

Residue level and health risk assessment of organophosphorus pesticides in country bean and bitter gourd collected from Cumilla, Bangladesh

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Abstract

Organophosphorus pesticides are one of the most commonly used pesticide classes in agriculture in the management of insect pests due to their high efficacy. The extensive use of organophosphorus pesticides can contaminate both the atmosphere and food, which may lead to health problems. In this study, the residue level of organophosphorus pesticides, and their health risk assessment was evaluated on country bean and bitter gourd. Out of forty analysed samples of the country bean, two samples contained multiple residues, seven samples contained single residue. On the other hand, out of forty samples of bitter gourd, one sample contained multiple residues and three samples contained single residue. Among the organophosphorus pesticides, diazinon was the most frequently detected organophosphorus pesticide in country bean and bitter gourd. The highest short-term risks or acute risk (aHI) was below 1.83%, and the highest long-term risk or chronic risk (HQs) was below 10.46% for the country bean and bitter gourd, respectively. Short- and long-term risk assessment results showed that the intake risks of country bean and bitter gourd were acceptable. The health hazard results showed that country bean and bitter gourd consumption in Cumilla, Bangladesh does not pose an acute or chronic risk to human health. However, a routine monitoring system must be established to control the contamination of vegetables with pesticides.

1. Introduction

Organophosphorus pesticides (OPPs) are organic derivatives of phosphoric acid, phosphonic acid, phosphorothioic acid, or phosphonothioic acids (Corbett, 1974; Gupta, 2005; Espinoza *et al.*, 2017; Eto and Zweig, 2018). In 1937, Schrader and his colleagues found contact insecticidal activity in certain organophosphorus compounds (Ramulu, 2020). Still, organophosphorus pesticides are one of the most commonly used pesticide classes in agriculture in the management of insect pests due to their high efficacy, comparatively high dissipation and low persistence compared to other pest class pesticides (González *et al.*, 2012; Eto and Zweig, 2018). More often, pesticides are misused, including overdoses, adulterated formulas, and harvesting without a minimum waiting period (Herrera-Herrera *et al.*, 2019). The extensive use of pesticides can contaminate both the atmosphere and food, which may lead to health problems (Sultana and Nakagoshi, 2001;

Islam *et al.*, 2009). In the case of OPPs, extensive exposure to higher concentrations can lead to cardiovascular, nervous, respiratory, metabolic, reproductive and even immune symptoms problems (Bello-Ramirez *et al.*, 2000; Kwong, 2002; Joshi and Sharma, 2011). Besides, these groups of compounds are likely to change embryonic health and lead in children to ADHD (Hassani *et al.*, 2017). The toxicity of OPPs metabolites is much more toxic than the parent compound itself. Thus, OPPs are known to be the most toxic class of pesticides in insects, animals and humans (Hassani *et al.*, 2017). According to the USA pesticide usage database, the most frequently used OPPs are malathion, dimethoate, chlorpyrifos, acephate, naled, diazinon, methyl-parathion, dicrotophos, phorate, phosmet, and azinphos-methyl (Grube *et al.*, 2011). In the developing world, pesticide selection is mostly older, broad-spectrum, organophosphate and carbamate class, because of their acute toxicity and cheaper than the

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newer types (Sultana and Nakagoshi, 2001).

Bangladesh is a land of agriculture, and many farmers cultivate vegetables in their home garden and the field on a commercial basis (Chowdhury *et al.*, 2011). Bitter gourd and country bean are major vegetables that are grown extensively in some particular growing areas in Bangladesh like Cumilla, Bogra, Jessore and Narsingdi (Rahaman *et al.*, 2008; Rahaman *et al.*, 2016). In the context of Bangladesh, farmers have frequently used pesticides to ensure the quality and higher yields of vegetables (Dasgupta *et al.*, 2007; Hossain *et al.*, 2015). The most alarming concern is that Farmers of Bangladesh often spray hazardous pesticides like OPPs up to five to six times than the recommended dosage in one crop growing season (Sultana and Nakagoshi, 2001; Dasgupta *et al.*, 2007; Hossain *et al.*, 2015). In recent years, bitter gourd and country bean cultivation have become expensive and risky for farmers and consumers due to the increased use of chemical pesticides to control pests (Akram *et al.*, 2010). Also, in most areas of Bangladesh, vegetables are marketed without maintaining a pre-harvest interval period. These market vegetables often contain pesticide residues due to overuse in the farm, which has adverse effects on human health (Dasgupta *et al.*, 2007; Chowdhury *et al.*, 2011). A study found pesticide residues in bitter gourds next to brinjal, which is the reason for the vegetable export reduction (Quasem, 2003). In order to ensure consumer safety, most countries have established a maximum residue limit (MRL) and an acceptable daily intake (ADI) of pesticide residues in food items (Chowdhury *et al.*, 2011). The impact of daily intakes of pesticide residues in food should be measured by an exposure or risk assessment.

In the present study, seven mostly used organophosphorus pesticides (acephate, chlorpyrifos, diazinon, dimethoate, fenitrothion, malathion and quinalphos) were selected to determine the level of residues in country bean and bitter gourd marketed in Cumilla, Bangladesh. This study also aimed to assess the short-term and long long-term health risks to consumers posed by these pesticide residues on country bean and bitter gourd.

2. Materials and methods

2.1 Chemicals

The standard (>99.6% pure) of acephate, chlorpyrifos, diazinon, dimethoate, fenitrothion, malathion and quinalphos were purchased from Sigma-Aldrich (St Louis, MO, USA) via Bangladesh Scientific Pvt. Ltd. Dhaka, Bangladesh. Analytical grade methanol, acetone, HPLC grade acetonitrile, sodium chloride

(NaCl), anhydrous magnesium sulphate (MgSO₄) and Primary Secondary Amine (PSA) were purchased from Bangladesh Scientific Pvt. Ltd.

2.2 Study area

The study area included five major markets in the Cumilla district (Figure 1). It is a district of Bangladesh situated approximately 100 kilometres southeast of Dhaka. This study was conducted by collecting country beans and bitter gourds for pesticide residues from several Upazilla markets in Cumilla District. The central markets of Chandina, Debidwar and Daudkandi Upazilla and Kangshanagar Bazar under Burichang Upazilla and Maynamati Bazar under Cumilla Sadar Upazilla were considered for sample selection. These markets are famous for vegetables and vegetables are grown in various areas in the Cumilla district and certain parts of the neighbour areas.

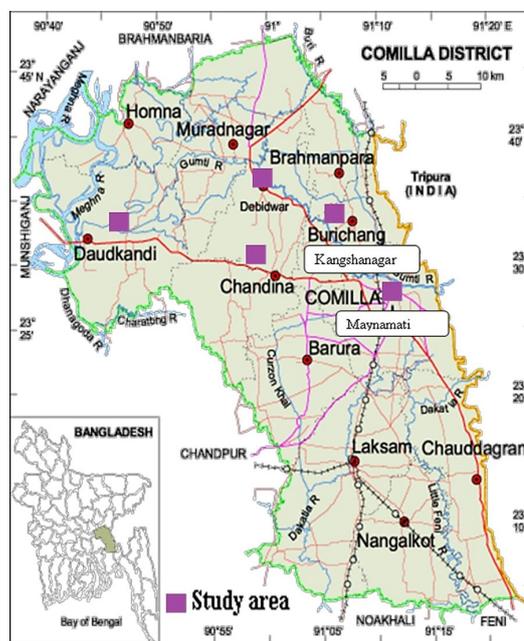


Figure 1. Map showing the places of sample collection in Cumilla district

2.3 Sample collection

For this research, 80 samples were collected (40 country beans and 40 bitter gourds). Eight country bean samples and eight bitter gourd samples were collected from each market (Chandina, Debidwar, Daudkandi, Kangshanagar and Maynamati) in the district of Cumilla. For the selected vegetables, each sample amounted to 1 kg. The samples were collected in individual polythene bags, and each bag has been appropriately labelled with the sample numbers and sources. The collected samples were transported on collection day to the Pesticide Analytical Laboratory of Bangladesh Agricultural Research Institute (BARI), Gazipur. Each sample was cut into small pieces and appropriately mixed for homogeneity.

Then, the chopped samples were processed at -20°C using airtight zipper bags before the extraction and clean-up process started.

2.4 Preparation of pesticide standard solution

Pesticide standard stock solutions of acephate, chlorpyrifos, diazinon, dimethoate, fenitrothion, malathion and quinalphos were prepared separately in acetone at a concentration of 1000 mg/L and stored at -20°C until use. A mixed standard solution of 50 mg/L in acetone containing all the pesticides as mentioned above was prepared by adding the appropriate volume of each stock solution in a 50 ml volumetric flask and made to the volume by addition of acetone. An intermediate mixed standard solution of 10 mg/L in acetone was prepared from the mixed standard solution of 50 mg/L. Then working standard solutions of 0.1, 0.2, 0.5, 1.0, 2.0, 3.0, and 5.0 mg/L in acetone were prepared by transferring the appropriate amount from 10 mg/L intermediate mixed standard solution into ten separate 10 mL volumetric flasks. All the standard solutions were kept in a freezer at -20°C until use.

2.5 Extraction and clean-up

In this study, the modified QuEChERS extraction technique was used for the extraction and clean-up of the collected vegetable samples (Prodhan *et al.*, 2015). The chopped vegetable samples were grounded thoroughly with a blender. A representative 10 g portion of the thoroughly homogenized sample was weighted in a 50 mL polypropylene centrifuge tube. Then 10 mL of acetonitrile (MeCN) was added into the centrifuge tube. The centrifuge tube was appropriately closed and shaken vigorously for 30 s by the use of a vortex mixer. Then, 4 g of anhydrous MgSO_4 and 1g of NaCl were added into the centrifuge tube, and it was shaken immediately by the vortex mixer for 1 min to prevent the formation of magnesium sulfate aggregates. Afterwards, the extract was centrifuged for 5 mins at 5000 rpm. An aliquot of 3 mL of the MeCN layer was transferred into a 15 mL centrifuge tube containing 600 mg anhydrous MgSO_4 and 120 mg Primary Secondary Amine (PSA). Then it was thoroughly mixed by vortex for 30 s and centrifuged for 5 mins at 4000 rpm (Laboratory Centrifuges, Sigma-3K30, Germany). After centrifuge, a 1 mL supernatant was filtered by a $0.2\ \mu\text{m}$ PTFE filter, and then it was taken in a clean GC vial for further analysis.

2.6 Instrumental analysis

The concentrated extracts were subjected to analysis by GC-2010 (Shimadzu) with Flame Thermionic Detector (FTD) for the detection of OPPs. The capillary column was AT-1 ($30\ \text{m} \times 0.25\ \text{mm} \times 0.25\ \mu\text{m}$) was used to separate the analytes. The split mode was used

for injection, and the injector and detector temperature were 250°C and 280°C with a split ratio: 30:0. The column temperature was programmed as follows: from 150°C for 1 min, from 150 to 220°C for 2 mins at $10^{\circ}\text{C}/\text{min}$. Helium was used as the carrier at a flow rate of 1.5 mL/min, and as a make-up gas at a flow rate of 30 mL/min for FTD with airflow at 145 mL/min. The flow rates of Helium and air were adjusted at 1.00 Pa, with a total run time was 10 mins.

2.7 Quality assurance procedure

The experiment was validated on parameters of specificity, linearity, determination coefficient (R^2) and limit of quantification (LOQ). The specificity was determined by specific retention time on the chromatogram for specific pesticides. The linearity and determination co-efficient was calculated continuously by standard fortified solutions of each pesticide at 0.05 to 0.5 mg/kg concentration levels. The limit of quantification was calculated as the lowest detection level for each pesticide. The limit of quantification (LOQ) was used to measure the sensitivity of the method.

2.8 Health risk assessment

The acute health risk to consumer (aHI) was measured using the estimated short-term intake (ESTI) and the acute reference dose (ARfD). The chronic health risk/hazard quotient (HQ) to consumers was measured using the estimated daily intake (EDI) and the acceptable daily intake (ADI). The hazard quotient indicates a potential risk if it reaches 100% and the higher aHI/HQ value is the higher risk. For the appropriate calculation, the ARfD and ADI values expressed as an mg/kg of daily intake for a 60 kg person for seven organophosphorus pesticides were obtained from the Joint FAO/WHO Meeting on Pesticide Residues (JMPR) database (<http://apps.who.int/pesticide-residues-jmpr-database>) (JMPR, 2004; JMPR, 2006). According to FAO/WHO, the average national per capita consumption of 23 g of leafy vegetables, 89 g of non-leafy vegetables and 14 g of fruit is an average of 126 g of fruit and vegetables per day in Bangladesh (FAO/WHO, 2003; Monoarul Haque *et al.*, 2014). The equation for calculation of ESTI, aHI, HQ and cHI were given below:

$\text{ESTI} = \text{the highest residue level} \times \text{food consumption}/\text{body weight}$

$\text{aHI} = \text{ESTI}/\text{ARfD} \times 100\%$

$\text{EDI} = \text{mean residue level} \times \text{food consumption}/\text{body weight}$

$\text{HQ} = \text{EDI}/\text{ADI} \times 100\%$

2.9 Statistical analysis

The level of pesticide residue in collected samples was analysed and calculated in mg/kg automatically by the Shimadzu GC software. All the data analysis and calculations were made by MS Excel 2013 software.

3. Results and discussion

3.1 Validation of the method

The linearity, determination coefficient (R^2), and LOQ were measured in external standard solution by using the peak areas obtained by GC-FTD analysis. The linear regression equation and determination coefficient R^2 values and LOQ were given in Table 1. The standard curve results showed that the linearity is excellent, with a determination coefficient of R^2 value higher than 0.9945. The limits of quantification (LOQs) were 0.01 mg/L, indicating the high sensitivity of this method. The proposed method was applied to the external standard solution for assessing its specificity. The specificity of seven selected organophosphorus pesticides was performed by comparing the retention times of each pesticide presented in a typical chromatogram obtained by GC-FTD using analysis of external added standard solution and matrix solvent (Figure 2).

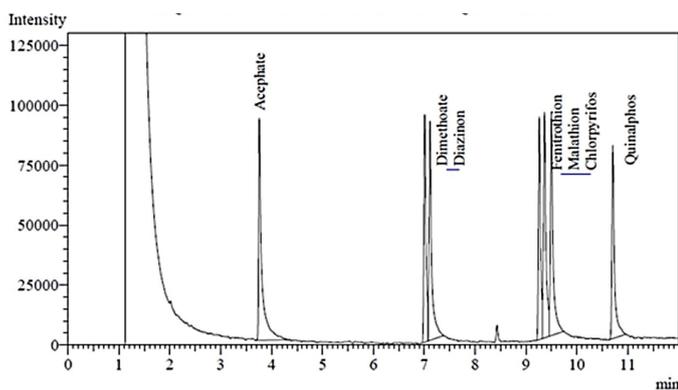


Figure 2. GC-FTD chromatogram of seven organophosphorus pesticides in external added standard spiked at 0.1 mg/kg

3.2 Pesticide residues in country bean

Out of forty samples of the country bean, nine samples (22.50% of the total number of samples)

contained diazinon residue and two samples (5% of the total number of samples) contained dimethoate residue (Table 2). In the country bean, the most frequently detected pesticide was diazinon (22.50%), followed by dimethoate (5%). Among the seven OPPs, acephate, chlorpyrifos, fenitrothion, malathion and quinalphos were not detected in any country bean samples were collected from five major vegetable markets of Cumilla district. These findings were very similar to Islam *et al.* (2014). They analysed 42 brinjals, cauliflower and country bean samples from the field and markets of the Narsingdi district where 15 samples were not detected any OPPs (Islam *et al.*, 2014). The study findings were well agreed with Hasan *et al.* (2017). In country bean samples collected from different markets of Dhaka, they have detected two kinds of insecticides (such as dimethoate and quinalphos). Out of 50 analysed country bean samples, ten samples (20%) contained dimethoate and quinalphos residues, 5 of which exceeded the maximum residue limits (MRLs). Most contaminated samples (8 samples) contained dimethoate residue (Hasan *et al.*, 2017). Ahmed *et al.*, (2017) analysed 170 samples collected from Jessore, Cumilla, Narsingdi, Tangail, Rangpur, Jamalpur, Gazipur and Dhaka for residue analysis in brinjal, yard long bean, bitter gourd, snake gourd, pointed gourd, okra, tomato, hyacinth bean and cabbage samples. Among the 170 samples, 21.78% were contaminated with insecticides, either single or multiple residues, in which 18.26% of samples had residues above MRL (Ahmed *et al.*, 2017).

According to Figure 3, diazinon was the most frequently detected pesticide followed by dimethoate in the country bean samples collected from 5 major vegetable markets (Chandina, Debidwar, Daudkandi, Kangshanagar and Maynamati) of Cumilla district of Bangladesh.

3.3 Pesticide residues in bitter gourd

Out of forty samples of bitter gourd, three samples (7.50% of the total number of samples) contained diazinon residue, one sample (2.50% of the total number of samples) contained multiple residues (Table 3) which

Table 1. Linear regression parameters and LOQ of the seven selected organophosphorus pesticides in the external added standard solution

Name of pesticide	Concentration (mg/kg)	Linear equation	R^2	LOQ (mg/kg)
Acephate	0.05-0.5	$y = 64654x + 294.93$	0.9995	0.01
Chlorpyrifos	0.05-0.5	$y = 61270x + 553.08$	0.9994	0.01
Diazinon	0.05-0.5	$y = 113063x + 882.86$	0.9945	0.01
Dimethoate	0.05-0.5	$y = 44867x + 716.55$	0.999	0.01
Fenitrothion	0.05-0.5	$y = 100414x + 977.86$	0.9975	0.01
Malathion	0.05-0.5	$y = 41036x + 340.41$	0.9945	0.01
Quinalphos	0.05-0.5	$y = 81217x + 211.14$	0.9993	0.01

R^2 = linear determination coefficient

Table 2. Summary of pesticide residue analysis in the country bean

Vegetable	Pesticide	No of the sample		Detected samples (%)	MRL* (mg/kg)	No of samples >MRL (mg/kg)	Residual Range (mg/kg)	Mean residue (mg/kg)
		Analysed	Detected					
Country bean	Acephate	40	0		0.01			
	Chlorpyrifos	40	0		0.01			
	Diazinon	40	9	22.50	0.01	9	0.079-0.138	0.1067
	Dimethoate	40	2	5	0.01	2	0.034-0.247	0.1405
	Fenitrothion	40	0		0.01			
	Malathion	40	0		0.02			
	Quinalphos	40	0		0.01			

*According to the EU Pesticide Database (EC-396/2005) (European Commission, 2005)

Table 3. Summary of pesticide residue analysis in bitter gourd

Vegetable	Pesticide	No of the sample		Detected samples (%)	MRL* (mg/kg)	No of samples >MRL (mg/kg)	Residual Range (mg/kg)	Mean residue (mg/kg)
		Analysed	Detected					
Bitter gourd	Acephate	40			0.01			
	Chlorpyrifos	40	1	2.50	0.01	1	0.056	0.056
	Diazinon	40	3	7.50	0.01	3	0.086-0.113	0.097
	Dimethoate	40	1	2.50	0.01	1	0.032	0.032
	Fenitrothion	40			0.01			
	Malathion	40			0.02			
	Quinalphos	40			0.01			

*According to the EU Pesticide Database (EC-396/2005) (European Commission, 2005)

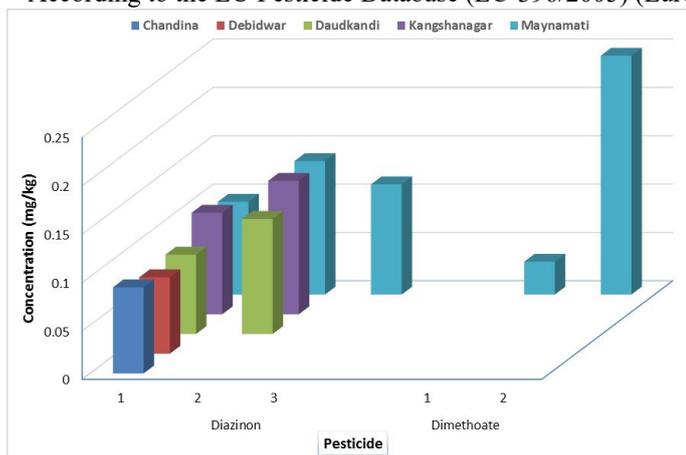


Figure 3. Frequency of organophosphorus pesticide residues with the amount in the country bean collected from different markets of Cumilla, Bangladesh

were above the European Union maximum residue limit (EU-MRLs). In the bitter gourd, the most frequently detected pesticide was diazinon (22.50%), followed by dimethoate (5%). Among the seven OPPs, acephate, fenitrothion, malathion and quinalphos were not detected in any bitter gourd samples collected from five major vegetable markets of Cumilla district. The findings of this study can also be compared to Akter *et al.* (2017). They found 22% of samples contained pesticide residues of diazinon, dimethoate, quinalphos, and chlorpyrifos residues in brinjal collected from the Mymensingh district. Among the detected OPP residues, only five samples exceeded the MRL set by EU (Aktar *et al.*, 2017). The present study is supported by Islam *et al.* (2019). They found that about 12% bitter gourd samples were contaminated with different organophosphorus

pesticides (Islam *et al.*, 2019).

Figure 4 shows diazinon was the most frequently detected pesticide followed by dimethoate and chlorpyrifos in bitter gourd samples collected from five major vegetable markets (Chandina, Debidwar, Daudkandi, Kangshanagar and Maynamati) of Cumilla district of Bangladesh. Out of 40 analysed bitter gourd samples only one bitter gourd sample collected from Kangshanagar vegetable market under Burichang Upazilla contained multiple residues of diazinon (0.093 mg/kg) and dimethoate (0.032 mg/kg). Another two bitter gourd samples collected from Debidwar and Daudkandi vegetables market contained a single residue of diazinon (0.113 and 0.086 mg/kg). Only one bitter gourd sample contained chlorpyrifos residue (0.056 mg/kg) collected from Maynamati Bazar under Cumilla

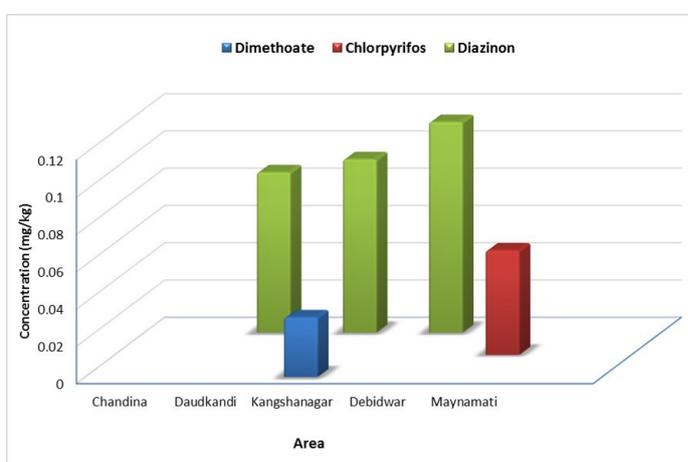


Figure 4. Frequency of organophosphorus pesticide residues with the amount in bitter gourd collected from different markets of Cumilla, Bangladesh

Sadar Upazilla. Nevertheless, other thirty-six bitter gourd samples (90% of the total number of samples) collected from these five major vegetable markets of Cumilla district contained no detectable residues of the sought pesticides.

3.4 Risk assessment

The mean concentrations of detected OPP pesticide residues were above their corresponding MRL levels (Table 2 and Table 3). The high-level residue in the country bean samples of some OPPs is an indicator of the recent use of diazinon and dimethoate on country bean farms. The OPPs health risk estimated in the country bean samples were represented in Table 4. For short-term risk analysis, all ESTI values were much lower than JMPR's ARfD values. Besides, the highest risk came from dimethoate with aHI w1.83%, followed by diazinon with aHI 0.68%. All the aHIs were lower than 100%, which indicates there was a negligible short-term or acute risk with the exposure to the tested pesticides via country bean consumption.

Moreover, in the long-term risk assessment, the risk quotient (HQs) was notably higher than acute risk indexes, which indicated the chronic risk from pesticide exposure via vegetable consumption should be considered. The long-term health risks (HQs) due to acephate, chlorpyrifos, diazinon, dimethoate, fenitrothion, malathion and quinalphos in the country bean was 0.025, 0.074, 3.174, 10.458, 0.148 and 0.025, respectively, suggesting that peoples in Cumilla have no significant health risk through the consumption of country bean since all the calculated values were less than 100%. Nevertheless, dimethoate presented the highest health risk index. In consequence, neither of the organophosphorus pesticide residues present in the country bean posed a risk for human health.

Table 4 and Table 5 contains the acute and chronic health risk values for acephate, chlorpyrifos, diazinon, dimethoate, fenitrothion, malathion and quinalphos in bitter gourd. For short-term risk assessment, all aHI values show much lower than 100%. That means, there was a negligible short-term or acute risk with the

Table 4. The short-term and long-term risks due to the average daily intake of organophosphorus pesticides through country bean consumption in Cumilla, Bangladesh.

Pesticide	Short-term risk			Long-term risk		
	ESTI (mg/kg/day)	ARfD (mg/kg/bw/day)	aHI (%)	EDI (mg/kg/day)	ADI (mg/kg/day)	HQ
Acephate	7.42E-06	0.1	0.007	7.42E-06	0.03	0.025
Chlorpyrifos	7.42E-06	0.1	0.007	7.42E-06	0.01	0.074
Diazinon	2.05E-04	0.03	0.682	1.59E-04	0.005	3.174
Dimethoate	3.66E-04	0.02	1.832	2.09E-04	0.002	10.46
Fenitrothion	7.42E-06	0.04	0.019	7.42E-06	0.005	0.148
Malathion	7.42E-06	0.3	0.002	7.42E-06	0.03	0.025
Quinalphos	/	/	/	/	/	/

ARfD and ADI were adopted from JMPR database. The symbol of “/” represented that there was no authorized value for ARfD/ADI, and the corresponding risk index could not be computed.

The country bean consumption value is based on their commended dietary intake per day for non-leafy vegetables of 89 g/day (data from FAO/WHO Dietary Guide (FAO/WHO, 2003).

When the detected residues in the sample were below the LOQ; for these values, data were treated with LOQ/2 (Yuan *et al.*, 2014).

Table 5. The short-term and long-term risks due to the average daily intake of organophosphorus pesticides through bitter gourd consumption in Cumilla, Bangladesh

Pesticide	Short-term risk			Long-term risk		
	ESTI (mg/kg/day)	ARfD (mg/kg/bw/day)	aHI (%)	EDI (mg/kg/day)	ADI (mg/kg/day)	HQ
Acephate	7.42E-06	0.1	0.007	7.42E-06	0.03	0.025
Chlorpyrifos	8.31E-05	0.1	0.083	8.31E-05	0.01	0.831
Diazinon	1.68E-04	0.03	0.559	1.44E-04	0.005	2.878
Dimethoate	4.75E-05	0.02	0.237	4.75E-05	0.002	2.373
Fenitrothion	7.42E-06	0.04	0.019	7.42E-06	0.005	0.148
Malathion	7.42E-06	0.3	0.002	7.42E-06	0.03	0.025
Quinalphos	/	/	/	/	/	/

ARfD and ADI were adopted from JMPR database. The symbol of “/” represented that there was no authorized value for ARfD/ADI, and the corresponding risk index could not be computed.

The country bean consumption value is based on their commended dietary intake per day for non-leafy vegetables of 89 g/day (data from FAO/WHO Dietary Guide (FAO/WHO, 2003).

When the detected residues in the sample were below the LOQ; for these values, data were treated with LOQ/2 (Yuan *et al.*, 2014).

exposure to the organophosphorus pesticides via bitter gourd consumption. However, in the long-term risk assessment, the risk indexes (HQs) were also much lower than 100%. So, the health risk from organophosphorus pesticide residues in bitter gourd was relatively smaller, which indirectly reflect the lower residue level of pesticides in bitter gourd. Health risk estimation revealed that dimethoate, diazinon and chlorpyrifos, though exceeded their MRLs in the country bean and bitter gourd, could not pose potential toxicity to the consumer. The acute and chronic risk index values showed that there was no health risk for consumers of Cumilla, Bangladesh, due to the intake of organophosphorus pesticide residues on these vegetables. Pesticide concentrations in vegetable products are known to be reduced by different home processing like washing, peeling, or cooking (Soliman, 2001; Wu *et al.*, 2007; Ling *et al.*, 2011; Yang *et al.*, 2012; Huan *et al.*, 2015). So, future studies should consider processing factors to compensate for reducing or removing pesticides.

4. Conclusion

This study found that country bean and bitter gourd contained organophosphorus pesticide residues in 16.25% of all samples collected from the major vegetable market of Cumilla district of Bangladesh. It was found that all detected samples exceeded the MRLs set by the EU. However, the short-term health risk or acute health indices (aHI) of those contaminated samples poses a negligible health risk for the consumer but chronic risk is considerably significant especially for dimethoate and diazinon. A modern traceability system can help to identify the source of contamination. Therefore, the observed levels of pesticide residues in the country bean and bitter gourd of Cumilla, Bangladesh do not pose a serious risk to consumers but pesticide contamination in vegetables may give rise to concern.

Conflict of interest

The authors declare no conflicts of interest.

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References

- Ahmed, M.S., Begum, A., Rahman, M.A., Akon, M.W. and Chowdhury, M.A.Z. (2017). Extent of Insecticide Residue Load in Vegetables Grown under Conventional Farming in Bangladesh. *The Agriculturists*, 14(2), 38–47. <https://doi.org/10.3329/agric.v14i2.31346>
- Akram, M.W., Rahman, M. and Ali, R. (2010). Evaluation of Some Management Practices for the Suppression of Cucurbit Fruit Fly in Bitter Gourd. *Journal of the Bangladesh Agricultural University*, 8 (1), 23–28. <https://doi.org/10.3329/jbau.v8i1.6393>.
- Aktar, M.A., Khatun, R. and Prodhon, M.D.H. (2017). Determination of Pesticide Residues in Eggplant Using Modified QuEChERS Extraction and Gas Chromatography. *International Journal of Agronomy and Agricultural Research*, 11(2), 22–31.
- Bello-Ramírez, A.M., Carreón-Garabito, B.Y. and Nava-Ocampo, A.A. (2000). A Theoretical Approach to the Mechanism of Biological Oxidation of Organophosphorus Pesticides. *Toxicology*, 149(2–3), 63–68. [https://doi.org/10.1016/S0300-483X\(00\)00222-5](https://doi.org/10.1016/S0300-483X(00)00222-5).
- Chowdhury, M.T.I., Razzaque, M.A. and Khan, M.S.I. (2011). Chlorinated Pesticide Residue Status in Tomato, Potato and Carrot. *Journal of Experimental Sciences*, 2(1), 1–5.
- Corbett, J.R. (1974). *The Biochemical Mode of Action of Pesticides*. London, United Kingdom: Academic Press.
- Dasgupta, S., Meisner, C. and Huq, M. (2007). A Pinch or a Pint? Evidence of Pesticide Overuse in Bangladesh. *Journal of Agricultural Economics*, 58 (1), 91–114. <https://doi.org/10.1111/j.1477-9552.2007.00083.x>.
- Espinoza-Navarro, O., Ponce-LaRosa, C. and Bustos-Obregón, E. (2017). Organophosphorous Pesticides: Their Effects on Biosentinel Species and Humans. Control and Application in Chile. *International Journal of Morphology*, 35(3), 1069–1074. <https://doi.org/10.4067/S0717-95022017000300041>.
- Eto, M. and Zweig, G. (2018). *Organophosphorus Pesticides*. 1st ed. Boca Raton, USA: CRC Press. <https://doi.org/10.1201/9781351075305>.
- European Commission. (2005). *Maximum Residue Levels of Pesticides in or on Food and Feed of Plant and Animal Origin*. EC-396/2005. Brussel, Belgium.
- FAO/WHO. (2003). *Diet, Nutrition and the Prevention of Chronic Diseases*. Report of a Joint FAO/WHO Expert Consultation. Geneva, Rome: FAO.
- González-Curbelo, M.A., Herrera-Herrera, A.V., Ravelo-Pérez, L.M. and Hernández-Borges, J. (2012).

- Sample-Preparation Methods for Pesticide-Residue Analysis in Cereals and Derivatives. *TrAC Trends in Analytical Chemistry*, 38, 32–51. <https://doi.org/10.1016/j.trac.2012.04.010>.
- Grube, A., Donaldson, D., Kiely, T. and Wu, L. (2011). Pesticides Industry Sales and Usage: 2006 and 2007 Market Estimates. Washington DC, USA: USEPA. https://www.epa.gov/sites/production/files/2015-10/documents/market_estimates2007.pdf.
- Gupta, R.C. (2005). Toxicology of Organophosphate and Carbamate Compounds. USA: Academic Press. <https://doi.org/10.1016/B978-0-12-088523-7.X5000-5>.
- Hasan, R., Prodan, M.D.H., Rahman, S.M., Khanom, R. and Ullah, A. (2017). Determination of Organophosphorus Insecticide Residues in Country Bean Collected from Different Markets of Dhaka. *Journal of Environmental and Analytical Toxicology*, 7(4), 489. <https://doi.org/10.4172/2161-0525.1000489>.
- Hassani, S., Momtaz, S., Vakhshiteh, F., Maghsoudi, A.S., Ganjali, M.R., Norouzi, P. and Abdollahi, M. (2017). Biosensors and Their Applications in Detection of Organophosphorus Pesticides in the Environment. *Archives of Toxicology*, 91(1), 109–130. <https://doi.org/10.1007/s00204-016-1875-8>.
- Herrera-Herrera, A.V., González-Sálamo, J., Socas-Rodríguez, B. and Hernández-Borges, J. (2019). Organophosphorus Pesticides (OPPs) in Bread and Flours. In Preedy, V.R. and Watson, R.R. (Eds.) Flour and Breads and their Fortification in Health and Disease Prevention. 2nd ed., p. 53-70. USA: Academic Press. <https://doi.org/10.1016/B978-0-12-814639-2.00005-8>.
- Hossain, M.S., Farkhrudin, A.N.M., Chowdhury, M.A.Z., Rahman, M.A. and Alam, M.K. (2015). Health Risk Assessment of Selected Pesticide Residues in Locally Produced Vegetables of Bangladesh. *International Food Research Journal*, 22(1), 110–115.
- Huan, Z., Xu, Z., Jiang, W., Chen, Z. and Luo, J. (2015). Effect of Chinese Traditional Cooking on Eight Pesticides Residue during Cowpea Processing. *Food Chemistry*, 170, 118–122. <https://doi.org/10.1016/j.foodchem.2014.08.052>.
- Islam, M.S., Prodan, M.D.H. and Uddin, M.K. (2019). Analysis of the Pesticide Residues in Bitter Gourd Using Modified QuEChERS Extraction Coupled with Gas Chromatography. *Asia Pacific Environmental and Occupational Health Journal*, 3 (5), 6–15.
- Islam, M.W., Dastogeer, K.M.G., Hamim, I., Prodan, M.D.H. and Ashrafuzzaman, M. (2014). Detection and Quantification of Pesticide Residues in Selected Vegetables of Bangladesh. *Journal of Phytopathology and pest Management*, 1(2), 17–30.
- Islam, S., Afrin, N., Hossain, M.S., Nahar, N., Mosihuzzaman, M. and Mamun, M.I.R. (2009). Analysis of Some Pesticide Residues in Cauliflower by High Performance Liquid Chromatography. *American Journal of Environmental Sciences*, 5(3), 325–329.
- JMPR. (2004). Pesticide Residues in Food 2004: Toxicological Evaluations. Rome, Italy: JMPR.
- JMPR. (2006). Pesticide Residues in Food 2006: Joint FAO/WHO Meeting on Pesticide Residues. Rome, Italy: JMPR
- Joshi, S.C. and Sharma, P. (2011). Male Reproductive Toxicity of Organophosphorous Compounds: A Review. *Toxicological and Environmental Chemistry*, 93(7), 1486–1507. <https://doi.org/10.1080/02772248.2011.581874>.
- Kwong, T.C. (2002). Organophosphate Pesticides: Biochemistry and Clinical Toxicology. *Therapeutic Drug Monitoring*, 24(1), 144–149. <https://doi.org/10.1097/00007691-200202000-00022>.
- Ling, Y., Wang, H., Yong, W., Zhang, F., Sun, L., Yang, M. L., Wu, Y.N. and Chu, X.G. (2011). The Effects of Washing and Cooking on Chlorpyrifos and Its Toxic Metabolites in Vegetables. *Food Control*, 22 (1), 54–58. <https://doi.org/10.1016/j.foodcont.2010.06.009>.
- Monoarul Haque, M., Amirul Hassan, M., Islam, K., Bhuiyan, M.R., Shahi, M.S.J.R. and Lipi, R.P. (2014). Diet Intake Pattern and Nutritional Status of Rural Population in Bangladesh. *Chattagram Maa-O-Shishu Hospital Medical College Journal*, 13(2), 51–54. <https://doi.org/10.3329/cmoshmcj.v13i2.21065>.
- Prodan, M.D.H., Papadakis, E.N. and Papadopoulou-Mourkidou, E. (2015). Determination of Multiple Pesticide Residues in Eggplant with Liquid Chromatography-Mass Spectrometry. *Food Analytical Methods*, 8(1), 229–235. <https://doi.org/10.1007/s12161-014-9898-3>.
- Quasem, M.A. (2003). Exports of Fresh Horticultural Crops from Bangladesh: Problems and Prospects. Dhaka, Bangladesh: Bangladesh Institute of Development Studies.
- Rahaman, M.A., Jahan, M., Islam, K.S. and Alam, S.N. (2016). Efficacy of three biopesticides against cucurbit fruit fly, *Bactrocera cucurbitae* Coquillett (Diptera: Tephritidae) and yield of bitter gourd. *Journal of Sylhet Agricultural University*, 3(2), 197–202.

- Rahaman, M.A., Prodhan, M.D.H. and Maula, M.A.K.M. (2008). Effect of botanical and synthetic pesticides in controlling epilachna. *International Journal of Sustainable Crop Products*, 3(5), 23–26.
- Ramulu, S. (2020). Chemistry of Insecticides and Fungicides. 3rd ed. India: Scientific Publishers.
- Soliman, K.M. (2001). Changes in Concentration of Pesticide Residues in Potatoes during Washing and Home Preparation. *Food and Chemical Toxicology*, 39(8), 887–891. [https://doi.org/10.1016/S0278-6915\(00\)00177-0](https://doi.org/10.1016/S0278-6915(00)00177-0).
- Sultana, P. and Nakagoshi, N. (2001). An Analysis of Pesticide Use for Rice Pest Management in Bangladesh. *Journal of International Development and Cooperation*, 8(1), 107–126.
- Wu, J., Luan, T., Lan, C., Hung Lo, T.W. and Chan, G.Y.S. (2007). Removal of Residual Pesticides on Vegetable Using Ozonated Water. *Food Control*, 18(5), 466–472. <https://doi.org/10.1016/j.foodcont.2005.12.011>.
- Yang, A., Park, J.H., Abd El-Aty, A.M., Choi, J.H., Oh, J.H., Do, J.A., Kwon, K., Shim, K.H., Choi, O.J. and Shim, J.H. (2012). Synergistic Effect of Washing and Cooking on the Removal of Multi-Classes of Pesticides from Various Food Samples. *Food Control*, 28(1), 99–105. <https://doi.org/10.1016/j.foodcont.2012.04.018>.
- Yuan, Y., Chen, C., Zheng, C., Wang, X., Yang, G., Wang, Q. and Zhang, Z. (2014). Residue of Chlorpyrifos and Cypermethrin in Vegetables and Probabilistic Exposure Assessment for Consumers in Zhejiang Province, China. *Food Control*, 36(1), 63–68. <https://doi.org/10.1016/j.foodcont.2013.08.008>.