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*Article*

# Multi insecticide residue analysis in vegetables collected from different regions of Bangladesh

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**Abstract:** An investigative study was conducted to determine the left over residue of five commonly used insecticides in cauliflower and yard long bean samples collected from local market of eight different regions like Bogra, Comilla, Dhaka, Gazipur, Jessore, Khagrachari, Rajshahi and Rangpur and carried out to the lab for analysis. Among 47 analyzed vegetable samples, 20 had insecticide residues, of them 11 had above maximum residue limit (MRL) set by FAO/WHO. Of 24 analyzed cauliflower samples, 11 contained residue of cypermethrin, diazinon, quinalphos, fenitrothion and malathion in which 8 had above MRL. Most of the sample contain diazinon and cypermethrin residue. Three samples showed 0.202-0.603 $mgkg^{-1}$diazinon resides and two samples contain 0.590-0.640 $mgkg^{-1}$cypermethrin residues which were above MRL. Cauliflower samples also showed two of three multi insecticide residues which were above MRL. Among 23 analyzed yard long bean samples, 9 showed the residue of five tested insecticides but only three had the residue of 0.410 $mgkg^{-1}$quinalphos, 0.651 $mgkg^{-1}$malathion and 0.591 $mgkg^{-1}$cypermethrin which were above MRL and the next 6 samples including three multi insecticide residues were of below MRL. Bogra, Jessore and Comilla showed more residues irrespective of single or multiple insecticide then other locations. No residue of the tested insecticide was found in the samples collected from Dhaka and Rajshahi in both the vegetables. Cauliflower in Khagrachari and yard long bean in Rangpur did not show any insecticide residue.

**Keywords:** insecticide; MRL; location; cauliflower; yard long bean

## 1. Introduction

Cauliflower and yard long bean are popular to consumers and extensively cultivated in Bangladesh. Both the vegetables are attacked by variety of insect pests. Many investigator suggested to use the chemical insecticides to protect the crops from the attack of the pest (Akhauri and Yadav, 2002; Prajapati *et al*., 2003; Chandrayudu *et al.*, 2006). Diversification into vegetable crops and increasing commercialization can support the development of the agriculture sector in several ways. But, in case of vegetables, data show the output growth in Bangladesh mainly through area expansion, without much improvement in yield. One potential drawback associated with a shift toward more intensive vegetable production is the common reliance of most vegetable producers on heavy application of pesticide (Hossain *et al.*, 2000). It has been reported that in cauliflower and yard long bean the attack of insect pests are severe and farmers sprayed insecticides quite frequently even every day (Kabir *et al.*, 1996; Anon., 2001). The improvement of the food production technology and mainly 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). Excessive deliberation of pesticide become toxic and hazard to the user, consumers and the environment which cause cancer, neurological problems, birth defect and fetal death (Sanborn *et al.*, 2007, Gilden *et al.*, 2010). 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. But very little references are available on the presence of pesticides in vegetables in Bangladesh (Khatoon *et al.*, 2004). The present study was undertaken to detect and quantify the amount of left over residue of insecticide in cauliflower and yard long bean from local market of eight different regions of Bangladesh.

## 2. Materials and Methods

The study was carried out in the pesticide analytical laboratory, Bangladesh Agricultural Research Institute, Joydebpur, Gazipur during 2009-2010 seasons. All the reagents and standard for cypermethrin, diazinon, quinalphos, fenitrothion and malathion were obtained from Sigma-Aldrich Laborchemikalien, Seelze, Germany via Bangladesh Scientific Pvt. ltd. Dhaka, Bangladesh. Standards of all the insecticide contained >99.6% purity. Cauliflower and yard long bean were collected from local market of eight different regions namely Bogra, Comilla, Dhaka, Jessore, Khagrachari, Rajshahi and Rangpur. The size of the sample of each vegetable was 1kg. Collected samples were kept in "Chilled box" and carried to the laboratory at the shortest time.

### 2.1. Extraction and separation

The collected vegetable samples (250g) were grounded thoroughly with the meat grinder. A sub sample of 20g was taken into a wide mouth jar then 100 ml of hexane was added to it. Anhydrous sodium sulphate ($Na_2SO_4$) was also added with sample until water was removed from the sample. The mixture was then macerated with high-speed homogenizer for 2 minutes. The homogenized material were then poured into 250 ml conical flask and placed into shaker for 12hrs continuous shaking. After shaking, the slurry was filtered through a Buchner funnel with suction. The flask and filter cakes were rinsed with 25ml of hexane each. The filtrate was then transferred into 250ml round bottom flask and was dried to 3-5ml by evaporation using a rotary vacuum evaporator. The concentrated filtrate was then transferred into volumetric flask making 10ml in volume which was then centrifuged at 16500 rpm for 5 minutes with laboratory refrigerator. After centrifuge, supernatant was collected for injection. Prior to the injection, this volume was again cleaned up by HPLC filter (0.2PTFE) which was ready for injection in Gas Chromatograph.

### 2.2. Detection and quantification of insecticide residue in samples

The concentrated extracts were analyzed by GC-2010 (Shimadzu) with Flame Thirmionic Detector (FTD) for detection of diazinon, quinalphos, fenitrothion and malathion. The capillary column AT-1, length 30m, ID 0.25mm and film thickness 0.25µm were used in case of FTD. Helium was used as carrier and make up gas in FTD. Electron captures detector (ECD), column Optima-1 and nitrogen gas were used for cypermethrin analysis. For GC-FTD, injection port temperature was $250^0$C, column oven initial temperature was $150^0$C which went up to $220^0$C following 10 min incrimental time. Detector temperature $280^0$C, stop time 10 min, current 1.0 pA, makeup flow 30 ml/min, hydrogen flow 1.5 mL/min and air flow 145 ml/min were also used for GC-FTD and in case of GC-ECD injection port température was $280^0$C, column oven initial temperature was $160^0$C which went up to $270^0$C following 10 min incremental time. Detector temperature was $300^0$C. Stop time was 18 min, current and makeup flow was same as GC-FTD.

Prior to the injection of the sample extract, standard solutions of different concentrations of all insecticides 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 insecticide. Each peak was characterized by its retention time. Sample results were expressed in $mgkg^{-1}$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:

$$\text{Residue in sample } (mgkg^{-1}) = \frac{\text{Conc. obtained in injected volume } (mgkg^{-1}) \text{ X Quantity of final volume (l)}}{\text{Amount of sample taken (kg)}}$$

## 3. Results

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

**Table 1. Quantity of residue of different insecticides estimated from cauliflower samples collected from different regions of Bangladesh.**

| Sample ID | | Quantity of detected residue ($mgkg^{-1}$) | | | | |
|---|---|---|---|---|---|---|
| **Location** | **Sample Code** | **Cypermethrin** | **Diazinon** | **Quinalphos** | **Fenitrothion** | **Malathion** |
| Bogra | BFC1 | ND | ND | ND | 0.350 | ND |
| | BFC2 | ND | 0.084 | 0.540 | ND | ND |
| | BMC3 | 0.640 | ND | ND | ND | ND |
| | BMC4 | ND | ND | 0.710 | ND | ND |
| Comilla | CFC1 | ND | ND | ND | ND | ND |
| | CFC2 | ND | 0.427 | ND | ND | ND |
| | CMC3 | ND | ND | ND | ND | ND |
| Dhaka | DMC1 | ND | ND | ND | ND | ND |
| | DMC2 | ND | ND | ND | ND | ND |
| Gazipur | CFC1 | 0.016 | ND | ND | ND | ND |
| | CFC2 | ND | 0.603 | ND | ND | ND |
| | CMC3 | ND | ND | ND | ND | ND |
| Jessore | JFC1 | 0.590 | ND | 0.029 | ND | 0.014 |
| | JFC2 | 0.379 | 0.054 | ND | ND | ND |
| | JMC3 | ND | ND | ND | ND | ND |
| | JMC4 | ND | ND | ND | ND | ND |
| Khagrachori | KMC1 | ND | ND | ND | ND | ND |
| | KMC2 | ND | ND | ND | ND | ND |
| Rajshahi | RMC1 | ND | ND | ND | ND | ND |
| | RMC2 | ND | ND | ND | ND | ND |
| Rangpur | RFC1 | 0.472 | ND | ND | ND | ND |
| | RFC2 | ND | 0.202 | ND | ND | ND |
| | RMC3 | ND | ND | ND | ND | ND |

In cauliflower, out of 23 analyzed samples 11 were contaminated with insecticides (i.e., cypermethrin, diazinon, quinalphos, fenitrothion, malathion). There were single insecticide residue in 8 samples and multi insecticide residues in 3 samples (Table 1). Diazinon was found at 5 locations (Comilla, Bogra, Gazipur, Jessore, Rangpur), cypermethrin was found at 4 locations (Bogra, Gazipur, Jessore, Rangpur), quinalphos was found at 2 locations (Bogra, Jessore), fenitrothion was found at Bogra and malathion was found at Jessore regions. Two samples of Jessore and one sample of Bogra showed multi insecticide residues, of which one sample of each location had residue which was above MRL. Four tested samples of Bogra, one contained multi insecticide residues (0.540 $mgkg^{-1}$quinalphos along with 0.084 $mgkg^{-1}$diazinon) and other three samples contained 0.350 $mgkg^{-1}$fenitrothion, 0.640 $mgkg^{-1}$cypermethrin and 0.710 $mgkg^{-1}$quinalphos residue which were above MRL. Among four samples of Jessore, two contained multi insecticide residues, of which one had 0.590 $mgkg^{-1}$cypermethrin residue along with quinalphos and malathion which was above MRL and other one had 0.379 $mgkg^{-1}$cypermethrin and 0.054 $mgkg^{-1}$diazinon residue which was below MRL and the next two had no residue. Among three samples of Rangpur, one had 0.202 $mgkg^{-1}$diazinon which was above MRL and one contained 0.472 $mgkg^{-1}$cypermethrin residue which was below MRL and another one had no residue. No residue of the tested insecticide was found in the samples collected from Rajshahi, Dhaka and Khagrachari (Table 1).

Nine yard long bean samples were found contaminated out of 23 analyzed samples from 8 different locations (viz., Bogra, Comilla, Dhaka, Gazipur, Jessore, Khagrachari, Rajshahi, Rangpur). There were 6 contaminated samples of single insecticide residue in which three samples were of cypermethrin. Only one sample of cypermethrin had 0.591 $mgkg^{-1}$residue from Jessore which was above MRL and the next two from Comilla and Jessore had below MRL. Two samples of Comilla contained insecticide residues. One had 0.651 $mgkg^{-1}$malathion which was above MRL and one had multi insecticide residues (cypermethrin with diazinon) which was below MRL. Two of three samples of Gazipur contained quinalphos residue (0.096 $mgkg^{-1}$ and 0.016 $mgkg^{-1}$) which were below MRL. Among four samples of Jessore one had multi insecticide residues (cypermethrin with quinalphos) which were below MRL and one had 0.591 $mgkg^{-1}$, cypermethrin residue which was above MRL and another two had no residue. One of three samples of Khagrachari contained 0.108 $mgkg^{-1}$cypermethrin and 0.019 $mgkg^{-1}$diazinon as multi insecticide residue which was below MRL. None of the cauliflower samples collected from Dhaka, Rajshahi and Rangpurregion was contaminated with tested insecticide (Table 2).

**Table 2. Quantity of residue of different insecticides estimated from yard long bean samples collected from different regions of Bangladesh.**

| Sample ID | | Quantity of detected residue(mgkg$^{-1}$) | | | | |
|---|---|---|---|---|---|---|
| **Location** | **Sample Code** | **Cypermethrin** | **Diazinon** | **Quinalphos** | **Fenitrothion** | **Malathion** |
| Bogra | BFY1 | ND | ND | ND | 0.0340 | ND |
| | BFY2 | ND | ND | 0.410 | | ND |
| | BMY3 | ND | ND | ND | ND | ND |
| Comilla | CFY1 | 0.432 | 0.039 | ND | ND | ND |
| | CFY2 | ND | ND | ND | ND | 0.651 |
| | CMY3 | ND | ND | ND | ND | ND |
| Dhaka | DMY1 | ND | ND | ND | ND | ND |
| | DMY2 | ND | ND | ND | ND | ND |
| Gazipur | CFY1 | ND | ND | 0.096 | ND | ND |
| | CFY2 | ND | ND | ND | ND | ND |
| | CMY3 | ND | ND | 0.016 | ND | ND |
| Jessore | JFY1 | ND | ND | ND | ND | ND |
| | JFY2 | 0.591 | ND | ND | ND | ND |
| | JMY3 | 0.190 | ND | 0.054 | ND | ND |
| | JMY4 | ND | ND | ND | ND | ND |
| Khagrachari | KMY1 | 0.108 | 0.019 | ND | ND | ND |
| | KMY2 | ND | ND | ND | ND | ND |
| | KMY3 | ND | ND | ND | ND | ND |
| Rajshahi | RMY1 | ND | ND | ND | ND | ND |
| | RMY2 | ND | ND | ND | ND | ND |
| Rangpur | RFY1 | ND | ND | ND | ND | ND |
| | RFY2 | ND | ND | ND | ND | ND |
| | RMY3 | ND | ND | ND | ND | ND |

## 4. Discussion

Five different organophosphorus and pyrethorid insecticide were analyzed to determine their residue levels in the collected cauliflower and yard bong bean samples from local market of eight different regions. Insecticide residues were detected in 43% of the 47 samples of cauliflower and yard bong bean and 23% of the total samples exceeded the MRL level provided by the food and agricultural organization (FAO) or world health organization (WHO). In cauliflower 46% samples were contaminated with cypermethrin, diazinon, quinalphos, fenitrothion and malathion residues in which 33% had above MRL. Diazinon was found in five samples showing 0.054-0.603 mgkg$^{-1}$residue, of which three had above MRL. Cypermethrin residue was also found in five samples containing 0.016-0.640 mgkg$^{-1}$residues, of which two were of above MRL. Fenitrothion residue was found only in one sample from Bogra which was above MRL containing 0.350 mgkg$^{-1}$residue and one sample from Jessore had 0.014 mgkg$^{-1}$residue of malathion which was below MRL. Quinalphos residue was found in three samples, of which two had 0.540 mgkg$^{-1}$and 0.710 mgkg$^{-1}$residues which were above MRL collected from Bogra and another one contained 0.029 mgkg$^{-1}$ residue which was below MRL collected from Jessore location. Among 23 analyzed yard bong bean samples 39% was contaminated with 5 insecticides irrespective of single or multiple residues in which 13% were above MRL set by European Commission. Only one samples of each location as Bogra, Jessore and Comilla had the residue of 0.410 mgkg$^{-1}$quinalphos, 0.591 mgkg$^{-1}$cypermethrin and 0.651 mgkg$^{-1}$malathion which was above MRL. The other detected reside levels contained 0.108-0.432 mgkg$^{-1}$cypermethrin, 0.019-0.039 mgkg$^{-1}$diazinon, 0.016-0.096 mgkg$^{-1}$quinalphos and 0.043 mgkg$^{-1}$fenitrothion which were below MRL. The sample of Bogra, Jessore and Comilla showed more insecticide residues. The detected levels of quinalphos, fenitrothion and malathion were found double of MRL set by FAO/WHO. Although multi insecticide residue was found in the samples of Comilla, Jessore and Khagrachari locations but their residue levels were below MRL. The presence of multiple insecticide residue indicated that farmers applied more than one insecticide on both the tested vegetables. The irrational use of insecticides on vegetables might cause the multiple residues of insecticides. Cauliflower had the higher residues of insecticides than yard long bean. Several investigators in India found organophosphate and pyrethroid residues in farmgate vegetables (Deka *et al.*, 2005; Battu Joia, 2006). Rahman *et al.* (2013) reported 26% samples (i.e., cauliflower, bean, brinjal, cabbage and red amaranth) were contaminated with quinalphos, chlorpyriphos and dimethoate residues which were above MRL. EL-Saeidand Selim (2013) found residues of organophosphorus, organochlorine, pyrethroid and carbamate in 15.89% of market vegetables (viz., cauliflower,

beans, egg plant, tomato, pepper, carrot, cucumber, squash, potato, onions and okra) that showed above the MRL. Ahmed *et al.* (2014) revealed that 34.67% hyacinth bean sample was contaminated with cypermethrin, quinalphos, fenitrothion and acephate residues, of which 12% had residue above MRL. The result of the present study was more or less similar with the findings of the above authors. This outcome is undesirable for safe food but it occurred in recent past years because of the abuse of insecticide on vegetables.

## 4. Conclusions

The detected residue levels of quinalphos, diazinon, cypermethrin and fenitrothion were more in cauliflower as compared to malathion, quinalphos, fenitrothion and diazinon levels were found triple times higher than MRL in cauliflower. In case of yard long bean the levels of quinalphos, fenitrothion and malathion were found double of the MRL. The presence of quinalphos, cypermethrin, fenitrothion, malathion and diazinon residues in cauliflower and yard long bean even at or above MRL indicated that farmers might be used insecticide irrationally and they did not maintain pre harvest interval of the applied insecticides. So, the government should take initiative for safe use of pesticide in vegetables. The samples of vegetables need to be assessed on a routine basis because a number of cauliflower and yard long bean samples were found contaminated with insecticide residue at levels that exceeded the maximum residue limit required for safe and quality food.

## Conflict of interest

None to declare.

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