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Assessment of residue levels and health risk of carbofuran, chlorpyriphos, abamectin, imidacloprid, and pretilachlor in locally available fruits in Dhaka, Bangladesh

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ABSTRACT

The study addresses pesticide contamination in local fruits as a critical food safety and public health issue. Although pesticides are essential in farming, high fruit residue levels are significant risks to consumer health. This study aims to evaluate pesticide residues in various fruit samples and assess their compliance with safety standards. A total of 15 different fruit samples were collected from the temporary floating market in Dhaka for residue analysis by standard (pesticide) in HPLC. This analysis focused on measuring the concentrations of prominent pesticides in the fruit samples, including carbofuran, chlorpyriphos, abamectin, imidacloprid, and pretilachlor, and followed Codex Alimentarius and Bangladesh Food and Drugs Authority guidelines. The findings revealed significant pesticide residues in various fruits. Oranges contained 20% carbofuran, chlorpyrifos, and abamectin, exceeding allowable limits. Apples showed 60% carbofuran and 40% chlorpyrifos concentrations, exceeding the maximum permissible residue limits (MRLs) set by Codex Alimentarius. Grapes were also found to contain significant levels of carbofuran, posing potential health risks, although the chlorpyrifos residues in black grapes were within permitted thresholds. However, most fruit samples exceeded the limits set by the Bangladesh Food and Drugs Authority. Furthermore, the Estimated Average Daily Intake (EADIs) for all fruits exceeded the recommended limits established by the World Health Organization (WHO) and the Food and Agriculture Organization (FAO). While few fruit samples meet standards, results show that some exceed allowable MRLs and call for stronger regulations, better monitoring in the market, and cooperation among policymakers, producers, and health regulating agencies to balance safety, health, and sustainable farming.

Contribution/Originality: This study uniquely evaluates pesticide contamination in fruits from local marketplaces in Dhaka, focusing on the simultaneous analysis of multiple pesticides (carbofuran, chlorpyriphos, abamectin, imidacloprid, and pretilachlor). It differs from previous research by integrating Estimated Average Daily Intake (EADIs) assessments against WHO and FAO guidelines to provide a holistic view of consumer health risks and regulatory compliance.

1. INTRODUCTION

Food security has been a pressing concern throughout the world, especially in a country like Bangladesh, where considerable efforts have been made to rectify it. Fahim et al. (2021) Bangladesh's government has implemented policies like the National Food and Nutrition Security Policy and established regulatory bodies like the Bangladesh Food Safety Authority to tackle food security and safety issues (Baquedano, Zereyesus, Valdes, & Ajewole, 2021;

Hosaini, 2022; Policy & Action, 2021; Salman, Haque, Hossain, Zaman, & Hira, 2023). However, despite these efforts, food adulteration persists as a significant challenge, posing grave health risks to millions of Bangladeshis. This paper explores the current state of food security in Bangladesh, delving into the extent of efforts to address food insecurity and combat the pervasive issue of food adulteration.

According to research, nearly 4.5 million Bangladeshis contract ailments by eating tainted food each year (Yeasmin et al., 2023). The Institute of Public Health Bangladesh found adulteration in all 43 consumer products, 40% in 30 food items, and over 100% in 13 (Nasreen & Ahmed, 2014; Nishat, 2017). Coloring agents, pesticides, formalin, carbide, urea, and derivatives are common pollutants. These substances can cause cancer, inflammatory illnesses, respiratory issues, and liver and kidney damage (Rather, Koh, Paek, & Lim, 2017; Thompson & Darwish, 2019).

Pesticides are chemical compounds employed to combat or alleviate the impacts of detrimental organisms. The substances mentioned are herbicides, insecticides, fungicides, and nematicides (Pathak et al., 2022). These chemicals are specifically formulated to prevent, eradicate, deter, or alleviate pests. Long-term consumption of pesticides can lead to respiratory, cognitive, dermatological, cancerous, and neurological impairments, as well as complications during pregnancies and congenital disabilities (Chowdhury et al., 2018; Sánchez-Bayo, 2021).

Pesticide exposure in Bangladesh is rising due to the expansion of irrigated agriculture. In Bangladesh, around 80 kinds of registered pesticides are routinely used, and 170 distinct trade names know them. These pesticides are frequently used in the agricultural and public health sectors (Rahman & Khan, 2005). Food contamination with pesticides is a widespread and consequential problem in Bangladesh, which is a serious health hazard. Pesticide residue in fruit and vegetable crops and yields is caused by the farmers' overuse of pesticides without sufficient standards or safety regulations (Ahmed, Siddique, Rahman, Bari, & Ferdousi, 2019; Sarker, Islam, Rahman, Nandi, & Kim, 2021). The volume of people suffering from foodborne maladies in Bangladesh is alarmingly high, according to multiple reports, including those from the Food and Agriculture Organisation (FAO) and the World Health Organisation (WHO) (Al Banna et al., 2021). It has been recognized that the use of pesticides in food is a significant cause of the increasing rates of cancer, liver disease, and renal disease in the nation (Rahman, Sultan, Rahman, & Rashid, 2015).

The main objective of the analysis of fruit samples by High-Performance Liquid Chromatography (HPLC) was to detect the residue of Carbofuran, Chlorpyriphos, Abamectin, Imidacloprid, and Pretilachlor pesticides. The study also investigated whether the levels of pesticides in fruit samples exceeded safety guidelines and the health effects of eating pesticide-contaminated fruit. Understanding these processes might improve regulatory policies and agricultural practices and reduce the risks to people's health.

2. MATERIALS AND METHODS

2.1. Sample Collection

A total of 15 samples of different types of fruit, such as apples (5), oranges (5), Grapes (2), and Sweet Orange (Malta) (3), were directly collected from a temporary floating fruit seller (on a rickshaw hawker) near Ananda Bazar, Gulistan of Dhaka, Bangladesh. The samples were labeled and transported to the laboratory. They were kept at -20 degrees Celsius until analysis.

2.2. Apparatus

The extracts of the sample were analyzed using High-Performance Liquid Chromatography (HPLC, LC solution software) (Ultra-Fast Liquid Chromatography, Shimadzu Corporation, Kyoto, Japan) equipped with an Ultraviolet UV detector (Spectral Photometric Detector (SPD)-10A) and an autosampler.

2.3. Reagents and Chemicals

The HPLC-grade solvents were used to prepare the mobile phase. The chemicals and reagents used in the analytical-grade study were purchased from local vendors.

2.4. Mobile Phase

The mobile phase consisted of a blend of acetonitrile and 1 mM phosphate buffer (pH 4.5) in a 75:25 (v/v) ratio. A phosphate buffer solution with a concentration of 1 mM was made by dissolving 178 mg of disodium hydrogen phosphate in water and diluting it to a final volume of 1000 mL. Orthophosphoric acid was used to alter the pH to 4.5. The mobile phase was passed through a 0.45 µm Millipore membrane filter and treated with an ultrasonic bath sonicator for 15 minutes before utilization. The diluent was used as the mobile phase to prepare the stock standard solution.

2.5. Standard and Sample (Fruits) Preparation and Analysis

Working standard solutions (equivalent to 2 to 12 µg/ml) of Carbofuran, Fipronil, and Pretilachlor were prepared by appropriate dilution of the stock standard solution with the mobile phase.

20 g of each fruit sample finely crushed by mortar and pestle. Then, the mixture of solvent (50mL) at the ratio ethyl acetate: hexane: acetone in 30: 10: 10 to the fruit samples. Then, vortex and sonicate the mixtures. Then, 10g of $MgSO_4$ and 0.05g of silica gel were added to the sonicated mixtures, and the mixtures were filtered with the Whatman-1 filter paper. The filtrates were taken to the test tubes and centrifuged at 3000 rpm (10 min). Then, the supernatant was concentrated by a rotary evaporator. About 2mL of acetonitrile was added to the fruit samples, sonicated again, taken to the vials, and analyzed by the HPLC.

2.6. Chromatographic Conditions

These analyses were carried out with the following chromatographic conditions.

Column: Phenomenex C18 analytical column (250 mm × 4.6 mm I.D., 5µm particle size).

Flow rate: 1 mL/min.

Column temperature: 25^0 C.

Injection volume: 20 µL.

Detection wavelength: 230 nm.

Mobile Phase ratio: Acetonitrile: Buffer 75:25.

2.7. Data Analysis

Pesticide residue levels were measured in mg/kg and compared to the Codex Alimentarius MRL and the Bangladesh Food & Drug Act requirements. Items exceeding Maximum Residue Limits (MRLs) were considered to have violated the regulations, while those meeting or falling below the MRLs were not in violation. Microsoft Excel version 21 (Microsoft Corporation) and STATA version 18.00 (StataCorp, College Station, Texas, USA) were used to calculate the proportion and median of pesticide residue levels in several types of fruits and figures were generated using R programming software (V.4.2.1).

2.8. Recovery Study

The quality assurance materials were developed by adding 0.01-0.1 mg/kg of the desired pesticides to fruit samples containing no pesticides. Each analytical batch was subjected to material analysis to confirm the reliability and correctness of the results. The retrieval of pesticides in the quality assurance materials fell within the 80-120% range, and the allowable relative standard deviations (RSDs) for repeated measurements were established to be less than 20%. The used approach had an enlarged uncertainty of U ≤ 25%.

Table 1. Residual levels of carbofuran chlorpyriphos abamectin imidacloprid pretilachlor in orange, apple, sweet orange (Malta), grape, and black grape samples.

Concentration (mg/kg)					
Sample	**Carbofuran**	**Chlorpyriphos**	**Abamectin**	**Imidacloprid**	**Pretilachlor**
Standard	15.630	18.23	0.084	0.433	1.1233
MRL	0.500	1.00	0.01-0.05	0.5-1.0	0.01-0.05
O-1	4.442 ± 0.258	BDL	BDL	BDL	BDL
O-2	BDL	BDL	1.224 ± 0.281	BDL	BDL
O-3	1.359 ± 0.0358	BDL	BDL	0.696 ± 0.0456	0.0589 ± 0.091
O-4	BDL	2.33 ± 0.111	0.171 ± 0.0863	BDL	BDL
O-5	BDL	1.568 ± 0.0160	BDL	BDL	BDL
A-1	BDL	BDL	BDL	1.589 ± 0.128	BDL
A-2	2.371 ± 0.162	BDL	BDL	BDL	BDL
A-3	BDL	1.025 ± 0.221	BDL	BDL	0.0313 ± 0.071
A-4	BDL	BDL	0.461 ± 0.085	BDL	BDL
A-5	BDL	BDL	BDL	BDL	BDL
M-1	BDL	BDL	BDL	2.587 ± 0.010	BDL
M-2	3.578 ± 0.1140	1.325 ± 0.088	BDL	BDL	3.343 ± 0.037
M-3	BDL	BDL	BDL	BDL	BDL
G-1	3.436 ± 0.470	BDL	0.130 ± 0.032	BDL	BDL
BG-1	BDL	2.897 ± 0.286	BDL	0.023 ± 0.0298	BDL

Note: The mean ± standard deviation (SD) values of triplicate measurements represent the results. Here, O=Orange, A=Apple, M=Sweet Orange(Malta), G=Grape, BG=Black grape; BDL = Below the detection limit (0.001 ppm); MRL = Maximum residual limit determined by EC regulation 396/2005 (From EU pesticide database) and food and drug act (Bangladesh) (EU Pesticide Database, 2005).

3. RESULTS

3.1. Compliance with Maximum Residue Limits (MRL) in Sampled Fruits

The analysis of pesticide contamination in orange, apple, Sweet Orange (Malta), grape, and black grape samples shows different levels of concern. The presence of carbofuran contamination, particularly in oranges and grapes, presents substantial health hazards, while the use of chlorpyrifos in apples gives rise to worries. Abamectin contamination is within permissible thresholds, whereas the quantities of imidacloprid present represent little acute health hazards. Nevertheless, pretilachlor contamination in oranges and Sweet Orange (Malta) raises significant health concerns. (Table 1).

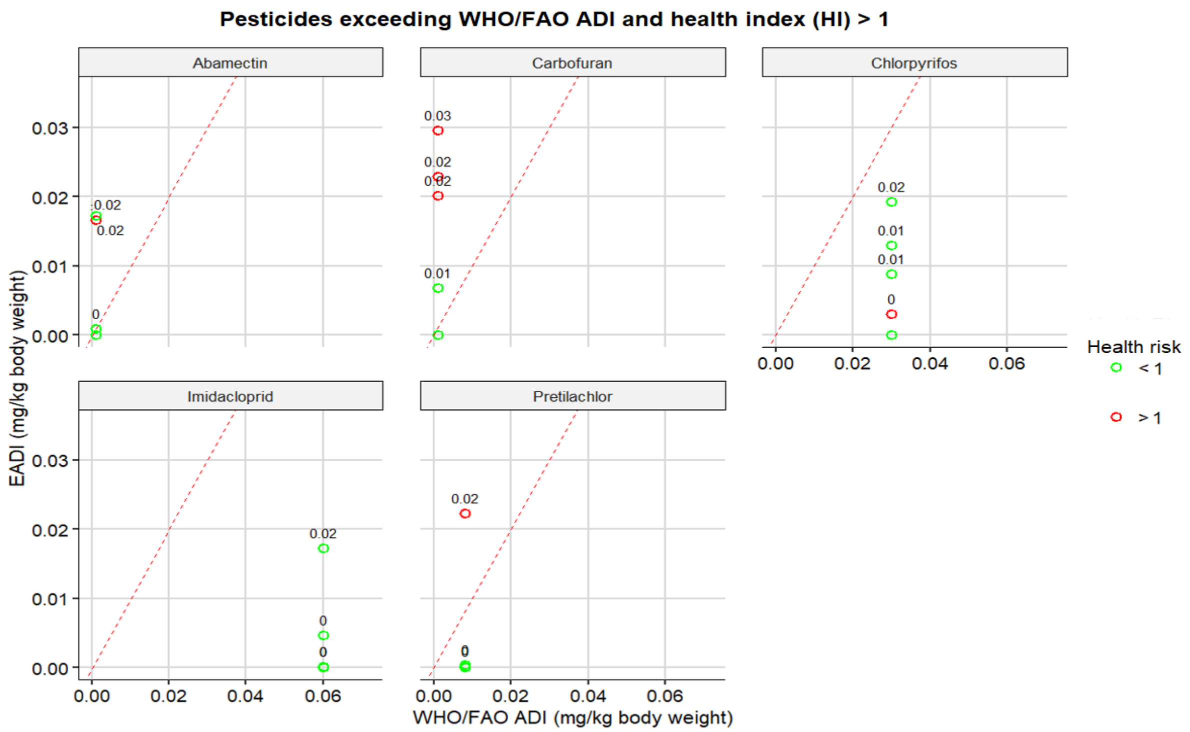

Figure 1. Overview of pesticides exceeding WHO/FAO acceptable daily intake (ADI) limits and health index (HI > 1).

Note: HRI = Human risk index; ADI = Acceptable dietary intake; EDI = Estimated dietary intake. The HRI was calculated by considering an average daily vegetable intake for a 60 kg adult as 0.400 kg/Person/Day.

The data presented in Figure 1 indicates variations in the levels of pesticide contamination across different kinds of fruits. Oranges exhibit carbofuran contamination at a concentration of 20%, chlorpyrifos at 20%, and abamectin at 20%. Carbofuran and abamectin exceed the World Health Organization/Food and Agriculture Organisation (WHO/FAO) recommended daily intake (ADI) and are health hazards. Contamination levels of imidacloprid and pretilachlor in oranges are currently 40% and 20% below the acceptable daily intake (ADI) standard, respectively. Carbofuran and chlorpyrifos levels in apples are high, with 60% of apples contaminated with carbofuran and 40% with chlorpyrifos. These levels are above the acceptable daily intake (ADI) and are hazardous to health. Abamectin contamination in apples is 40%, and imidacloprid and pretilachlor are 20%. These levels are below the Acceptable Daily Intake (ADI) standard. The exhibit in Sweet Orange (Malta) shows that carbofuran contamination is 20% and pretilachlor contamination is 20%. These levels are above the Acceptable Daily Intake (ADI) and are, therefore, harmful to health. Grapes are the most contaminated with carbofuran, with 100% potential health hazard. Grapes, however, do not contain chlorpyrifos contamination. The contamination level of Abamectin is 100%, however it remains below the Acceptable Daily Intake (ADI). Grapes are free from imidacloprid contamination, and the level of pretilachlor contamination is negligible, falling below the acceptable daily intake (ADI). The black grapes show no presence of carbofuran contamination. However, they exhibit chlorpyrifos contamination, below the Acceptable Daily Intake (ADI) but present in 100% of the samples. Similarly, imidacloprid is present in 100% of the samples but below the ADI.

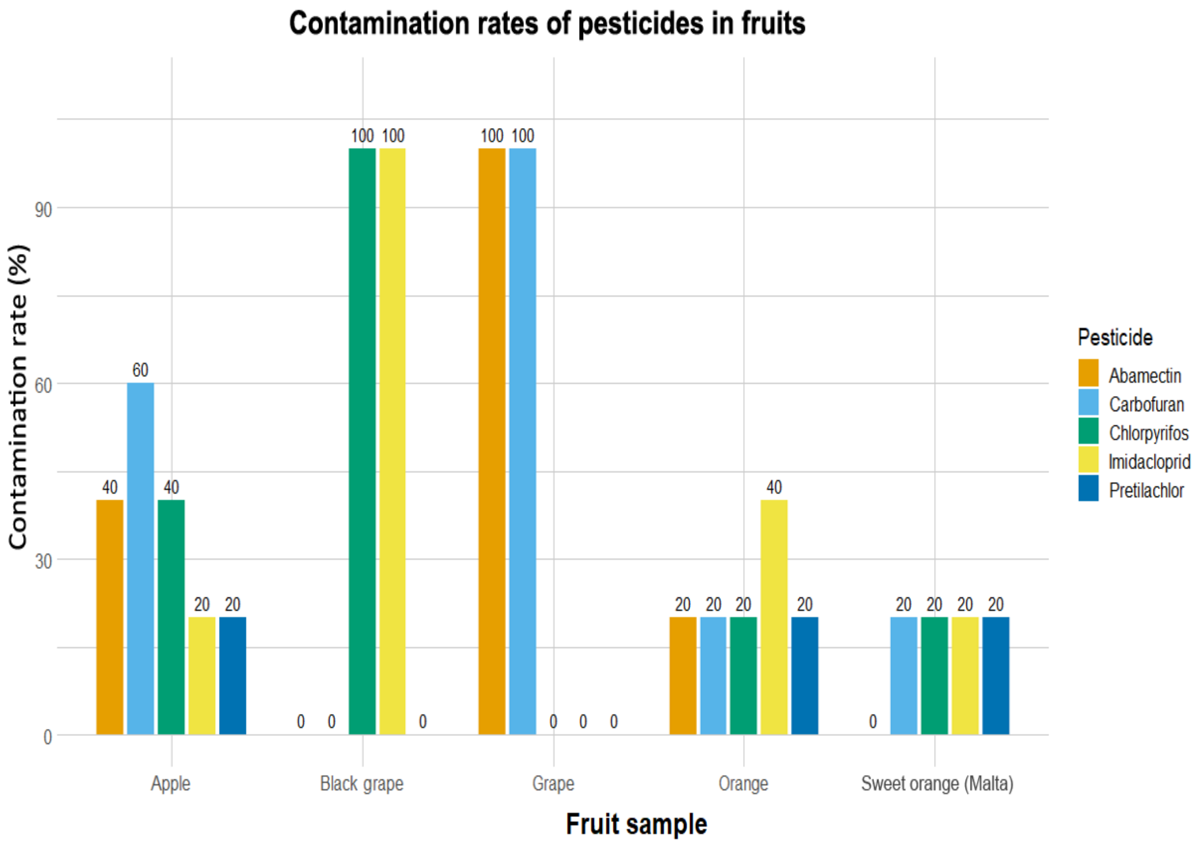

Figure 2. Pesticide residual contamination in several types of fruit samples.

The Figure 2 illustrates that Grape shows the most elevated levels of contamination among all pesticides, with Carbofuran, Chlorpyriphos, and Abamectin reaching a maximum of 100% contamination. Black Grape has varying amounts of contamination, with Imidacloprid reaching 100% contamination, while Abamectin shows cases of 0% contamination. Pretilachlor reliably maintains a contamination level below 20% for all varieties of fruit. Both apples and sweet oranges (Malta) show modest amounts of contamination from several pesticides.

4. DISCUSSION

Analysis of Carbofuran, Chlorpyriphos, Abamectin, Imidacloprid, and Pretilachlor residual concentrations in fruits indicated that all the gathered samples were contaminated. This outcome is concerning as all the contamination levels exceeded Bangladesh's Food and Drug Act standards and EC regulation (de Kok Food et al., 2007). All contaminated samples contained multiple residues.

Carbofuran is the main chemical component detected in orange samples, showing significant contamination. A published article has reported Similar findings in crops (Alam et al., 2015). Pesticide residues in fruits, a pesticide banned and/or restricted in other countries, is a severe concern and threatens consumers' health. Carbofuran, an insecticide and nematicide, was prohibited by the US EPA in 2009 due to considerable risks to food and water (Agency, 2011).

The contamination rates of chlorpyrifos in Sweet Orange (Malta) and Black Grapes are considerable, suggesting a significant presence of the pesticide in these products. Chlorpyrifos has the potential to pollute soil, water, and air, which can lead to detrimental effects on ecosystems and species. The issue at hand is concerning due to the established connection between chlorpyrifos and a range of health issues, such as neurological effects, persistent developmental disorders, and autoimmune disorders (Hongsibsong et al., 2020; Nandi, Vyas, Akhtar, & Kumar, 2022; Risher & Navarro, 1997; Wołejko, Łozowicka, Jabłońska-Trypuć, Pietruszyńska, & Wydro, 2022).

Grape samples have been found to contain abamectin. Recorded levels of abamectin in grape samples have been found to exceed the maximum residue limits (MRLs) (Authority et al., 2018). However, a study conducted in the Journal of Pharmaceutical and Pharmacognosy Research revealed the levels of abamectin in Moscatel grapes of the Valley of Ica-Peru, beneath the Maximum Residue Limit (MRL) of 0.010 ppm (Alvarado et al., 2023).

Orange, Apple, Sweet Orange (Malta) and Black Grape samples contain imidacloprid. Other researchers have reported similar findings (Aćimović et al., 2014). Pretilachlor has significant effects on orange samples and can cause many side effects, such as nausea, vomiting, neurological problems, and even death. Its mode of action inhibits the formation of fatty acids, proteins, lipids, and flavonoids, and long-term exposure to this neurotoxic, genotoxic, and carcinogenic herbicide poses these risks. (Nepali et al., 2023).

Analysis of the fruit samples showed elevated levels of pesticides, where several of the samples had residues from more than one type of pesticide. Pesticides in food can cause various adverse health effects depending on the pesticide, the level and duration of exposure.

Organophosphates, such as Carbofuran, Chlorpyrifos, Abamectin, Imidacloprid, and Pretilachlor, caused symptoms of pupil constriction, increased urination, diarrhea, excess sweating, tearing, enhanced arousal, and excessive salivation. The finding that the fruit samples contain multiple pesticide residues emphasizes the potential health risks from their exposure (Gupta, 1994). Symptoms of exposure to organophosphates, including Carbofuran, Chlorpyriphos, Abamectin, Imidacloprid, and Pretilachlor, include constricted pupils (miosis), increased urination, diarrhea, excessive sweating (diaphoresis), excessive tearing (lacrimation), heightened arousal (excitation), and excessive saliva production (salivation) (Moore, 2009).

Sometimes, uneducated itinerant sellers resort to excessively applying pesticides to their fruits to minimize losses caused by spoilage. The degrees of exposure to customers may vary when they arrive at their homes and undergo appropriate washing.

Further extensive research should be conducted to ascertain the quantities of various pesticide residues on additional vegetables and fruits sourced from multiple regions in Bangladesh at several sample intervals. It is essential to regularly monitor the use of popular pesticides on vegetables because this study showed high levels of residual pesticides present, which could be a health risk. Bangladesh should embrace the implementation of comprehensive legislation to regulate pesticide use and enhance knowledge and technical expertise among the farming community.

5. CONCLUSION

In conclusion, pesticide contamination in various fruit samples shows the complex relationships between agriculture, food safety, and public health. Most fruits meet regulatory pesticide limits, but some exceed them, showing the unpredictability of pesticide contamination. It underlines the requirement for keeping defensive measures and stringent well-being norms to limit well-being dangers resulting from the causes of pesticide deposits. Nevertheless, available data may not adequately reflect the realm of pesticide contamination, and habits of street vendors, commonly ill-informed, economically disadvantaged, and low educated, can often lead to inconsistent and frequently abusive use of pesticides according to personal preference. Elevated Hazard Quotients heighten the need for detailed risk assessments and regulatory action in some fruits. Policymakers, food safety/ public health authorities, and agricultural stakeholders must join efforts to address these challenges to ensure food safety and public health. With a focus on sustainable pest management and open communication, we can protect consumer health in an ever-changing agricultural scene.

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Authors' Contributions: Conceived and designed the experiments; wrote, analyzed, and interpreted the data, M.M.U.H.; conceived and designed the experiments; wrote, S.A.; monitoring, wrote and supervised the study, M.Z.S. All authors have read and agreed to the published version of the manuscript.

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