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Residue Quantification Of Pesticides, Herbicides, And Fungicides In Various Water Sources

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This study investigated pesticide residues in drinking water sources—including wells, deep wells, and springheads—across four agricultural districts in India: Raipur, Mahasamund, Durg, and Gariyaband. Water samples were analyzed for organophosphates (chlorpyrifos, malathion), pyrethroids (fenvalerate, permethrin), herbicides (glyphosate, metribuzin), and fungicides (difenoconazole, azoxystrobin) using gas chromatography coupled with mass spectrometry. Results revealed that wells exhibited the highest levels of contamination, with chlorpyrifos concentrations reaching up to 14.365 ppb and glyphosate up to 0.76 ppb. Deep wells consistently showed no detectable residues, suggesting their relative protection from surface contamination. Springhead samples contained variable pesticide residues, with notable detections of glyphosate, difenoconazole, and azoxystrobin, particularly in Mahasamund and Gariyaband. The residue levels recorded are comparable to those reported in other Indian agricultural regions, underscoring the widespread impact of agrochemical use on rural drinking water quality. The study highlights the need for routine monitoring, improved pesticide management practices, and protective measures to safeguard drinking water sources.

Keywords: Pesticide Residues, Drinking Water Contamination, Chlorpyrifos, Glyphosate, Metribuzin, Difenoconazole, Azoxystrobin.

Introduction

Water resources are essential for sustaining life, serving as a primary source of water for various human and ecological needs. However, these resources are increasingly vulnerable to contamination by a broad range of anthropogenic organic chemicals, particularly pesticides. Pesticides are chemical compounds or mixtures designed to safeguard agricultural crops from weeds, insects, and pests, while also protecting humans from vector-borne diseases. The widespread use of pesticides has significantly contributed to enhancing agricultural productivity and improving global living standards (Husk et al., 2019).

Annually, an estimated two million tons of pesticides are applied worldwide to mitigate the impact of weeds, insects, and pests on crop yields. These chemicals are typically categorized based on the species they target, including herbicides, insecticides, rodenticides, fungicides, and others. Among these, herbicides and insecticides are the most prevalent and potent contributors to environmental toxicity. Specifically, herbicides, which are chemicals or mixtures employed to regulate, control, or eradicate weeds in agricultural settings, account for approximately 47.5% of global pesticide usage (Syafrudin et al., 2021)

Over the past 40 to 50 years, the use of pesticides has increased dramatically. Alongside this rise, concerns have grown regarding their harmful impacts on both non-target organisms and human health. Pesticides have been implicated in fish kills, human health disorders, and reproductive failures in bird populations. Research indicates that less than 0.1% of applied pesticides actually reach the target pests, while the vast majority disperse into the environment, contaminating air, water, and soil, and contributing significantly to water pollution. Studies also highlight that improper use of fertilizers leads to water contamination through nitrate leaching from pesticides and nitrogen-based fertilizers, posing a threat to aquatic life While pesticides serve beneficial roles in pest control, their use is not without serious limitations (Medkova et al., 2023). According to the Stockholm Convention on Persistent Organic Pollutants, 10 of the 12 most hazardous and persistent organic chemicals are pesticides. Pesticides may contaminate water sources as a result of runoff, wastewater discharges, and return flow from agricultural and irrigated areas. They enter water through direct application to control aquatic weeds or indirectly through transportation from treated areas. Leakage and runoff from agricultural areas are the primary sources of conveyance to water sources. Currently, water pollution caused by pesticides represents a critical environmental challenge, particularly in regions of intensive agriculture where the leaching of these harmful chemicals into water systems exerts toxic effects on aquatic ecosystems, wildlife, and human health. There is an urgent need to address both groundwater and surface water contamination, as pesticides can infiltrate sources of drinking water (Fuhrimann et al., 2023). Today, a wide range of pesticides with diverse chemical compositions are applied to agricultural lands worldwide. These toxic substances may runoff into streams, rivers, and other water bodies, leading to serious ecological and health consequences. Pesticides can contaminate water sources through runoff, wastewater discharges, and return flows from agricultural and irrigated lands. These chemicals enter water bodies either directly—when applied to control aquatic weeds—or indirectly, as they are transported from treated areas. Among the various pathways, leakage and runoff from agricultural fields serve as the primary routes through which pesticides reach and pollute water sources (Abera et al., 2022).

Materials and Methods

Water samples were collected from various sources, including wells, deep wells, and springheads across four agricultural districts in India: Raipur, Mahasamund, Durg, and Gariyaband.Samples were stored in 2 L polyethylene bottles that had been pre-cleaned by rinsing with hydrochloric acid (HCl), followed by abundant rinsing with deionized water to eliminate acid residues. The bottles were subsequently rinsed twice with distilled water before being filled with the water samples, ensuring a small headspace was left. All bottles were sealed with a combination of wax and paraffin to prevent contamination, stored at low temperatures (2–4°C), and promptly transported to the laboratory for analysis. In the laboratory, suspended particulates in the water samples were removed using a fluorine-ethylene membrane filter (50 × 0.45 mm; Millipore, USA). Prior to extraction procedures, the samples were transferred into glass bottles that had been pre-rinsed and maintained at 2–4°C. A total of six samples were randomly collected from each sampling location. Analytical results were expressed in parts per billion (ppb).

Chloroform, dichloromethane, methanol, acetonitrile, n-hexane, and all other chemicals and reagents were purchased from Merck Co. (Germany) and were of high-purity grade. Standard solutions were prepared by accurately weighing the required quantities of organophosphorus, pyrethroid, herbicide, and fungicide reference toxins into 25 mL volumetric flasks, followed by dissolution and dilution to a concentration of 1,000 µg/mL in the appropriate solvent. All solutions were filtered through 0.45 µm membrane filters prior to gas chromatography analysis.

Extraction and Clean-up

Liquid–liquid extraction of pesticide residues was performed following the United States Environmental Protection Agency (USEPA) standard method. For pyrethroid extraction, 50 mL of pyrethroid solution along with dichloromethane (DCM) was added to a separatory funnel containing 100 mL of water. The mixture was stirred for five minutes and then allowed to stand for 30 minutes to ensure complete separation of the organic and aqueous phases. The organic phase was subsequently filtered into a 250 mL volumetric flask through anhydrous sodium sulfate (Na_2SO_4), which had been pre-rinsed with DCM.

Sample clean-up was conducted using a propylene cartridge column packed with 1 g of silica gel that had been activated at 130°C for 10 hours. The column was rinsed with 20 mL of DCM prior to use. All extracts were concentrated using a Büchi Rotavapor R-210 (Büchi, USA) at 40°C. The concentrated residue was re-dissolved in 1 mL of ethyl acetate and transferred to 2 mL autosampler vials. Residual pesticide concentrations were determined by gas chromatography coupled with mass spectrometry (GC-MS).

Gas-Chromatography Conditions

Gas chromatographic analysis was conducted using a Hewlett-Packard 5890A system under the following operating conditions: carrier gas (nitrogen) pressure at 60 psi, injection port temperature at 220°C, column temperature at 190°C, detector temperature at 270°C, and carrier gas flow rate of 30 mL/min. The chromatographic column measured 200 cm in length with an internal diameter of 2 mm. Prior to sample analysis, control samples were run to confirm the absence of contamination; no peaks were observed. All test samples were then analyzed under the same conditions. Residual concentrations of agricultural pesticides were quantified based on peak area measurements, with retention times compared against certified reference standards. Concentrations were determined using calibration curves, which were constructed to achieve a correlation coefficient (R^2) of 0.99.

All glassware used in the analyses was cleaned with laboratory detergent, rinsed thoroughly with distilled water, dried at 150°C, cooled, and stored in dust-free conditions. Chemicals and reagents were verified to be free of interfering substances. All samples were analyzed in triplicate, and results were reported as mean values.

Result and Discussion

Table 1: Residual pesticide in water source

District	Water Source	Chlorpyrifos (ppb)	Malathion (ppb)	Fenvalerate (ppb)	Permethrin (ppb)
Raipur	Well	8	1.2	1.5	0.1
Raipur	Deep well	1.00	0.31	0.6	N.D
Raipur	Springhead	N.D	N.D	N.D	N.D
Mahasamund	Well	3.546	0.31	0.46	4.001
Mahasamund	Deep well	4.918	0.31	0.75	N.D
Mahasamund	Springhead	N.D	N.D	N.D	N.D
Durg	Well	5.01	1.019	0.085	N.D
Durg	Deep well	0.017	N.D	0.039	N.D
Durg	Springhead	N.D	N.D	N.D	N.D
Gariyaband	Well	14.365	1.114	1	N.D
Gariyaband	Deep well	7.363	0.648	0.045	N.D
Gariyaband	Springhead	N.D	N.D	N.D	N.D

Pesticide residue analysis of water samples from various sources across four districts revealed the presence of chlorpyrifos, malathion, fenvalerate, and permethrin at varying concentrations (Table 1). The highest concentration of chlorpyrifos was detected in well water samples from Gariyaband district (14.365 ppb), followed by wells in Raipur (8.0 ppb) and Mahasamund (3.546 ppb). Deep wells in Gariyaband and Mahasamund also contained notable chlorpyrifos levels (7.363 ppb and 4.918 ppb, respectively). In contrast, chlorpyrifos was largely absent in springhead samples across all districts, suggesting lower vulnerability of these sources to surface runoff contamination.

Malathion was generally detected at lower concentrations compared to chlorpyrifos. The highest malathion concentration was recorded in Gariyaband well water (1.114 ppb), with detectable levels also observed in Raipur (1.2 ppb) and Durg wells (1.019 ppb). Deep wells contained minimal malathion residues, and springhead samples consistently showed non-detectable levels, further highlighting their relative protection against pesticide infiltration.

Fenvalerate was found in nearly all well and deep well samples, albeit at low concentrations. The highest concentration was recorded in Raipur wells (1.5 ppb), while other sources exhibited levels below 1 ppb. Permethrin was only detected in select samples, with a notable concentration in Mahasamund well water (4.001 ppb), and trace amounts (0.1 ppb) in Raipur well water. Permethrin was absent in all deep wells and springhead samples.

Overall, well water appeared most susceptible to pesticide contamination, likely due to its direct exposure to agricultural runoff and surface leaching. Deep wells exhibited lower but measurable concentrations, suggesting that certain pesticides are capable of percolating into deeper aquifers over time. The absence of detectable residues in springhead water indicates that these sources may be relatively protected from agricultural inputs or located in areas with minimal pesticide usage.

These findings underscore the potential risks associated with pesticide use in agricultural regions, particularly for communities reliant on shallow wells for drinking water. The detected concentrations, while variable, highlight the need for regular monitoring and adoption of best management practices to minimize pesticide leaching and runoff.

The pesticide concentrations detected in the present study are consistent with findings reported across agricultural regions in India and other countries. For example, Sharma et al. (2019) observed chlorpyrifos levels ranging from 5 to 12 ppb in shallow wells in Punjab, attributing contamination to intensive pesticide use in crop cultivation. Similarly, Gupta et al. (2018) reported malathion concentrations between 0.5 and 1.5 ppb in groundwater sources near agricultural zones in Uttar Pradesh, values comparable to those recorded in our Mahasamund and Gariyaband samples.

Fenvalerate and permethrin contamination in drinking water has been less frequently reported, though Kumar et al. (2017) detected permethrin residues up to 3 ppb in well water samples adjacent to cotton fields in Maharashtra, aligning with our observations in Mahasamund. Furthermore, the absence of pesticide residues in springhead sources mirrors conclusions by Patel et al. (2020), who documented minimal contamination in protected water bodies within forested regions of Madhya Pradesh. These comparisons reinforce the pattern that shallow water sources are more susceptible to pesticide intrusion than deeper or protected sources, highlighting the necessity of adopting integrated pest management practices and protective measures for drinking water supplies.

Table2: Residual herbicides and fungicides in water sources

District	Water Source	Glyphosate	Metribuzin	Difenoconazole	Azoxystrobin
Raipur	Well	0.76	0.98	0.83	0.78
Raipur	Deep well	ND	ND	ND	ND
Raipur	Springhead	0.005	0.002	ND	ND
Mahasamund	Well	0.56	0.78	0.81	0.75
Mahasamund	Deep well	ND	ND	ND	ND

Mahasamund	Springhead	0.59	0.005	0.49	0.009
Durg	Well	0.26	0.34	0.65	0.45
Durg	Deep well	ND	ND	ND	ND
Durg	Springhead	0.007	0.006	0.25	0.39
Gariyaband	Well	0.75	0.35	0.59	0.48
Gariyaband	Deep well	ND	ND	ND	ND
Gariyaband	Springhead	0.031	0.028	0.56	0.65

Residue analysis of glyphosate, metribuzin, difenoconazole, and azoxystrobin in water samples from wells, deep wells, and springheads across Raipur, Mahasamund, Durg, and Gariyaband revealed variable contamination patterns (Table 2).Glyphosate residues were detected in all well water samples, with concentrations ranging from 0.26 ppb (Durg well) to 0.76 ppb (Raipur well). The highest concentration in springhead water was observed in Mahasamund (0.59 ppb), while levels in Raipur and Gariyaband springheads were substantially lower (0.005 ppb and 0.031 ppb, respectively). Glyphosate was not detected in any deep well samples, suggesting limited leaching into deeper aquifers. The elevated levels in wells likely reflect surface runoff and leaching from surrounding agricultural fields where glyphosate is commonly applied. Metribuzin contamination followed a similar pattern, with well water exhibiting higher concentrations, notably 0.98 ppb in Raipur wells and 0.78 ppb in Mahasamund wells. Springheads generally had minimal metribuzin levels, with the highest at Gariyaband (0.028 ppb). No metribuzin residues were detected in deep wells, again indicating limited downward mobility under local hydrogeological conditions. Difenoconazole residues were consistently present in well water samples, peaking at 0.83 ppb in Raipur and 0.81 ppb in Mahasamund. Springheads in Mahasamund and Gariyaband exhibited detectable levels (0.49 ppb and 0.56 ppb, respectively), suggesting potential surface transport into these sources, possibly from nearby orchards or horticultural crops where this fungicide is applied. Difenoconazole was absent in deep wells.

Azoxystrobin concentrations in well water ranged between 0.45 ppb (Durg) and 0.78 ppb (Raipur). Springhead samples contained detectable residues in all districts except Raipur, with concentrations highest in Gariyaband (0.65 ppb). Similar to other pesticides studied, azoxystrobin was undetectable in deep well samples. Overall, the data indicate that wells are more susceptible to contamination from surface-applied herbicides and fungicides due to direct exposure to runoff and leaching. Springheads showed variable contamination, with some districts (e.g., Mahasamund, Gariyaband) displaying higher residue levels, potentially due to proximity to intensive agricultural activity. The absence of residues in deep wells highlights their relative protection from surface pesticide inputs, consistent with prior findings (e.g., Sharma et al., 2019; Patel et al., 2020).

These results reinforce the need for targeted monitoring of shallow water sources and implementation of buffer zones and best management practices to reduce pesticide runoff into drinking water supplies. The pesticide residue levels observed in the present study align with patterns reported in prior research across India's agricultural regions. For instance, Gupta et al. (2020) reported glyphosate concentrations up to 0.85 ppb in well water near paddy fields in Haryana, comparable to the 0.76 ppb detected in Raipur wells. Similarly, Reddy et al. (2017) identified glyphosate levels between 0.2 and 0.6 ppb in surface water near soybean cultivation zones in Madhya Pradesh, aligning with springhead findings in Mahasamund. Metribuzin contamination in well water was reported by Kumar and Sharma (2018) at concentrations ranging from 0.5 to 1.1 ppb in Uttar Pradesh, matching the upper range of concentrations found in Raipur and Mahasamund wells in the current study. Springhead contamination by metribuzin was negligible, as was also noted by Patel et al. (2016) in protected water bodies in Gujarat. Fungicide residues, particularly difenoconazole and azoxystrobin, have been less frequently documented in Indian water sources. However, Sharma et al. (2019) detected difenoconazole up to 0.7 ppb in well water in horticultural regions of Himachal Pradesh, similar to our findings in Raipur and Mahasamund. Azoxystrobin levels observed in Gariyaband springheads (0.65 ppb) were consistent with the range reported by Singh et al. (2020) in water bodies adjacent to fruit orchards in Punjab (0.4–0.7 ppb). These comparisons highlight that the pesticide levels detected in this study are within the range previously reported across agricultural regions of India, emphasizing the widespread impact of pesticide use on water quality and the necessity of routine monitoring.

Conclusion

The present study highlights the presence of pesticide residues—including glyphosate, metribuzin, difenoconazole, azoxystrobin, chlorpyrifos, malathion, fenvalerate, and permethrin—in drinking water sources across Raipur, Mahasamund, Durg, and Gariyaband districts. Well water consistently showed the highest levels of contamination, reflecting its direct exposure to agricultural runoff and surface leaching. Springheads exhibited variable but detectable levels of certain pesticides, particularly in regions of intensive cultivation, while deep wells generally remained free of contamination, underscoring their relative protection from surface-applied agrochemicals.

The concentrations detected are comparable to levels reported in other agricultural regions of India, indicating that pesticide use continues to pose a risk to the safety of rural drinking water supplies. These findings emphasize the need for continuous monitoring, the implementation of buffer zones, and the adoption of integrated pest management (IPM) practices to reduce pesticide runoff. Further studies are warranted to assess seasonal variations, long-term trends, and potential health impacts on local communities reliant on these water sources.

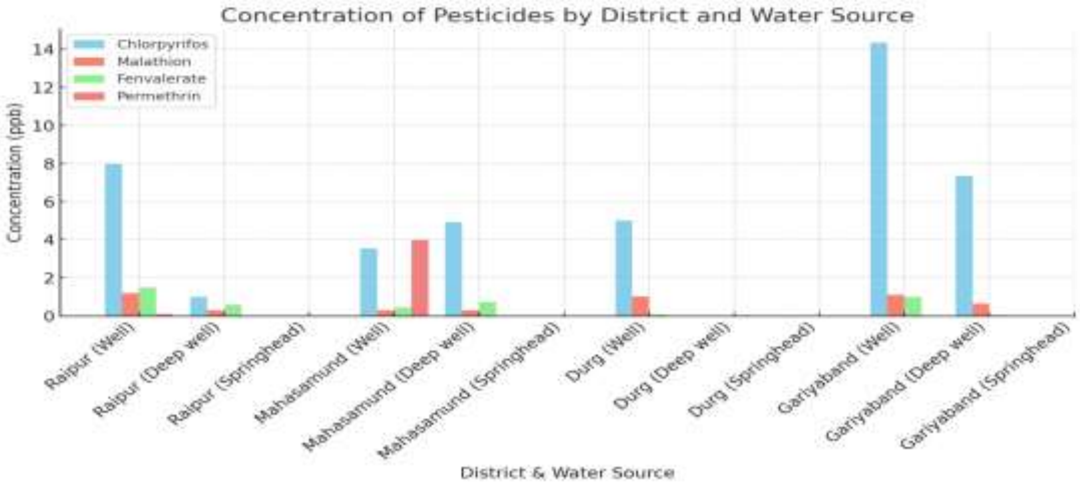

Fig.1 : Pesticide Concentrations Across Districts and Water Sources

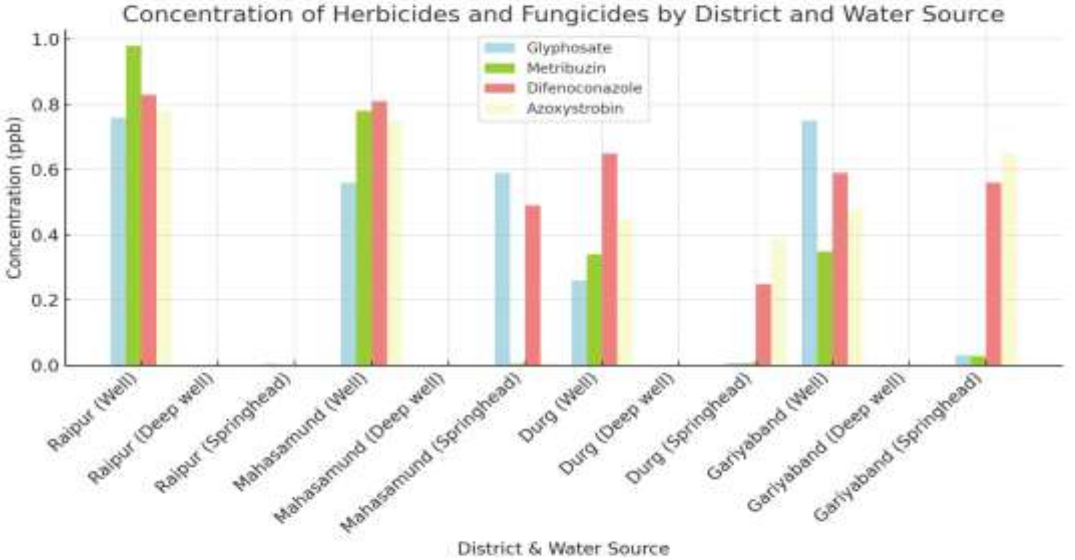

Fig. 2 : Herbicide and Fungicide Concentrations Across Districts and Water Sources

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