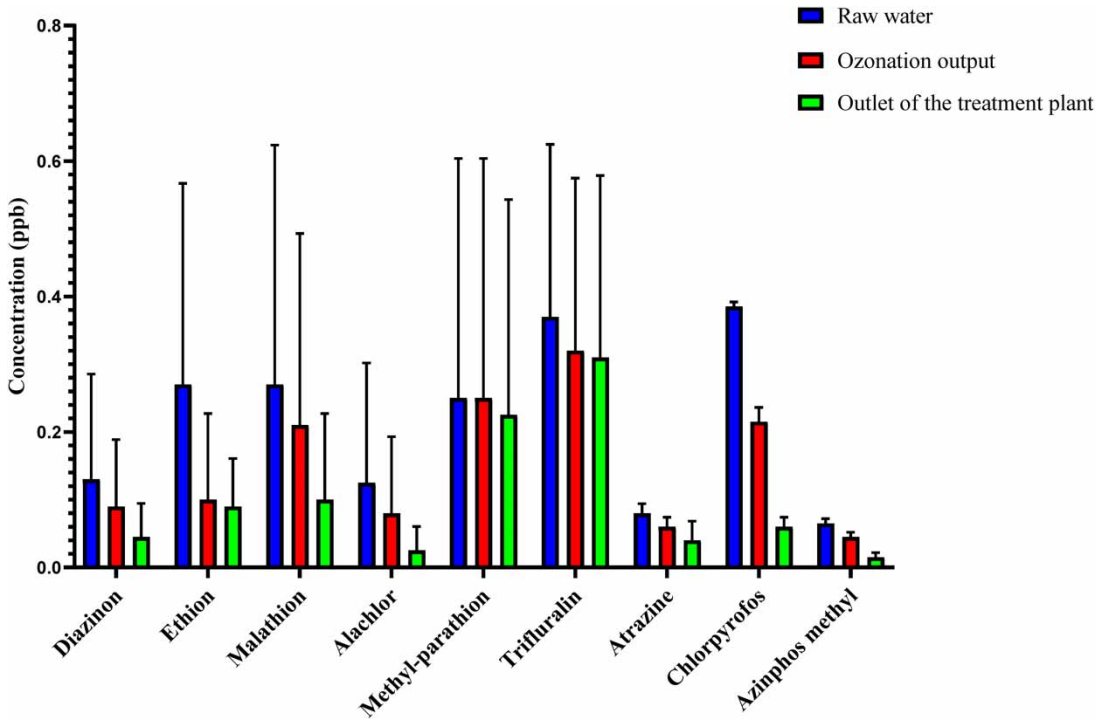


Figure 1 | Selected pesticides variation on the source of water treatment plant (Mean with SD).

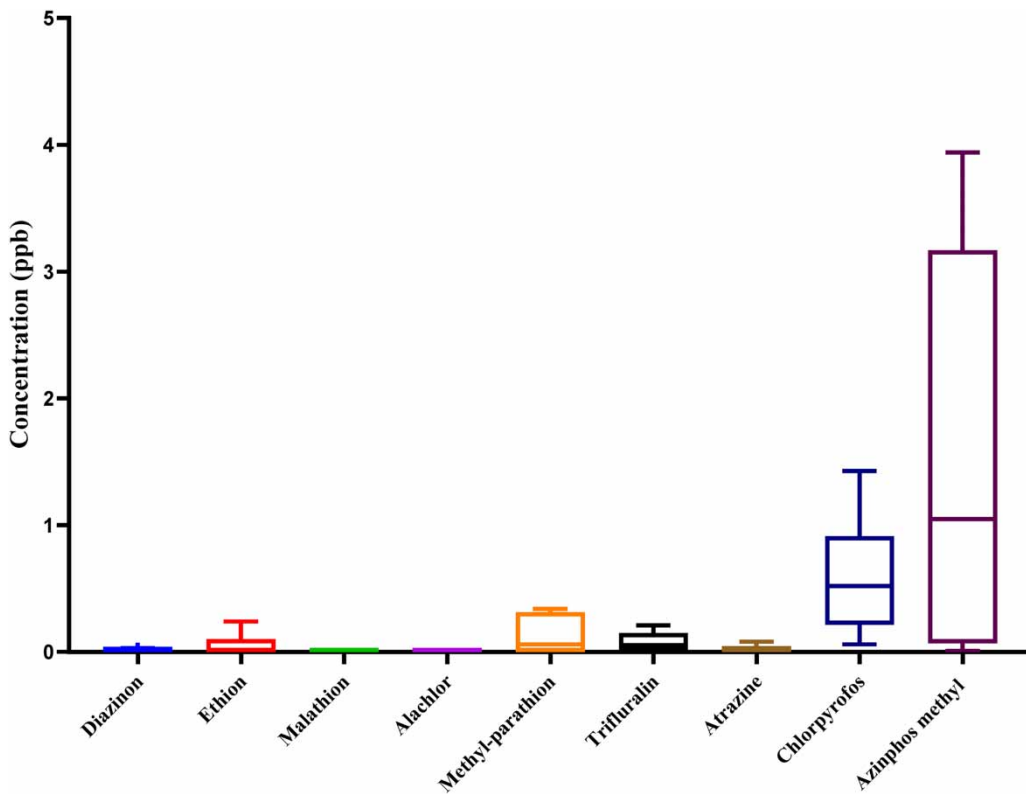
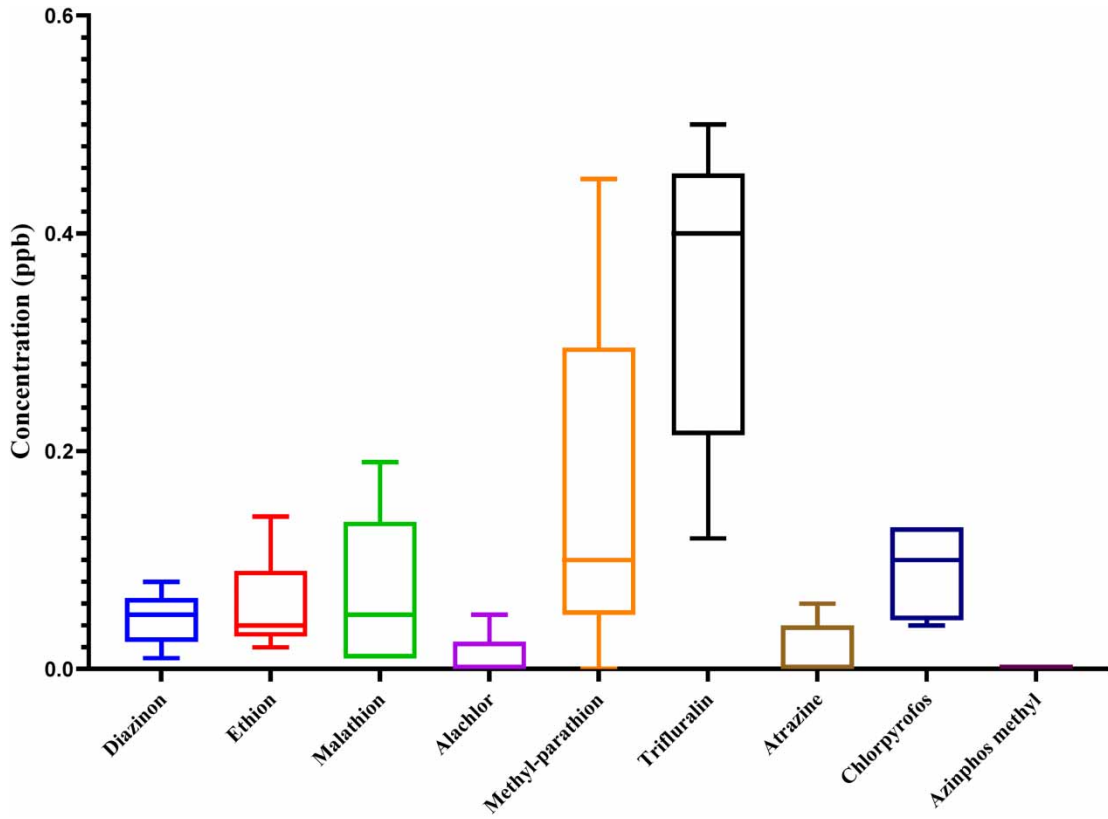
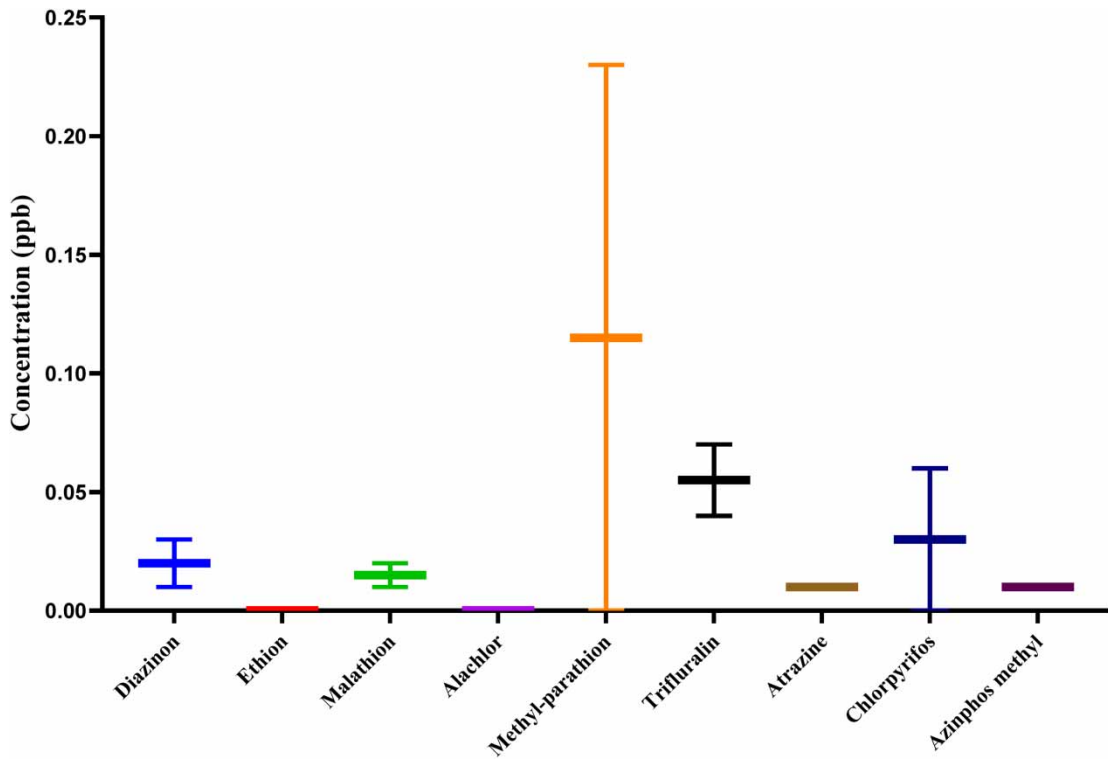


Figure 2 | Selected pesticides variation on the source of groundwater wells (10–90 percentiles).



**Figure 3** | Selected pesticides variation on the source of treated surface water (which is supplied from the water dams) (10–90 percentiles).



**Figure 4** | Selected pesticides variation on the source of mixture of surface water and groundwater (10–90 percentiles).

### 3.1.2. Organophosphate insecticide monitoring in water resources

**3.1.2.1. Diazinon.** The results of our study show that diazinon contamination (an organophosphorus insecticide) is present in treated surface water, with a concentration of 0.046 µg/L (Figure 3). The mixture of surface and underground water has a slightly lower concentration of 0.02 µg/L (Figure 4), while the concentration in groundwater wells is even lower at 0.017 µg/L (Figure 2). This study found that the concentration of diazinon, an organophosphorus pesticide with the potential for both acute lethality and chronic poisoning, is higher in treated surface water than in groundwater for a variety of reasons. A number of variables, including the type of irrigation, the amount of rainfall, the timing of the application, the characteristics of the soil, and farm management techniques, have an impact on the concentration of the pesticide diazinon, which is used extensively in agriculture and residential pest control. Diazinon is not directly applied to groundwater, which is protected from contamination by soil and other natural barriers, so there is less chance of contamination (Sosan *et al.* 2008; Wang *et al.* 2017).

Another issue is that different sources of drinking water may or may not contain diazinon, depending on where you live (Hosseini *et al.* 2021). For instance, in one study, diazinon was found to be absent from some wells used to test drinking water, while it was found to be present in surface water between 10 and 91% of the time in other locations (Zhang *et al.* 2012). One study found that after spraying, drinking water typically contained  $0.005 \pm 0.007$  µg/L of diazinon (Hosseini *et al.* 2021). Diazinon was found in Bangladeshi lakes at a concentration of 7.86 µg/L, which was higher than what we found in our study (Hossain *et al.* 2015). During an Italian study on the risk assessment of organophosphorus pesticides in the aquatic environment of the Sel river estuary, the average concentration of diazinon was 0.001 µg/L (Montuori *et al.* 2022). It is important to note that the actual concentration of diazinon in many water samples is unknown because it is lower than reported by analytical methods (Wang *et al.* 2017).

Children should not be at risk for any adverse effects from exposure to diazinon in drinking water for up to 10 days at a concentration of 20 g/L, according to the US EPA (Burgess *et al.* 2008). The drinking water equivalent level (DWEL) for diazinon is 0.04 mg/L, which is based on a reference dose of 0.0002 mg/kg day and assumes a 70 kg adult drinking 2 L of water per day (Burgess *et al.* 2008).

**3.1.2.2. Ethion.** The average concentration of ethion (an organophosphate insecticide) in treated surface water was 0.056 µg/L (Figure 3), in underground well water it was 0.052 µg/L (Figure 2), and in mixed water of underground wells and surface water it was not detected (Figure 4). The outcomes demonstrate that there is only a very slight difference in the concentration of ethion between the treated surface water and the untreated surface water. The study did not provide information on the concentration of ethion in mixed water of underground well and surface water. The dilution effect of the two water sources (Huang *et al.* 2020) may have caused a concentration below the study detection limit, in which case the concentration of ethion in the combined water may be zero. The standard level of ethion in water is not provided in the given search results.

An estimate of chronic exposure to ethion has been set by the US EPA at 1 µg/L in surface water. It is crucial to remember that this value is an estimate and that the actual level of ethion in drinking water is probably much lower than what surface water modeling suggests. The average concentration of ethion in water and any health risks connected to exposure to this pesticide need further investigation (Regulations 1983; Acrylamide 2009).

**3.1.2.3. Malathion.** According to Figures 2–4, the average concentration of malathion (organophosphate insecticide) in treated surface water was 0.068 µg/L, while the average concentration in surface and groundwater combined was 0.015 µg/L. Malathion was present in water from groundwater wells, but the amount was negligible at 0.005 µg/L. The fact that malathion enters treated surface water sources like lakes and streams through surface runoff after application explains why there are higher concentrations of the pesticide than in groundwater. On the other hand, wells and other groundwater sources are less likely to be contaminated with malathion because the pesticide must travel through layers of soil and rock to get to the water source (Brenner 1992; Hassan *et al.* 2022).

Malathion concentrations reported in other studies were significantly higher than the average concentration of malathion in surface water treated for this study (0.068 µg/L). For instance, a study on rivers and surface water in the Cienega region of Jalisco, Mexico, found that malathion concentrations ranged from 311.76 to 863.49 µg/L (Vasseghian *et al.* 2022). Between 0.11 and 138 µg/L of malathion was found on average in the water sources in Jimma and Addis Abeba (Vasseghian *et al.*

2022). It is crucial to remember that the amount of malathion in water sources can change depending on a number of variables, including the distance from agricultural fields, the use of malathion in the neighborhood, and the type of water source.

Standard levels for malathion were not found in the search results. Malathion is an insecticide that can sometimes be found in well water as well as in surface water, such as streams. The maximum residue limits (MRLs) considered by the EPA for malathion in aquatic environments were set at 0.1 µg/L (Hassan *et al.* 2022). However, a systematic analysis of malathion levels in water sources from around the world showed that its concentration in drinking water can reach 26,684 µg/L (Vasseghian *et al.* 2022), which is significantly higher than the MRL. However, the average level of malathion in water sources investigated in this study was lower than the MRL. Human and aquatic health may be affected by high levels of malathion in drinking water and surface water sources. For this reason, it is very important to pay attention to the amount of malathion in water sources and take necessary precautions to reduce its contamination.

**3.1.2.4. Methyl-parathion.** The average concentration of methyl-parathion (organophosphate insecticide) in treated surface water was found to be 0.158 µg/L (Figure 3), while in groundwater it was 0.135 µg/L (Figure 2). The average concentration of methyl-parathion in mixed groundwater and surface water was 0.11 µg/L (Figure 4). The results of this study show that all three water sources are contaminated with methyl-parathion, with the highest concentration in treated surface water.

The concentration of methyl-parathion in treated surface water is higher than in groundwater and combined groundwater and surface water because of its solubility in the aqueous phase. Water hardness, temperature, and pH are other very important factors (Memon *et al.* 2008). Another reason is that treated surface water is more environmentally exposed and more likely to be exposed to pesticides such as methyl-parathion. Surface water can be contaminated by runoff from agricultural fields where pesticides are often used. Groundwater, on the other hand, is protected by soil and other natural barriers that limit the movement of pesticides through the water. The concentration of methyl-parathion in the mixture of groundwater and surface water was lower than in the treated surface water, probably due to the dilution effect of the groundwater. In general, the concentration of methyl-parathion in water sources is influenced by several factors, such as the type of water source, distance from agricultural land, and pesticide use in the area. Several studies have reported the presence of methyl-parathion in water sources. A study in Pakistan found that 5.4% of groundwater sources were contaminated with methyl-parathion (Shakerkhatibi *et al.* 2014). According to a study in the United States, levels of methyl-parathion in natural waters near agricultural areas reached 0.46 µg/L and peaked in the summer (Organization & WHO 2004). The recommended value of methyl-parathion in drinking water is 0.2 µg/L. According to the World Health Organization (WHO; Fawell & Lund 2003), the concentration of methyl-parathion in the water samples was below the WHO drinking water guidelines. However, it is important to note that the concentration of methyl-parathion in water samples can vary depending on the location and time of sampling.

**3.1.2.5. Chlorpyrifos.** The average concentrations of chlorpyrifos (an organophosphate insecticide) in groundwater, treated surface water, and the mixed water were 0.59, 0.09, and 0.03 µg/L, respectively.

According to these results, those who consume groundwater may be more adversely affected by chlorpyrifos. Chlorpyrifos is more soil-adhesive and less soluble in low water (EPA 2001; Eaton *et al.* 2008). In addition, because groundwater moves slowly, chlorpyrifos can build up and linger there for a longer time. In contrast, surface waters are more exposed to sunlight and oxygen, which can break down chlorpyrifos and lower their concentration. Furthermore, the likelihood of surface water being diluted by rainwater and other sources increases, which can lower the chlorpyrifos concentration.

The search results show reports of chlorpyrifos concentrations in both surface and groundwater. US EPA reports that the majority of chlorpyrifos detections in surface water were below 0.1 µg/L, with a maximum reported concentration of 0.4 µg/L (Smegal 2000). In one study, the majority of measurements for chlorpyrifos in groundwater were below 0.01 µg/L, and detectable levels were found in less than 1% of the wells tested (Fawell & Lund 2003). However, other studies have found higher levels of chlorpyrifos in surface water, with minimum and maximum values of 43.46 and 79.7 µg/L, respectively (Arain *et al.* 2018). Additionally, surface water in Sri Lanka contained a maximum amount of 0.109 µg/L of chlorpyrifos (Menike *et al.* 2012). Although the average concentration of chlorpyrifos in surface water is 0.09 µg/L, which is lower than the EPA standard level, the EPA has set a standard level of 0.1 µg/L for chlorpyrifos in drinking water. Chlorpyrifos levels in groundwater typically exceed the EPA standard level by 0.59 µg/L. In general, the amount of chlorpyrifos in surface and underground water varies depending on the area and the level of pollution brought on by industrial and agricultural activities.



**3.1.2.6. Azinphos-methyl.** Azinphos-methyl is an organophosphate insecticide that is present in groundwater at a concentration of 1.5 µg/L (Figure 2). It is a light brown, waxy or white crystalline solid dissolved in a liquid carrier (Lewis *et al.* 2004). According to the sources, it has not been found in treated surface water (Figure 3). However, it has been detected in mixed surface water and groundwater at a concentration of 0.01 µg/L (Figure 4). Because azinphos-methyl has a long half-life, it readily adsorbs to soil and can contaminate groundwater (Loewy *et al.* 1999, 2006; Lewis *et al.* 2004). Therefore, the concentration of azinphos-methyl in groundwater is usually higher.

Azinphos-methyl is often found in groundwater at varying levels depending on geography and regulations. According to the California Department of Pesticide Regulation (CDPR), the maximum annual mean concentration of azinphos-methyl in groundwater is 0.40 µg/L. The US EPA sets a maximum concentration of 0.40 µg/L in groundwater. Therefore, the levels of azinphos-methyl found in the groundwater samples used in this study exceeded the CDPR and US limits (Lewis *et al.* 2004). This suggests that the groundwater sample is azinphos-methyl contaminated and may be hazardous to both human health and the environment.

It is very important to closely monitor azinphos-methyl concentrations in water sources and take the necessary precautions to ensure safe levels. Azinphos-methyl is considered an unregulated contaminant and its health effects are still under investigation (Loewy *et al.* 1999).

### 3.1.3. Selective herbicide monitoring in water resources

**3.1.3.1. Alachlor.** The average concentration of alachlor (an herbicide) in treated surface water was found to be 0.01 µg/L (Figure 3). In groundwater (Figure 2) and mixed groundwater and surface water (Figure 4), the concentration of alachlor was not reported.

The US EPA has set a maximum contaminant level (MCL) for alachlor in drinking water of 2 µg/L (Acrylamide 2009), but our results were below this limit. This herbicide can have harmful effects on aquatic life and human health at high concentrations, so a lower concentration of alachlor in surface water is a good conclusion. However, it is important to remember that even very low levels of alachlor can have negative effects over time, so water sources must be continuously monitored to ensure that they are safe. One study found that less than 10% of groundwater and surface water samples collected in a specific agricultural study area contained alachlor (Frick & Dalton 2007).

**3.1.3.2. Trifluralin.** Based on Figures 2–4, trifluralin herbicide had an average concentration of 0.34 µg/L in treated surface water, 0.076 µg/L in groundwater, and 0.05 µg/L in the combination of groundwater and surface water. The results of this study indicate that individuals who drink treated surface water are significantly more at risk than those who drink well water from the ground or a combination of well and surface water. Trifluralin is relatively immobile and does not easily permeate the soil, so it is more likely to remain in the upper layers of the soil and be transported to surface waters by runoff or erosion (Leovac *et al.* 2015; Nahar *et al.* 2023; Silburn 2023).

Trifluralin exhibits sorption behavior in water and is immobile and stable in soil, which may help explain why treated surface water has higher concentrations of trifluralin than groundwater or a mixture of groundwater and surface water. Trifluralin is an herbicide commonly used in agriculture to control weeds. The presence of trifluralin in water bodies can have negative impacts on aquatic life and human health (Johnson *et al.* 2011). Studies on animals show that trifluralin (5 mg/L) is toxic to the liver and kidneys. It is also crucial to remember that 2 µg/L is the recommended level for drinking water (Lee *et al.* 2008).

The maximum allowable level of trifluralin in water varies depending on the nation and the type of water, according to the search results that were provided. Trifluralin, for instance, has a maximum acceptable concentration (MAC) in drinking water in Canada (45 µg/L) (Toft *et al.* 1987), trifluralin levels in the samples of water used in this study are less than this amount.

**3.1.3.3. Atrazine.** Atrazine, a herbicide, was detected at an average concentration of 0.016 µg/L in treated surface water and 0.023 µg/L in groundwater. Based on the data presented, the concentration of atrazine in the mixed water was 0.01 µg/L. According to the search results, the physical and chemical properties of atrazine result in higher concentrations in groundwater than in treated surface water. Atrazine has a long half-life, low vapor pressure, low pKa, high mobility, and is stable in the environment. As a result, it accumulates in underground and surface water. Atrazine is a common agricultural herbicide that can contaminate groundwater through runoff and leaching in agricultural areas. Concentrations

of atrazine in groundwater may be higher than in treated surface water due to soil accumulation and groundwater leaching (Bakaraki Turan *et al.* 2022).

One study found atrazine levels in North American surface water to be 0.17 µg/L, in contrast to our reported result of 0.016 µg/L (Beaulieu *et al.* 2020). Different sampling locations, methods, analytical methods, seasons, and agricultural practices in the study area may be factors in the variation in reported surface water atrazine levels. Based on the results presented, EPA set a drinking water standard of 3 µg/L for atrazine (Acrylamide 2009). This is the highest level of atrazine that can be present in water without posing a risk to human health. However, a study that found higher rates of breast cancer in rats exposed to the chemical led the California Office of Environmental Health Hazard Assessment to set a public health goal of 0.15 µg/L for atrazine in drinking water (Home). The measured concentrations are within a safe range, according to the measurement data from three sources used in the current study.

### 3.2. Risk assessment

It is well known that the majority of selected pesticides used are subject to numerous transport and conversion products. As a result, they frequently enter aquatic environments via surface runoff, air drift, or soil percolation, affecting the diversity and abundance of non-target species and having complex effects on ecosystems. They also frequently do not stay at their target site, changing interactions between trophics. Many pesticides (such as pesticides and herbicides) also eventually find their way into groundwater, where their transformation products can linger for years (Sankararamkrishnan *et al.* 2005). Using a standard approach, risk analysis was determined after analyzing the data. Tables 3–5 display the findings of the risk analyses conducted on the groundwater, treated surface water, and mixed water (surface water and groundwater), respectively.

**Table 3** | Hazard Quotient and Cancer Risk for Selected pesticides in groundwater well

<b>HQ for children</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	<b>HI</b>
$2.63 \times 10^{-3}$	$3.13 \times 10^{-3}$	$7.50 \times 10^{-6}$	0	$1.61 \times 10^{-2}$	$3.03 \times 10^{-4}$	$2.04 \times 10^{-5}$	$3.55 \times 10^{-4}$	-	$2.22 \times 10^{-2}$
<b>HQ for adult</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	<b>HI</b>
$2.39 \times 10^{-3}$	$2.87 \times 10^{-3}$	$6.75 \times 10^{-6}$	0	$1.48 \times 10^{-2}$	$2.80 \times 10^{-4}$	$1.87 \times 10^{-5}$	$3.25 \times 10^{-4}$	-	$2.04 \times 10^{-2}$
<b>CR for adult</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	
-	-	$5.19 \times 10^{-7}$	-	-	$2.73 \times 10^{-4}$	-	-	-	

**Table 4** | Hazard Quotient and Cancer Risk for Selected pesticides in treated surface water

<b>HQ for children</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	<b>HI</b>
$6.94 \times 10^{-3}$	$2.85 \times 10^{-3}$	$9 \times 10^{-5}$	$1.8 \times 10^{-5}$	$1.61 \times 10^{-2}$	$1.93 \times 10^{-3}$	$8.55 \times 10^{-6}$	$5.39 \times 10^{-5}$	-	$2.80 \times 10^{-2}$
<b>HQ for adult</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	<b>HI</b>
$6.2 \times 10^{-3}$	$2.6 \times 10^{-3}$	$8.18 \times 10^{-5}$	$1.7 \times 10^{-5}$	$1.50 \times 10^{-2}$	$1.77 \times 10^{-3}$	$7.8 \times 10^{-6}$	$4.93 \times 10^{-5}$	-	$2.57 \times 10^{-2}$
<b>CR for adult</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	
-	-	$6.43 \times 10^{-6}$	-	-	$1.7 \times 10^{-3}$	-	-	-	

**Table 5** | Hazard Quotient and Cancer Risk for Selected pesticides in groundwater well in mixed groundwater and surface water

<b>HQ for children</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	<b>HI</b>
$3 \times 10^{-05}$	0	$2.25 \times 10^{-5}$	0	$1.32 \times 10^{-2}$	$2 \times 10^{-4}$	$8.57 \times 10^{-6}$	$1.8 \times 10^{-5}$	-	$1.64 \times 10^{-2}$
<b>HQ for adult</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	<b>HI</b>
$2.75 \times 10^{-3}$	0	$2 \times 10^{-5}$	0	$1.2 \times 10^{-2}$	$1.87 \times 10^{-4}$	$7.71 \times 10^{-6}$	$4.93 \times 10^{-5}$	-	$1.5 \times 10^{-2}$
<b>CR for adult</b>									
Diazinon	Ethion	Malathion	Alachlor	Methyl-parathion	Trifluralin	Atrazine	Chlorpyrifos	azinphos methyl	
-	-	$1.54 \times 10^{-6}$	-	-	$1.82 \times 10^{-4}$	-	-	-	

### 3.2.1. Non-carcinogenic risk assessment

Regarding the non-carcinogenic risk, the risk is minimal in all scenarios for all compounds (HQ was less than 1). There were no adverse health effects if the ratio of potential dose to expected adverse effects (HQ) was less than 1; if the HQ was greater than 1, then adverse health effects were possible. Among the pesticides, methyl-parathion pesticide was reported to have the highest non-carcinogenic risk for both children and adults who used groundwater, surface water, and this mixture. Due to the high HI, children and adults who use treated surface water are more exposed to the non-carcinogenic risk of selected pesticides than the group that uses groundwater and the group that uses mixed groundwater and surface water. According to a report conducted by IRAN on the contamination level and human non-carcinogenic risk assessment of diazinon pesticide residue in drinking water resources, both children and adults had lower non-carcinogenic risk of diazinon exposure through water consumption than is advised (HQ = 1). This is in line with our findings as well (Ebrahimzadeh *et al.* 2022).

### 3.2.2. Cancer risk assessment

In terms of the carcinogenic aspect, the literature indicates that there is no risk for humans with regard to diazinon, ethion, alachlor, methyl-parathion, atrazine, chlorpyrifos, and azinphos-methyl. Additionally, the SF is not available, so these were not determined in this case. According to our results (Tables 3–5), adult carcinogenic risks for malathion and trifluralin were reported in all three consumer groups, except for malathion in the group of groundwater consumers above the value of 1.0E-06 recommended by US EPA for carcinogens. The highest carcinogenic risk (1.70E-03) was observed for trifluralin in the group of consumers of treated surface water and the lowest carcinogenic risk (5.19E-07) was for malathion in the groundwater consumers.

Conclusions of a study carried out by Mekonen *et al.* examined the levels of malathion and diazinon in samples of drinking water from Addis Ababa and the Jimma zone in Ethiopia. They claimed that malathion and diazinon pose chronic risks to all consumers, but that acute risks have not yet been documented (Marín-Benito *et al.* 2014). The results of a study revealed that there was a high carcinogenic risk regarding malathion and children were at the most non-carcinogenic risk regarding diazinon (Wang *et al.* 2022).

In light of the above, it is clear that the assessment of health risks from specific pesticides in drinking water sources varies considerably around the world and is influenced by a number of variables such as pesticide loading sources, pesticide degradation rates, and weather conditions. It is also important to identify the source of pesticides pollution and control its discharge. Even though pesticides are in compliance with regulations, their potential risk should not be ignored. In order to preserve and protect aquatic ecosystems and human health, risk assessment for pesticides is essential. By assessing risk, the competent authority can make better decisions and implement policies. Thus, it is necessary to continuously investigate the risks associated with pesticides (that are used in human life) for human health. Consequently, it is important to raise awareness about the use of pesticides and herbicides by farmers across the watershed. By respecting the Legedadi pond's buffer zone and establishing buffer strips for different water sources, especially those that are used as drinking water, we can lower the risks of these pesticides. For reducing water contamination, Waskom (1994) explains that it is essential to raise awareness among pesticide users and applicators, maintain the application equipment in good working order, and avoid using pesticides improperly. Comprehensive monitoring and remediation focused on the source are needed to

reduce the long-term health risks posed by pesticide residues. Pesticides can be removed from drinking water using a variety of techniques, including ultraviolet light, ultrasonic processes, photocatalysis, solar radiation, chemical oxidation, advanced oxidation, biological treatment technologies, and more. This study evaluated the risk of OPPs, and it appears that a comprehensive examination of other pesticides, especially chlorinated ones, should be conducted in water treatment plants and a thorough risk assessment of these pesticides for consumers needs to be undertaken.

#### 4. CONCLUSION

In this study, a thorough analysis of pesticides in drinking water was conducted. According to results, although the concentration of all measured compounds was highest in treated surface water, two organophosphate insecticides, chlorpyrifos and azinphos-methyl, had the highest concentrations in groundwater.

In this study, the daily intake and exposure to specific pesticides through drinking water consumption of the two selected populations of adults and children were assessed for their potential health risks. The findings revealed that HQ for measured selected pesticides was reported to be less than one for both children and adult groups. Trifluralin showed the highest carcinogenic risk ( $1.70E-03$ ) in the group of consumers of treated surface water, and malathion showed the lowest carcinogenic risk ( $5.19E-07$ ) in the group of groundwater consumers. Residents in this study area may face greater health risks as a result, and serious attention must be paid to this issue. In order to avoid drinking water shortages or waterborne diseases that pose a threat to human health, it is anticipated that these results will assist federal and local governments in adopting new guidelines for evaluating the quality of water. Additionally, this shows that immediate steps must be taken to lower these levels and ensure the drinking water's quality.

#### AUTHOR CONTRIBUTION

A. A. T.: investigation, data curation, resources, conceptualization, methodology, and writing. Z. T.: investigation, data curation, resources, methodology, writing, original draft. A. S. B.: supervision, literature searching, writing, and editing. A. A.: supervision, literature searching, writing, review, and editing.

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The authors declare their consent to participate in this article.

#### CONSENT TO PUBLISH

The authors declare their consent to publish this article.

#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

#### CONFLICT OF INTEREST

The authors declare there is no conflict.

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