

Article

Analysis of pesticide residue in vegetables collected from nine different regions of Bangladesh using Gas Chromatography

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Abstract: The study was conducted to analyze the amount of residue of six commonly used pesticides (e.g. chlorpyrifos, quinalphos, diazinon, acephate, dimethoate and fenitrothion) in vegetables viz., hyacinth bean and eggplant samples collected from local market of nine different locations viz. Cumilla, Bogura, Rangpur, Rajshahi, Khagrachari, Cox's bazaar, Barishal, Jamalpur and Dhaka for the comparison between the detected residue level with maximum residue limit (MRL) set by European Union. Among 36 analyzed samples of hyacinth bean six samples (16.67%) were contaminated with chlorpyrifos (0.082 mg/kg) and dimethoate residue (0.192 mg/kg - 0.961 mg/kg), all of them were above maximum residue limit. Out of 36 analyzed samples of eggplant 3 samples (8.33%) were contaminated with quinalphos (0.081mg/kg) and dimethoate residue (0.032 mg/kg - 0.217 mg/kg) which were above MRL. Most of the samples contained with dimethoate residue in both the vegetables. The presence of pesticide residue exceeding MRL in vegetables is harmful to human and other living organisms. Analyzed samples of Cumilla, Bogura, Rangpur and Rajshahi showed pesticide residue (i.e. dimethoate, chlorpyrifos and quinalphos) over MRL but the samples of other five locations like Cox's bazar, Khagrachari, Barishal, Jamalpur and Dhaka did not show any detectable residue which might safe for consumption.

Keywords: pesticide; residue; MRL; hyacinth bean; eggplant

1. Introduction

Bangladesh has an agricultural economy. Although food production has more than doubled since 1971, food security is still remains a major issue for it's huge population. To grow more agricultural production the use of agrochemicals is increasing rate at present. More intensive vegetable production is the common reliance of most vegetable producers on heavy application of pesticide (Hossain *et al.*, 2000). Among the vegetables i.e. hyacinth bean and eggplant grown in Bangladesh, are nutritious, valuable and very popular to consumers. It has been reported that in eggplant and hyacinth bean the attack of insect pests are severe and farmers sprayed pesticides quite frequently even every day (Anon. 2001). The improvement of the food production technology and the demands of the new markets for better products both in quality and quantity have been made among others with intensive uses of pesticides for productivity warranty (Trevisan *et al.*, 2004). Pesticide being toxic can become a potential hazard to the manufacturers, the user, the public at large and the environment. Pesticide can produce negative impacts, both social and private (Antle and Pingali, 1994). Extensive deliberate use of pesticides has resulted in contamination of our vital supplies, air, water and food. The risk to humans may be short term as well as long term depending on the persistence of the pesticide and the exposure period. Pesticide residue in food has become a consumer's safety issue and the consumers have the right to know how much pesticide get incorporated in the food they eat. The detection, identification and quantification of pesticide in the food becoming the public interest. But very little references are available on the presence of pesticides in vegetables in Bangladesh (Khatoon *et al.*, 2004; Ahmed *et al.*, 2013). Considering these, the present study was undertaken

to analyze and quantify the amount of residue of pesticide in hyacinth bean and eggplant collected from local market of nine different regions of Bangladesh.

2. Materials and Methods

The standard for chlorpyrifos, quinalphos, diazinon, acephate, dimethoate and fenitrothion were obtained from Sigma-Aldrich Laborchemikalien, Seelze, Germany via Bangladesh Scientific Pvt. Ltd. Dhaka, Bangladesh. Standards of all the pesticides contained >99.6% purity. Available vegetable samples viz., eggplant and hyacinth bean were collected from local market of different regions namely Cumilla, Bogura, Rangpur, Rajshahi, Khagrachari, Cox's bazaar, Barishal, Jamalpur and Dhaka.

2.1. Extraction, separation and clean up

The QuEChERS (Quick, Easy, Cheap, Effective, Rugged and Safe) method was used to extract pesticides from vegetables. The method was slightly modified. Collected vegetable samples were chopped on a chopping board with a sharp knife. 10 g of chopped sample was transferred into a 50 mL polypropylene centrifuge tube. Then 10 mL of acetonitrile (MeCN) was added into centrifuge tube. The centrifuge tube was closed properly and shaken vigorously for 30s by the use of a vortex mixture for proper mixing. Again 4 g of anhydrous MgSO₄ and 1g of NaCl (Sigma-Aldrich, Analytical grade) were added and vigorous shaking was done for 1 minute to prevent the formation of magnesium sulfate aggregates. Then centrifuge tube containing sample with chemical mixture was kept at centrifuge machine (Sigma-3K30, Germany) and centrifuged at 5000 rpm for 5 minutes. After centrifugation 3 mL of the supernatant (MeCN layer) was transferred into 15 mL micro centrifuge tube containing 120 mg Primary Secondary Amone (PSA), 600 mg anhydrous MgSO₄ and 100 mg of charcoal for cleaning up and thoroughly mixed by vortex for 30s. Then centrifuge tubes containing sample extract was placed in centrifuge machine and centrifuged at 4000 rpm for 5 minutes. After centrifuge, a 1 mL supernatant was filtered through a 0.2 µm PTFE filter using a syringe and taken in a clean vial for analysis in Gas Chromatography.

2.2. Detection and quantification of pesticide residue in samples

The concentrated extracts were analyzed by GC-2010 (Shimadzu) with Flame Thirionized Detector (FTD) for detection of diazinon, quinalphos, fenitrothion, acephate, chlorpyrifos and dimethoate. The capillary column used was AT-1, length 30m, ID 0.25mm and film thickness 0.25µm. Helium was used as carrier and make up gas in FTD. The instrument parameters for GC-FTD were as follows: Injection port temperature was 260⁰C, Column oven initial temperature was 160⁰C which went upto 240⁰C following 8 min incremental time. Detector temperature was 280⁰C, Stop time: 12 min, Current: 0.5 pA, Makeup flow: 30 ml/min, H₂ Flow: 1.5 mL/min and Air flow: 145 mL/min.

Before the injection of the sample extract, standard solutions of different concentrations of each pesticide were prepared and injected with selected instrument parameters. The samples were calibrated (retention time, peak area etc.) against three to four pointed calibration curve of standard solution of concerned pesticide. Each peak was characterized by its retention time. Sample results were expressed in mg/kg automatically by the GC software which represented the concentration of the final volume injected. From this value, the actual amount of pesticide residue present in the sample was determined by using the following formula:

Residue in sample (mg/kg)

$$= \frac{\text{Conc. obtained in injected volume (mg/kg)} \times \text{Quantity of final volume (L)}}{\text{Amount of sample taken (kg)}}$$

3. Results

The analytical results of both the vegetable samples for the detection of pesticide residue have been summarized in Tables 1 and 2.

Table 1. Quantity of residue of pesticide estimated form hyacinth bean collected form nine different regions of Bangladesh.

SL No.	Locations	Samples analyzed	Contaminated samples	Detected residue (mg/kg)	EU MRLs (mg/kg)
1	Cox's bazar	4	-	ND	-
2	Khagrachari	4	-	ND	-
3	Barishal	4	-	ND	-
4	Jamalpur	4	-	ND	-
5	Dhaka	4	-	ND	-
6	Cumilla	4	1	dimethoate: 0.192	0.01
7	Bogura	4	2	dimethoate:0.961	0.01
				clorpyriphos: 0.082	0.05
8	Rangpur	4	1	dimethoate: 0.921	0.01
9	Rajshahi	4	2	dimethoate: 0.632, 0.303	0.01

MRLs=Maximum residue limits; EU= European Union; ND=Not detected

Among 36 analyzed samples of hyacinth bean six samples (16.67%) were contaminated with chlorpyriphos and dimethoate residues. The level of chlorpyriphos residue was 0.082 mg/kg and the level of dimethoate residues were 0.192 mg/kg, 0.961 mg/kg, 0.921 mg/kg, 0.632 mg/kg and 0.303 mg/kg, respectively, all of them were above EU-MRLs. Dimethoate and clorpyriphos residue were found from four locations viz., Cumilla, Bogura, Rangpur and Rajshahi but the other five locations like Cox's bazar, Khagrachari, Barishal and Dhaka did not show any detectable residue.

Table 2. Quantity of residue of pesticide estimated form brinjal collected from nine different regions of Bangladesh.

SL No.	Locations	Samples analyzed	Contaminated samples	Detected residue (mg/kg)	EU MRLs (mg/kg)
1	Cox's bazar	4	-	ND	-
2	Khagrachari	4	-	ND	-
3	Barishal	4	-	ND	-
4	Jamalpur	4	-	ND	-
5	Dhaka	4	-	ND	-
6	Cumilla	4	-	ND	0.01
7	Bogura	4	1	dimethoate:0.032	0.01
8	Rangpur	4	1	quinalphos: 0.081	0.01
9	Rajshahi	4	1	dimethoate: 0.217	0.01

MRLs=Maximum residue limits; EU= European Union; ND=Not detected

Out of 36 analyzed samples of eggplant, 3 samples (8.33%) were contaminated with quinalphos (0.081mg/kg) and dimethoate residues (0.032mg/kg and 0.217mg/kg) which were above UE-MRLs. Dimethoate residue was found in Bogura and Rajshahi region. Rangpur had quinalphos residue and the rest regions as Cox's bazar, Khagrachari, Barishal, Jamalpur, Cumilla and Dhaka did not found any detectable residue.

4. Discussion

A total of 36 analyzed samples of hyacinth bean collected from local market of nine different locations viz. Jessore, Cumilla, Norsingdi, Bogura, khagrachari, cox's bazaar, Mymensingh, Rangpur, Dinajpur, Ishurdi, Gazipur and Dhaka revealed that 16.67% samples were contaminated with dimethoate and chlorpyriphos residue. All of the detected levels of residue were above maximum residue limits provided by European Union (2016). Most of the contaminated samples had dimethoate residue. Chlorpyriphos residue was about to double (0.082 mg/kg) of the Eu-MRLs in Bogura region. But the detected dimthoate residues were 30- 92 times higher than EU-MRLs. In case of 36 analyzed samples of eggplant, 8.33% samples were contaminated with quinalphos (0.081 mg/kg) and dimethoate residues (0.032 mg/kg and 0.217 mg/kg) which were also above EU-MRLs. Irrational and repeated use of pesticides in vegetables might cause the higher level of residues even at above MRL (Anon. 2001). Virginia and Bajet (1996) found residue of ogranophosphate insecticides (e.g. methomyl, triazophos, methyl parathion and diazinon) using rapid field kit from market basket samples of eggplant, tomato,

cabbage and chinese peachy that exceeded MRL. El-Saeid and Selim (2013) found residues of organophosphorus, organochlorine, pyrethroid and carbamate from market vegetables (e.g. beans, eggplant, cauliflower, tomato, pepper, carrot, cucumber, squash, potato, onions and okra) that showed above the MRL in 15.89% of the total tested samples. Ahmed *et al.* (2013) found 38.67% brinjal samples were contaminated with six insecticides (viz., acephate, fenitrothion, quinalphos, cypermethrin, diazinon and malathion), of which 17.33% samples had residue above MRL irrespective of single or multiple residues. Ahmed *et al.* (2014) reported that 34.67% samples of hyacinth bean were contaminated with four insecticides (i.e. acephate, quinalphos, cypermethrin and fenitrothion), in which 12% of the samples contained residue above MRL. The results of the present study agreed with the works of the above authors. Detected residue of dimethoate, chlorpyrifos and quinalphos were found in the samples of four locations viz., Cumilla, Bogura, Rangpur and Rajshahi but the samples of other five locations like Cox's bazar, Khagrachari, Barishal, Jamalpur and Dhaka did not show any detectable residue which might safe for consumption.

5. Conclusions

Analyzed 72 samples of hyacinth bean and eggplant, 12.5% of the total samples were found contaminated with dimethoate, chlorpyrifos and quinalphos residues. Among the nine contaminated samples, 10% samples had dimethoate residue. Only one sample from Bogura contained chlorpyrifos residue in hyacinth bean and one sample from Rangpur had quinalphos residue in eggplant. But the major concern was, all of the detected residues had above EU-MRLs. The presence of pesticide residue exceeding MRLs in vegetables are detrimental to human consumption. So, regular monitoring programme on pesticide residue study in vegetables is needed for safe food production.

Conflict of interest

None to declare.

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