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Original Research

Analysis of organophosphorus pesticide residue content in brinjal and cucumber vegetables in the Narsingdi district in Bangladesh

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Abstract: This study investigated the prevalence of organophosphorus (OPs) pesticide residues in brinjal (eggplant) and cucumber samples collected between September 2021 and June 2022 in Narsingdi District, Bangladesh. A total of 36 unwashed vegetable samples (18 samples of brinjals and 18 samples of cucumbers) were collected from three sources, namely farmers' fields, retail markets and a controlled export zone. The samples were analyzed for residues of four OPs pesticides, viz. diazinon, dimethoate, malathion and chlorpyrifos, using the QuEChERS method followed by Gas Chromatography-Mass Spectrometry (GC/MS) technology. The results revealed a concerning level of pesticide contamination, particularly with dimethoate. Over half (58%) of all samples contained dimethoate, of which more than a third (39%) exceeded the European Union's Maximum Residue Limits (MRL). Chlorpyrifos was also detected in 50% of the samples, but only 8% of the samples exceeded the MRLs. Malathion residues were found in a small number of samples (11%), all of which were within the MRLs. Notably, diazinon was not detected in any of the samples. Cooking (boiling at 100°C for 30 minutes) reduced the pesticide content in some samples, but this was not always sufficient to bring the pesticide content below the MRLs. This highlights the limitations of cooking as a sole decontamination method. Encouragingly, no pesticide residues were found in samples collected from the export zone, indicating stricter adherence to regulations in these controlled environments. These findings highlight the potential health risks such as nervous system disorders, vomiting, blurred vision, coma, etc. and even death associated with consuming vegetables contaminated with pesticide residues above the recommended limits. These factors emphasize the need for stricter regulations on the use of pesticide, the promotion of Integrated Pest Management (IPM) techniques and consumer education on safe prewashing practices.

Keywords: Chlorpyrifos, Diazinon, Dimethoate, Malathion, MRL levels, Raw and cooked vegetables

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Introduction

Agriculture is a main sector for Bangladesh that contributes to food production. Among the different crops in Bangladesh, vegetables are vital parts of a healthy diet, and vegetable production increased by 450% (2.9 to 15.95 million tons from 2008 to 2018) in 2018 compared to the last decades [1]. The use of pesticides plays a crucial role in increasing the production of these vegetables. However, the journey from farm to table can involve a hidden risk of pesticide residues. Farmers in Bangladesh largely rely on different kinds of pesticides that are crucial for protecting crops from pests and diseases, but some of these chemicals can linger on produce after harvest. Currently, major concerns about the potential health effects of consuming fruits and vegetables with higher concentrations of pesticide residue are raised. Farmers use these pesticides without maintaining the proper dose or harvesting interval. Improper use of OP pesticides on farmland can have short-term, intermediate and even long-term effects on human health such as headaches, burning eyes, pain in muscles, joints or bones, rashes or itchy skin and blurred vision [2]. Ensuring safe food is one of the most important pillars for achieving food security [3]. Considering public health, there is a threshold recommended by international regulatory authorities that specifies the Maximum Residual Limit (MRL) in foods after harvesting or processing [4]. There are guidelines for harvesting time after the application of pesticides, but farmers do not adhere to the recommended time intervals and doses [5]. As a result, residues remain in foods even after cooking. Several studies have been conducted on raw vegetables and fruits, but very few studies have been performed after cooking, especially in Bangladesh.

Brinjal and cucumber are very popular vegetables consumed in Bangladesh [6]. However, considering the production process, both of these vegetables, especially brinjal, are highly susceptible to insect infestation [7]. To protect these crops from pest infestation, farmers are increasingly using different groups of pesticides such as organophosphorus (OPs), pyrethroids, carbamates and organochlorines. Sometimes they spray these pesticides just before marketing. Among these pesticide groups, OPs pesticides, such as chlorpyrifos, dimethoate, diazinon and malathion, are the most common [8]. This study focused on pesticides in the OPS group, which are widely used to control insects and mites on vegetables. OPs are absorbed by humans when they eat or touch treated crops. Although OPs are effective in killing a broad range of pests, residues of OPs can remain on vegetables, fruits, grains and other plants. It is crucial to control the amount of OPs used in vegetables to safeguard human health [9-10]. These pesticides target the nervous system by inhibiting an enzyme called acetylcholinesterase [11]. Exposure to high levels of OPs can lead to headache, dizziness, nausea and even death [12].

Extensive use of these pesticides without maintaining the dose and considering the harvest interval makes the food system more vulnerable. This research aimed to measure the amount of OPs pesticide residues before and after cooking of selected vegetables from Narsingdi district of Bangladesh.

Materials and methods

Survey site and period

This study was conducted from September 2021 to June 2022 at the National Food Safety Laboratory, Institute of Public Health (IPH), Ministry of Health and Family Welfare, Government of the People's Republic of Bangladesh. The study samples were collected from three different upazilas of Narsingdi district, namely Belabo, Raipura and Narsingdi Sadar in Bangladesh (as shown in Figure 1).

Study sample

Samples of the most popular vegetables brinjal and cucumber (unwashed) were collected randomly, 36 samples in total, with each sample being collected 3 times at every visit. The samples were fresh (raw), cooked (30 minutes at 100°C) and boiled without any spices, salt, or any other ingredients. The samples were collected from three different sources, namely farmers, retail market and the control area of the export zone in Narsingdi district. The minimum weight of each sample was 500 gm (fresh weight).

Selection of pesticides

To measure the amount of pesticide residues, four different pesticides from the OPs group were considered which were diazinon, dimethoate, malathion, and chlorpyrifos.

Reagents

Each standard for pesticides was acquired from Dr. Ehrenstorfer Co. (Augsburg, Germany). Acetonitrile, toluene, sodium chloride, PSA Bond silica (primary and secondary amine), graphite carbon black (GCB) and anhydrous magnesium sulphate were obtained from Sigma-Aldrich.

Apparatus

An Agilent 7683B autosampler (Agilent Technologies, USA) and a (single quadrupole) SQ detector were employed in the Agilent Technologies 6890 Network GC System chromatograph (Wilmington, USA). A capillary column built with HP-5 (30 m × 0.25 mm I.D., 1 µm film thickness)

Sample extraction and determination of pesticide residues

Total analysis was performed at the Food Safety Laboratory of the Institute of Public Health (IPH). The Maximum Residue Limits or tolerances set by the European Union were compared with the results. The OPs pesticides were analyzed for both raw and cooked vegetable samples. The extraction and clean-up were performed according to the method of Quick Easy Cheap Effective Rugged Safe (QuEChERS) [13]. Gas chromatography/mass spectrometry (GC/MS) was used for the quantification and detection of pesticide residues.

GC/MS analysis

The collected vegetable samples were chopped and blended separately in an electric blender with micro cutters to obtain an isolated homogeneous mixture of vegetables. In a 50 mL centrifuge tube, an aliquot of 10 g of homogenized sample was mixed with 10 mL of acetonitrile. The mixture was vortexed for one minute followed by adding 4 g of magnesium sulphate and 1 g of sodium chloride. The sample was centrifuged at 5000 rpm for 5 minutes and the supernatant was removed for purification. During clean-up, 2 mL of supernatant was transferred into another tube containing 50 mg of primary and secondary amine (PSA), 50 mg of graphite carbon black (GCB) and 150 mg of magnesium sulphate. After proper agitation and centrifugation at 10000 rpm for 5 minutes, aliquots of the extract were evaporated through a nitrogen system and reconstituted with 1 mL of toluene for GC-MS analysis.

In addition, each pesticide standard solution (1 mg/mL) was prepared by diluting acetonitrile at a different concentration for standard curve preparation. A Shimadzu

Figure 1. MAP of the samples collected area of the Narsingdi district in Bangladesh

gas chromatograph (GC-MS QP 2010 Ultra, Japan) equipped with a mass selective detector and a Restek (Bellefonte, PA) Rxi-5MS analytical column with fused silica (30 m length × 0.25 mm inner diameter × 1.0 µm film thickness) were used for the analysis.

The temperature was programmed for organophosphate pesticide from an initial value of 90 °C, ramped to 180 °C at 25 °C/min and to 270 °C at 3 °C/min, then was increased to 300 °C at 20 °C for 3 min, with a total run time of 40 min. For analyte detection, the analysis was performed in selected ion monitoring (SIM) mode and the ions were considered for each pesticide.

Results

Method validation

The linearity of the calibration curves was evaluated at 50, 100 and 200 ng/mL. The coefficient of determination (R2) obtained was higher than 0.97 for all pesticides (Figure 2). The accuracy and precision parameters of the study method were obtained by recovery analysis throughout the entire procedure. The recovery rates were between 80 and 120%. Therefore, the present study method provided a good possibility to quantify pesticide residues in vegetables. The method validation for the analysis of dimethoate in brinjal and cucumber samples resulted in a Limit of Detection (LOD) of 19.66 ppb and a Limit of Quantification (LOQ) of 65.52 ppb. The dimethoate concentration in the brinjal samples (123.85 ppb) exceeded the LOQ, while the cucumber samples (12.93 ppb) remained below it, highlighting the sensitivity of the method for detecting and quantifying pesticide residues (Table 1).

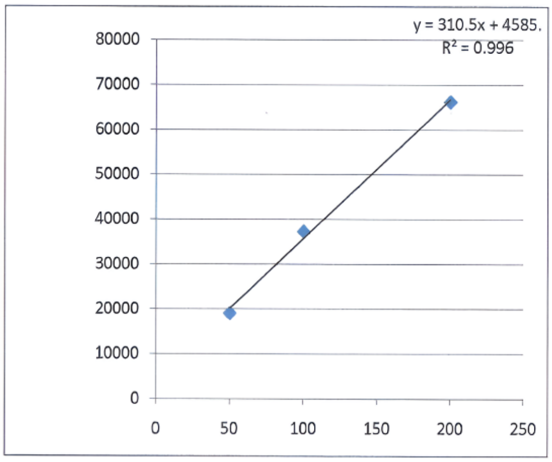


Figure 2. Linear regression analysis for determining LOD and LOQ values of dimethoate obtained from brinjal and cucumber samples.

Table 1. The LOD and LOQ values for dimethoate in brinjal and cucumber, along with calibration data and sample analysis

Dimethoate	BAR OP MR	Vegetables					
		MF	22-MAY	r2	lod	loq	range
Standard Conc. ppb	50	100	200	0.996334	19.65593	65.51976	50-200 ppb
Area	19025	37271	66150				
22 MAY	Area	Concentration (ppb)					
Brinjal	43045	123.8466					
Cucumber	8601	12.93064					

The overall scenario of pesticide residues in brinjals and cucumbers is shown in Figure 3. Among the 36 samples, 21 (58%) contained dimethoate, 14 (39%) of which exceeded the MRL set by the European Union database. Among the 36 samples, 18 (50%) contained chlorpyrifos, with only 3 (8%) samples exceeding the MRL, while 4 (11%) samples contained malathion, for which none (0%) exceeded the MRL. However, diazinon was not present at all in the samples of this study. Considering all the pesticides, 15 (42%) of the 36 samples exceeded the MRL for one or more pesticides, while 12 and 1 samples surpassed the MRL for dimethoate and chlorpyrifos, respectively, and 2 samples surpassed the MRL for both of these pesticides.

Pesticide residue at the farmer level and in retail market samples

Dimethoate residue in brinjal

Figure 4 shows that in the brinjal samples collected from different sources at the farmer level (field) and retail markets, all the raw samples exceeded the MRL recommended by the EU, with Fermer-1 (0.123 mg/kg), Retailer-2 (0.11 mg/kg) and Retailer-3 (0.109 mg/kg) samples containing residues 10-12 times higher than the recommended level. However, after cooking, all samples showed a more or less reduction in their residual levels, with some of them falling below the recommended level, while some of them were still above the level of MRL, such as the cooked samples of Farmer-1 (0.101 mg/kg).

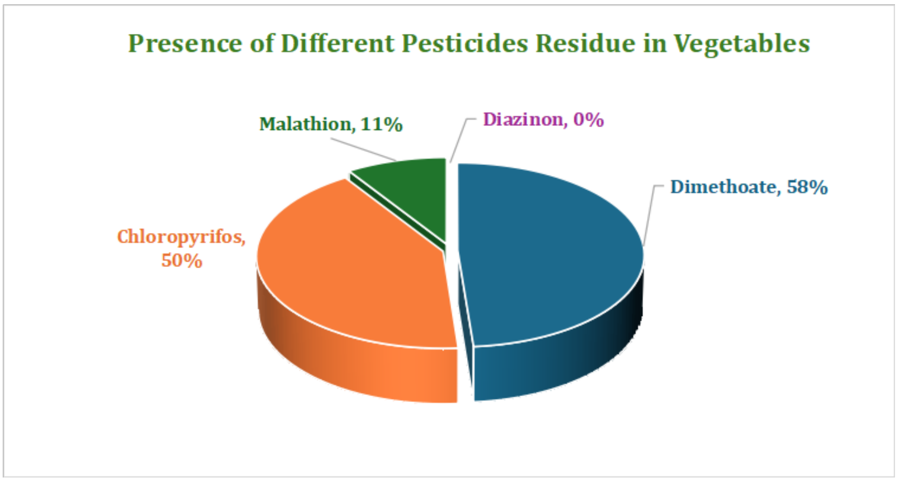
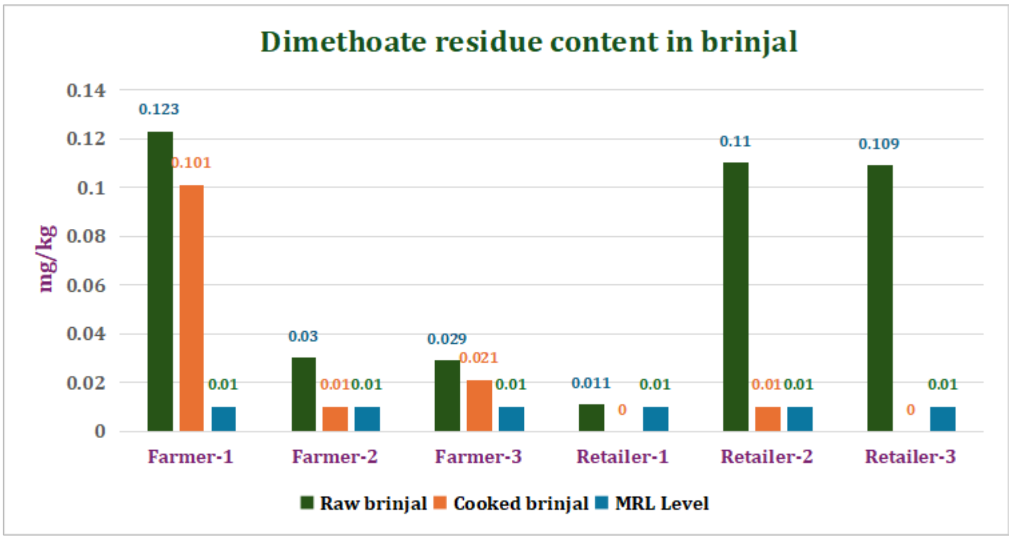


Figure 3. Different pesticides remaining as residues after the harvest of brinjal and cucumber in the Narsingdi district of Bangladesh



*MRL according to the EU pesticide database

Figure 4. Residual amount of dimethoate in raw and cooked brinjal samples collected from farmers and retail markets

Chlorpyrifos residue in brinjal

As for the chlorpyrifos residue level in brinjal, only one sample collected from Retailer-1 (0.02 mg/kg) exceeded the MRL by more than two times when it was raw, while none of the samples had a residue level higher than the MRL after being cooked. In addition, some samples were free of chlorpyrifos residues; these were the samples from Farmer-1 (both raw and cooked) and Retailer-3 (cooked) (Figure 5). Therefore, the contamination rate and the presence of pesticide residues caused by chlorpyrifos are markedly lower than those caused by dimethoate in brinjal samples for both raw and cooked conditions.

Dimethoate residue in cucumber

Figure 6 shows the dimethoate residues found in cucumber samples, with four out of the six raw samples exceeding the above MRL: Farmer-2 (0.014 mg/kg), Fermer-3 (0.011 mg/kg), Retailer-2 (0.019 mg/kg) and Retailer-3 (0.016 mg/kg). However, after cooking, most of the samples exhibited a reduction in residual levels, while the residual level in the Fermer-2 (0.012 mg/kg) and Retailer-3 (0.014 mg/kg) samples were still slightly above the MRL.

Chlorpyrifos residue in cucumber

The residual content of chlorpyrifos was comparatively less than the amount of dimethoate detected in cucumber under both the raw and cooked conditions, which showed a similar pattern to the residue content in brinjal. Among the samples, only one sample collected from Retailer-2 (0.013 and 0.011 mg/kg raw and cooked, respectively) slightly exceeded the MRL (Figure 7).

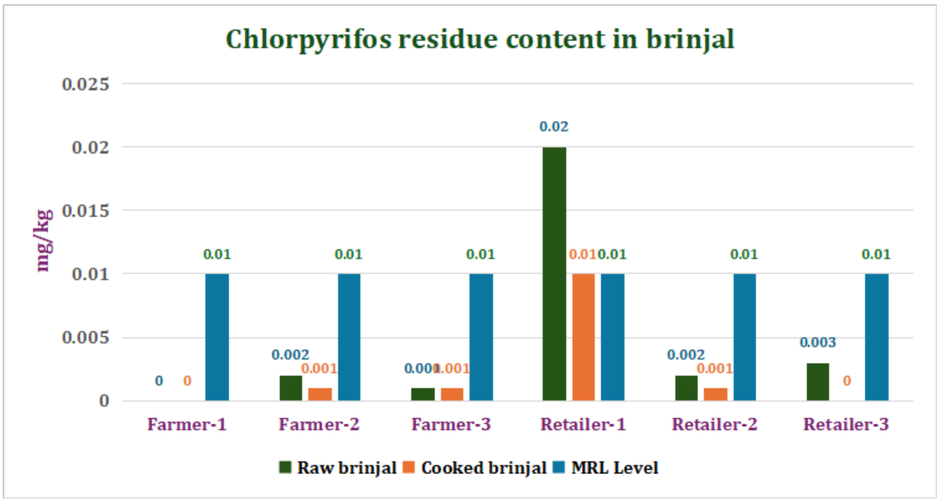


Figure 5. Residual amount of chlorpyrifos in raw and cooked brinjal samples collected from farmers and retail markets

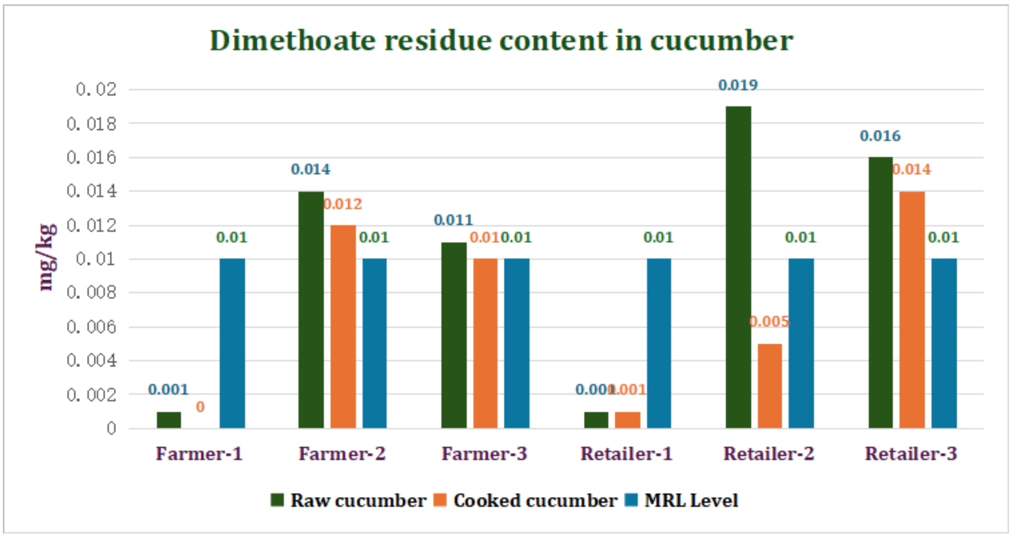


Figure 6. Residual amount of dimethoate in the raw and cooked cucumber samples collected from the farmers and retail markets

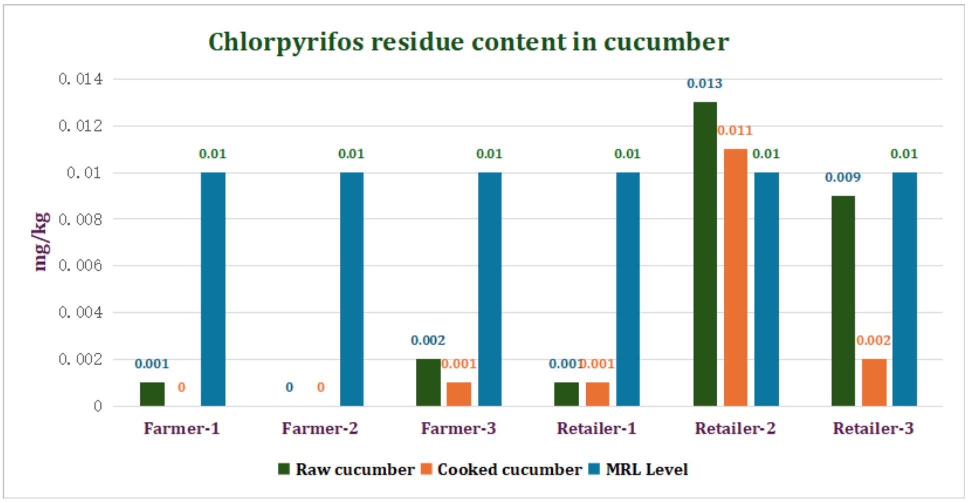


Figure 7. Residual amount of chlorpyrifos in raw and cooked cucumber samples collected from farmers and retail markets

Table 2. Residual amount of malathion in brinjal and cucumber samples collected from farmers and retail markets

Samples	Malathion residue content in				MRL level
	Raw brinjal	Cooked brinjal	Raw cucumber	Cooked cucumber	
Farmer-1	0.001	0	0.001	0	0.01
Farmer-2	0	0	0	0	0.01
Farmer-2	0	0	0	0	0.01
Retailer-1	0	0	0.001	0	0.01
Retailer-2	0	0	0.002	0	0.01
Retailer-3	0	0	0	0	0.01

Malathion and diazinon residues in brinjal and cucumber

In both the brinjal and cucumber samples, malathion was detected in only four (11%) samples, each two in the brinjal and cucumber samples. However, all samples contained an amount of residue of this pesticide that was below the recommended MRL (Table 2). In contrast, diazinon could not be found in any of the samples taken from any of the other sources, which showed that the vegetables in this area were free from diazinon residue contamination.

Pesticide residue in the export zone samples

In Narsingdi district, there are several areas in which vegetable cultivation is controlled and these areas are specially approved for export. Samples were taken from the controlled environment and the retail market of each area. No pesticide residue was found in the samples from the export zone.

Discussion

This study investigated the presence of pesticide residues in brinjal (eggplant) and cucumber samples collected from different sources in Bangladesh, including farmers' fields, retail markets and export zones. The findings revealed a concerning scenario of pesticide contamination, particularly with dimethoate (58%), followed by chlorpyrifos (50%), in these vegetables when they were raw (fresh). The prevalence of all OPs pesticides in the samples was more than a third (42%) above the MRL set by the European Union, with dimethoate solely representing the highest exceedance of the MRL at 39%. This indicates the widespread use and potential overuse of dimethoate in Bangladeshi agriculture. On the other hand, although cooking reduces the pesticide content in some samples, it is not always sufficient to reduce the pesticide content below the MRL. This highlights the limitations of cooking as the sole decontamination method. However, malathion residues were only present in 4 (11%) samples without exceeding the MRL, and unlike other studies, diazinon residues were not detected in any of the samples, indicating a possible shift in pesticide use practices. Encouragingly, no pesticide residues were found in the samples collected from the export zone, indicating that the stricter regulations in these controlled environments are being adhered to.

These results align with those of several previous studies [14-17] reporting similar patterns of pesticide contamination in vegetables from Bangladesh. The observed decrease in pesticide residues after washing and peeling, as also reported by [18-19], necessitates emphasizing proper prewashing practices for consumers.

The high prevalence of pesticide residues, especially above the MRL, highlights significant health risks for consumers. These findings underscore the urgent need for stricter enforcement of pesticide use regulations, particularly adherence to the dimethoate harvest interval (a minimum interval of 12 days between the harvest and the last spray, as recommended by the WHO). Additionally, promoting Integrated Pest Management (IPM) techniques can effectively reduce dependence on harmful chemicals. Educational campaigns should focus on empowering consumers to make informed choices, such as adopting safe prewashing practices and purchasing vegetables from reliable sources. Further investigations are also necessary to assess the long-term health impacts of chronic exposure to pesticide residues.

This study focused on a specific region in Bangladesh. A broader sampling across the country would provide a more comprehensive picture of the national scenario. Additionally, the study did not explore the reasons behind the high prevalence of dimethoate use.

Conclusion

These findings highlight the concern about OPs pesticide contamination in brinjal and cucumber plants in Narsingdi district of Bangladesh. Combined efforts are crucial for promoting responsible use of pesticides, raising consumer awareness and implementing stricter regulations to ensure the safety of the food supply.

Authors' contribution

T Mahjabin: Conceptualization, methodology, data curation, formal analysis, resources, validation, visualization, writing - original draft; MA Razzak: Project administration, supervision, investigation; conceptualization, funding acquisition, validation; M Rahman: Visualization, investigation, resources, validation; ANF Ahmmed: Supervision, investigation, resources, validation, writing - review and editing; R Ferdous: Data curation, formal analysis, software, validation, visualization; writing - original draft, writing - review and editing. All the authors reviewed the manuscript and provided their feedback.

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Conflict of interest

The authors declare that there are no conflicts of interest.

Compliance with ethical standards

The study was conducted while maintaining all possible ethical considerations. The farmers and sellers were informed before the sample was collected.

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