ANALYSIS OF PESTICIDES RESIDUES IN FRUITS AND VEGETABLES EXPORTED TO THE EUROPEAN UNION

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ABSTRACT

Presence of pesticide residues in agricultural commodities can cause acute and chronic toxicities in human and thus, exceeding their Maximum Residue Limits (MRL) in fresh fruits and vegetables can negatively affect the export market. The purpose of this study was to develop a monitoring programme to evaluate and regulate the pesticide usage of farmers to produce safe products for human consumption while protecting the export market. Two surveys were conducted to identify the practices of the farmers and exporters who cultivate and process the commodities respectively for exportation to the European Union market. A total of 340 samples of fresh fruits and vegetables were analyzed for the presence of pesticide residues. According to the results, 26% of tested samples showed the presence of pesticide residues. Among them 65% samples exceeded the Maximum Residue Limit of the European Union (EU-MRL). Number of pesticide residue forms identified in samples was 24 out of which 50% them are unauthorized in the European Union. Contamination of leafy vegetables was relatively higher than that of the other food categories analyzed in this study. The most detected pesticide residues are Profenophos and Tebuconazole.

Key words: Contamination, Leafy vegetables and fruits, Maximum Residue Limit, Pesticide

INTRODUCTION

The maximum allowable levels of pesticide residues in foods are often stipulated by regulatory bodies in many countries. The European Union (EU) decides Maximum Residue Limits (MRLs) for all food and animal feed and they have stipulated the lowest MRL in comparison to other countries and regions (Lakshani *et al*., 2017 European Commission, 2005). Out of nearly 104 active ingredients of pesticides registered in Sri Lanka, 22 active ingredients are banned in EU. Over use of pesticides and use of unauthorized pesticides have resulted in reduction of International trade with EU member countries (European Commission 2002) and the same also affects consumer health in Sri Lanka (Marasinghe *et al*., 2011).

In order to minimize pesticide residues in food crops, Integrated Pest Management (IPM) and Good Agricultural Practices (GAP) could be utilized in agriculture production. Good Agricultural Practices in Sri Lanka (SLGAP) is being implemented by the Department of Agriculture (DOA). A total of 60 Sri Lankan fruits and vegetables farmers have been certified under the SLGAP program up to June, 2017.

EU being one of the highest food safety standards implemented union in the world ensure that the food available for consumers are safe. The Rapid Alert System for Food and Feed (RASFF) in EU detects the risks to public health associated with food and feed and notify relevant authorities regarding the unsafe aspects. As such during the period from 2014 to 2016 nine notifications have been received by the Department of Agriculture.

Five of the above notifications were raised for leafy vegetables (Gotukola) followed two notifications for long beans. Papaya, Passion fruit, Chili and Pepper received only one notification each. In consequent to the increasing notifications, the European Commission made an audit on "Control of Pesticides in Food of Plant Origin intended for Export to the European Union" in 2016 (Director General for Health and Food Safety, 2016).

Therefore, quantification of pesticide residues in fruits and vegetables which are to be exported was conducted in this study to monitor and regulate the use of pesticides.

MATERIALS AND METHODS

Survey (1) was conducted to identify the practices at farmer fields and survey (2) was conducted to identify the practices at pack houses of exporters. All farms and pack houses identified for the study were the suppliers of fruits and vegetables to the European member countries. Two separate questionnaires were prepared for the two surveys. Pack houses and farmer fields were visited randomly and personally interviewed with farmers as well as pack house authorities. List of farmer fields and exporter facilities were gathered by the Extension & Training Centre of the Department of Agriculture. The samples were collected from August 2016 to December 2017, while surveys were conducted in mid-2017.

Survey 1: Farmer level

Data were collected from randomly selected fourteen (14) farmers who export vegetables to EU consisting of nine (9) Bitter Gourd and five (5) Snake Gourd farms. All the 14 farms were located in Kalutara, Gampaha and Puttalam districts. Correct use of pesticides (using correct dosage and recommending Pre-harvesting intervals) and the adoption of IPM practices were considered at this survey.

Survey 2: Exporter level

Second survey was conducted to identify the practices at pack houses which are obtaining fruits and vegetables from registered farmer fields. Random analysis of pesticide residues and traceability at the export items were also observed. Data were collected from the twelve (12) pack houses located in Wattala, Kandana, Puttalam, Maharagama, Nugegoda, Angoda, Seeduwa, Katunayake and Negombo.

Sampling and quantification of pesticide residues

Three hundred and forty (340) samples of fruits and vegetables were randomly collected from farmer fields and pack houses of fruits and vegetable exporters and from the fruits and vegetable consignments ready to be exported to the European countries, at the National Plant Quarantine Services (NPQS). The sample collection at the consignments was done through the National Plant Quarantine service of Department of Agriculture. Sampling was done according to the composite methods of sampling as per the recommendations of the Codex Alimentarius Commission (FAO/WHO, 1999). Samples

were collected in polyethylene bags and stored in cool boxes. Different agricultural commodities were collected (vegetables, fruits, and leafy vegetables) and transferred to the analytical laboratories of Industrial Technology Institute (ITI) and Bureau Veritas Consumer Product Sri Lanka (Pvt.) Ltd, (BV) for pesticide residue analysis. While Bureau Veritas Consumer Product Sri Lanka (Pvt.) Ltd. (BV) is certified with ISO/IEC 17025 for analysis of pesticides in fruits and vegetables and use Gas chromatography – Mass Spectrometry as the analytical equipment, ITI has already fulfill the requirements for obtaining accreditations and use Liquid Chromatography- Mass Spectrometry for the same.

RESULTS AND DISCUSSION

Results of the Survey 1 and Survey 2 are tabulated in Table 1 and Table 2, respectively.

Table 1. Farmers practices of the selected farmers on the use of agro-chemicals

Farmer category	No. of famers met	Insects (No. of cases)	Insecticides used	Fungicides used	Weedicides used	Follow recommendations	Follow Pre-harvest Interval (PHI)	IPM
Snake guard	5	Melon fly (03) Aulacophora, Caterpillar	Chlorantraniliprole (03) Acephate	(Tryfloxystrobin +tebuconazole) Captan Chlorothalonil	NA	Yes	Yes	Bagging, Protein bait, Pheromone trap
Bitter guard	9	Aphids(02) Leaf miner Melon fly(08)	Chlorantraniliprole (05) Abamectin (03) thiamethoxam (02)	(Tryfloxystrobin +tebuconazole) (02) Manncozeb	NA	Yes	Yes	Bagging, Protein bait, Pheromone trap

Table 02. Results of the Survey (2) – Exporter level

No. of exporters visited for the survey	Fruits & vegetables obtained only from registered fields	Pesticide residues tested before exportation	Presence of traceability	Chemicals used for storage	Chemicals used for washing
12	01(8%)	01(8%)	02(17%)	None	None

Pest distribution in snake-gourd and bitter-gourd fields

Figure 1 shows that, melon fly, leaf miner, caterpillars, aphids and aulacophora beetle were the main pests of snake gourd and bitter gourd in the farmer fields who cultivate fruits and vegetables for EU market. Melon fly (*Bactrocera cucurbitae*) is a serious pest of horticultural crops, especially cucurbits.

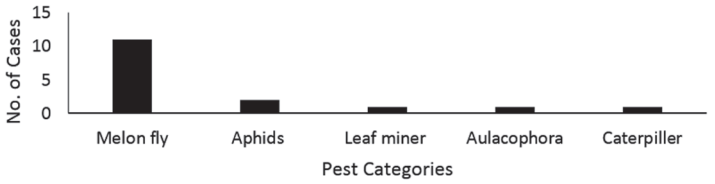

Figure 1. Pest distribution in snake gourd and bitter gourd fields

Pesticide recommendations for pest/ crop combinations, correct dosages and Pre-Harvest Interval (PHI):

Figure 2 shows that Chlorantraniliprole has been predominantly used over the other insecticides by farmers. Since having three days of pre-harvest interval, there are no effects of remaining residues on treated surfaces and also for consumers and environment. All active ingredients used by the farmers are authorized in both Sri Lanka and EU and also recommended for common pest management.

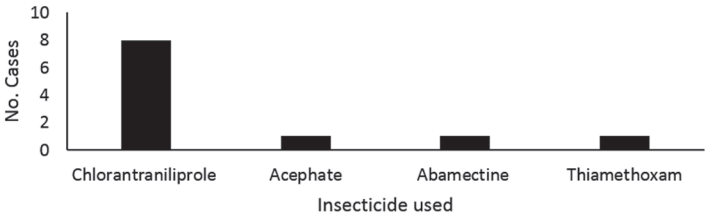

Figure 2. Field usage of insecticides for snake gourd and bitter gourd

Usage of fungicides in farmer fields of snake gourd and bitter gourd are given in Figure 3. A pre-mixture of Tryfloxystrobin Tebuconazole is recommended for powderly mildew in cucurbits. This fungicide has been used in farmer fields of snake gourd and bitter gourd. Other fungicides such as Captan, Chlorothalonil and Mancozeb are also recommended by the DOA for cucurbits and are authorized in the EU. Hence results reveal that farmers are abide by the DOA recommendations for the pest management.

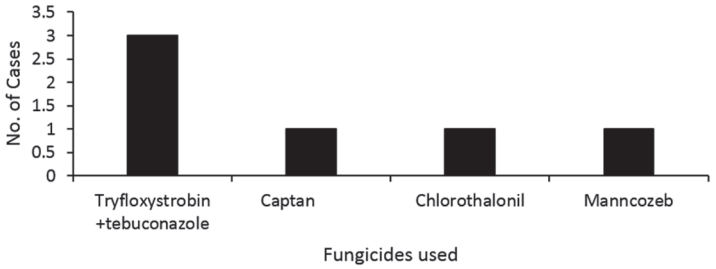

Figure 3. Usage of fungicides in snake gourd and bitter gourd fields

Fruits and vegetables obtained from registered farmer fields

Since monitoring program for snake gourd and bitter gourd of which are cultivated for exportation is already being functioned and simultaneously exporters were registered under Department of Agriculture including their pack-houses and farmer fields, all exporters were obtaining their snake gourd and bitter gourd consignments from supervised farmer fields. However, fruits and vegetables other than snake guard and bitter gourd has not been obtained only from the registered farmer fields. Eight percent (8%) of pack houses had obtained their fruits and vegetables only from registered farmer fields while the other 92% has obtained bulk of fruits and vegetables from both local markets and unregistered farmer fields.

Pesticide residue testing before exportation

Only 8% processors/exporters were found to conduct pesticide residue tests before exporting to the EU market while the remaining 92% had exported their consignments without any pesticide residue testing (Table 2). However, test reports of residue analysis confirmed that there were no harmful residues of pesticides in consignments; giving an opportunity to maintain the stability in EU market. The major limiting factor of the residue testing before exportation is the high cost of analysis of pesticide residues in Sri Lanka. Hence, an establishment of a laboratory set up for analysis of pesticide residues at an affordable cost would be an opportunity in promoting exports.

Traceability of pack-houses

Results revealed that, 17% of pack houses had possessed the traceability reports while other 83% of pack houses has not either maintained and/or were not able to trace back their commodities.

Identification and quantification of pesticide residues

Table 3 shows that 28% of samples of vegetables, collected from pack-houses and farmer fields intended for export to EU were contaminated (≥0.01 mg/l) with one or more pesticides of current scope. Further, it shows that pesticide contamination of leafy vegetables (Gottukola) in 2017 is much lower than that in 2016.

Tables 3. Pesticide residues detected in vegetables sampled from pack-houses and famer fields

Vegetable/ Fruit	Number of samples analyzed	Number of samples contaminated	Pesticide residue levels detected mg/kg
Angunakola	2	2	Acephate-0.02 Etofenprox-0.011
Beans	15	6	Etofenprox-0.013 – 0.28 Carbendazim-0.02 Diazinon - 34.72 Thiamethoxam – 0.05 Fipronil – 17.6
Bell pepper	1	1	Carbendazim-0.018
Bitter gourd	33	1	Profenophos – 0.01 Imidacloprid – 0.01
Brinjal (Eggplant)	8	1	Carbendazim-0.012
Capsicum	2	2	Acephate-0.03 Acetamiprid-0.07
Chilli	5	2	Diazinon – 0.01 Quinalphos 0.01
Cucumber	3	2	Thiamethoxam-0.013– 0.066
Gotukola (2016)	23	15	Profenophos- 0.11 – 69.61 Tebuconazole 0.02 – 1.2 Hexaconazole 0.06 Thiamethoxam 0.3 – 0.6 diazinon 0.07
Gotukola (2017)	8	6	Profenophos-0.031-2.54 Azoxystrobin-0.011 Etofenprox-0.02 Carbendazim-0.09, Thiophanate-methyl-0.012,
Kankun	11	02	Profenophos-0.02 Thiamethoxam-1.72 Dimethoate-0.34

Tables 3(continued). Pesticide residues detected in vegetables sampled from pack-houses and famer fields

Vegetable/ Fruit	Number of samples analyzed	Number of samples contaminated	Pesticide residue levels detected mg/kg
Kathurumurunga	12	01	Profenophos-1.13
Long Bean	6	2	Thiophanate-methyl-0.12 Edifenphos-0.02, Profenophos-2.87
Mukunuwenna	15	10	Thiamethoxam 0.04-115.1 Profenophos 0.08-1.34 Tebuconazole 0.02-0.03 Quinalphos-0.02 Imidacloprid-13.93
Okra	19	02	Imidacloprid-0.16 Thiamethoxam-0.01
Onion/Onion flowers	3	1	Thiodicarb-0.03
Snake guard	16	03	Imidacloprid-0.005 Thiamethoxam-0.02-0.17
Spinach	12	2	Profenophos-0.03 Thiamethoxam-0.78
Thalanabatu	3	1	Etofenprox-0.01 Profenophos-0.05
Winged Bean	9	ND	-
Other (Ash plantain/ Banana blossom (Keselmuwa)/ Drumstick (Murunga)/ Drumstick leaves/ Raja-ala/ Okra/ Tomato	19	ND	-
Total	225	62 (28%)	

ND < Limit of Detection(LOD); LOD -0.01mg/kg

Angunakola (*Wattakaka volubilis*), Gotukola (Centella asiatica), Kankun (*Ipomoea aquatica*), Kathurumurunga (*Sesbania grandiflora*), Mukunuwenna (*Alternanthera sessilis*), Thalanabatu (*Solanumm elongena*), Ash plantain (*Musa* spp.), Thibbatu (*Solanum torvum*), Kohila (*Lasia spinosa*)

Table 4 shows that 20% of samples of fruits, collected from pack-houses and farmer fields were also contaminated (≥0.01 mg/l) with pesticide residues.

Tables 4. Pesticide residues detected in fruits sampled from pack-houses and famer fields

Vegetable/ Fruit	No of samples analyzed	No of samples contaminated with pesticide residues	Range of pesticide residues detected- mg/kg
Banana	2	1	Profenophos-0.03
Guava	03	ND	
Mango	05	ND	
Papaya	8	3	Thiophanate-methyl-0.06 Captan-0.04 Dimethoate- 0.03
Wood-apple/ Pineapple	2	ND	-
Total	20	4 (20%)	

Table 5 shows that 25% of samples of fruits and vegetables, collected from consignments at the NPQS were contaminated with one or more pesticides at or above 0.01 mg/kg.

Table 5. Pesticide residues detected in fruits and vegetables sampled from consignments at National Plant Quarantine Service

Vegetable/ Fruit	No of samples analyzed	No of samples contaminated with pesticide residues	Range of pesticide residues detected
Beans	2	ND	
Long bean	4	1	Novaluron - 0.08
Winged bean	2	ND	
Snake gourd	8	3	Imidacloprid (0.01-0.06) Dimethoate (0. 34)
Okra	15	ND	
Mukunuwcnna	10	7	Profenophos-(0.02-0.05) thiamethoxam (0.22), Imidacloprid (0.01)
Gotukola	14	10	Phenthoate (0.02), Profenophos (0.03-5.38) Fipronil (0.01), Tebuconazole (0.02- 1.08) Quinalphos (0.04), Chlorpyriphos (0.02) Malathion (0.01-0.04), Metribuzin (0.16) Thiamethoxam (4.13-8.58)
Bitter gourd	9	ND	
Drumstick leaves	2	ND	
Kankun	3	ND	

Table 5(continued). Pesticide residues detected in fruits and vegetables sampled from consignments at National Plant Quarantine Service

Vegetable/ Fruit	No of samples analyzed	No of samples contaminated with pesticide residues	Range of pesticide residues detected
Kehelmuwa (banana blossom)	1	1	Profenophos (0.02)
Thibbatu	1	ND	
Thalanabatu/ Kohila/ Tomato/ Brinjal/ Capsicum	8	1	Profenophos(0.09) Carbofuran (0.53)
Spinach	7	1	Malathion (0.01), Thiamethoxam (0.06)
Thampala	4	ND	
Kathurumurunga	3	ND	
Green chilli	2	ND	
Total	95	24 (25%)	

ND < Limit of Detection (LOD); LOD -0.01mg/kg

Distribution of contaminated samples

Leafy vegetables were the highest group of samples which contaminated with detectable levels of pesticide residues. viz., 56 out of 89 samples (63%) were contaminated with one or more pesticides followed by vegetables (33%) and fruits (4%) (Table 6).

Table 6. Distribution of contaminated samples of fruits, leafy vegetables and vegetables

Category	No. of samples analyzed	No. of samples contaminated
Vegetables	196	29
Leafy vegetables	124	56
Fruits	20	4
Total	340	89

Analytical results revealed that only 26% of the samples out of the total samples which includes vegetables and fruits were positive for pesticide residues.

Detection of pesticide residues in leafy vegetables

While leafy vegetables have the highest contamination with pesticide residues, the comparative results showed that Gotukola and Mukunuwenna were the most contaminated with level at 73% and 68%, respectively (Figure 4). Spinach, Kathurumurunga and other

leafy vegetable samples showed very low percentage of contamination with pesticide residues.

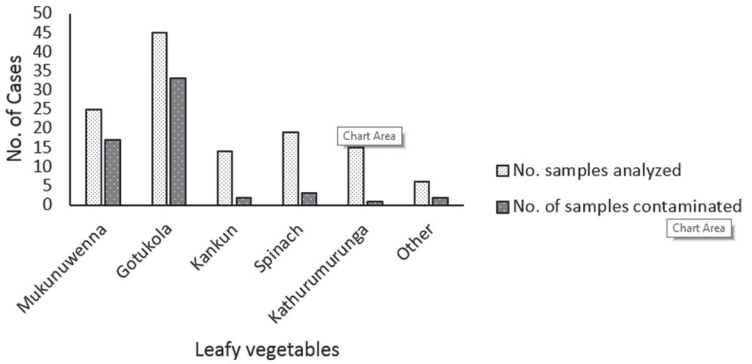

Figure 4. Number of leafy vegetable samples analyzed and contaminated

Pesticide residues in other vegetables

While pesticide contamination in winged bean (0), bitter gourd (2%) and okra (6%) are significantly controlled, snake gourd, beans and brinjal showed a significant contamination with pesticides at 25%,33% and 12% of samples analyzed, respectively (Figure 5).

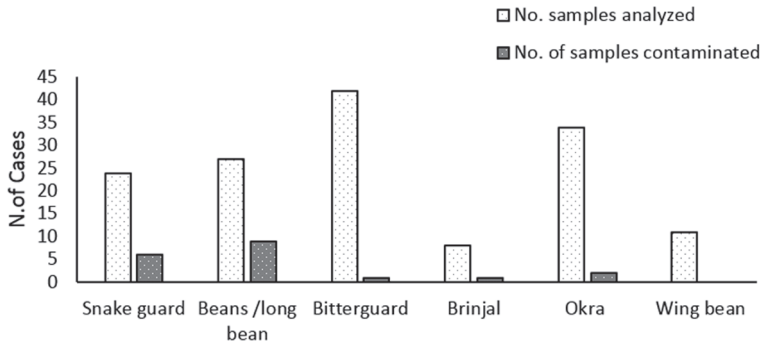

Figure 5. Comparison of number of samples analyzed and contamination for selected vegetables

Pesticide residues in fruits

Results revealed that out of banana, guava, mango, papaya, wood apple and pineapple samples tested for pesticide residues and only papaya has a significant contamination of pesticides amounting to 37% of papaya samples analyzed.

Percentage of samples exceeding the Maximum Residue Limits of EU (EU-MRLs)

Maximum Residue Limit for each pesticide as stipulated by EU is reported in Table 7. It was observed that 65 samples out of 89 were contaminated (72%) with levels exceeding the EU-MRL. When considers the total number of samples analyzed, only 19% samples exceeded the relevant EU-MRLs.

Pesticides detected as residues

Among twelve EU-authorized pesticides, Thiamethoxam was the most prominently detected compound; viz. 11 samples were contaminated in the whole sample analysis (Figure 6). Thiamethoxam is followed by Imidachloprid. Both Thiamethoxam and Imidachloprid are insecticide used for the control of aphids, whiteflies, trips, termites, mealy bugs and scales in vegetables. Thiophanate methyl, which was recommended for the plant diseases of anthracnose, powdery mildew and leaf spot was the most detected fungicide. EU MRLs and authorized pesticides in EU could be searched through EU Database (EU Pesticide Database).

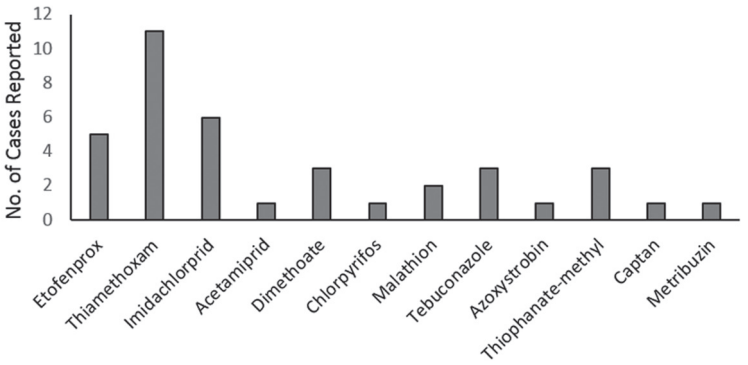

Figure 6. No. of cases reported on EU authorized pesticides

Meanwhile, as shown in the Figure 7, out of EU unauthorized pesticides nearly 12 pesticide compounds were detected in the sample analysis from farmer fields, pack houses and samples taken from the consignments ready for exportation at the NPQS, and Profenophos seemed to be the most prominent pesticide residue detected. Since, the production targets are aimed at the EU market, the detection of EU unauthorized pesticides in agricultural products appear to be a major risk factor for the EU export hub.

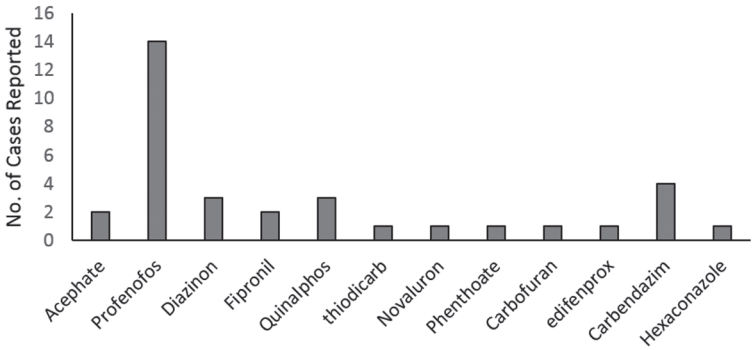

Figure 7. No. of cases reported on EU unauthorized pesticides

Pesticides in leafy vegetables

Table 7. Types of pesticide contaminants in leafy vegetables

Pesticide	No. of leafy samples contaminated	EU MRL mg/kg	Range	Mean± Standard deviation	Whether EU-authorized or not banned
Profenophos	32	0.01	0.02-69.61	5.27 ± 16.26	EU-authorized
Imidacloprid	2	2.0	0.01-13.93	6.97	
Fipronil	1	0.01	0.01	0.01	Banned
Diazinon	1	0.01	007	0.07	Banned
Thiamethoxam	6	0.01	0.01 – 115.10	19.24 ± 46.96	
Tebuconazole	12	0.5	0.02 – 72.2	6.38 ± 20.73	
Hexaconazole	1	0.01	0.06	0.06	Banned
Quinalphos	2	0.01	0.02 – 0.04	0.03	Banned
Captan	1	0.03	0.04	0.04	
Acephate	1	0.01	0.02	0.02	Banned
Etofenprox	2	3.0	0.01 – 0.02	0.015	
Azoxystrobin	1	0.01	0.011	0.011	
Carbendazim	1	0.01	0.09	0.09	Banned
Thiophanate-methyl	1	0.1	0.012	0.012	
Dimethoate	1	0.01	0.34	0.34	
Chlorpyriphos	1	0.05	0.02	0.02	
Malathion	2	0.02	0.01 – 0.04	0.03	
Metribuzin	1	0.1	0.16	0.16	

EU-MRL – Maximum Residue limits of EU

Altogether 124 leafy vegetable samples were analyzed for pesticide residues and 56 were contaminated with the pesticides given in Table 7. Nearly one third of these contaminated pesticides are EU unauthorized pesticides. More than 80% of pesticide residues detected in leafy vegetables are exceeded their relevant EU MRLs. Profenophos which belongs to Class II of WHO Hazard Level has been detected as the highly detected pesticide residue in most of the leafy vegetable samples and being having 14 days Pre-Harvest Interval (PHI). Profenophos has been listed as an EU unauthorized pesticide. It appears that the current production system of leafy vegetables is not conforming to the EU marketing requirements. Since there were no pesticide recommendations for the leafy vegetables up to end of 2017 (Department of Agriculture, 2015), farmers used to apply pesticides by their choice. Since leafy vegetables are harvested within short time intervals (i.e. weekly), the PHI of these pesticides may not be conforming to leafy vegetables (Sharaniya and Loganathan, 2016). Therefore, the harvest may contain higher residues, which in turn pave the way for high risk for the EU export market.

Department of Agriculture has stepped to expand SLGAP while conducting thorough monitoring programs for some of other crop categories including leafy vegetables which are intended to be exported to the EU market.Use of pesticides with low persistence such as bio-pesticides and use of other techniques without using pesticides are encouraged resulting in low accumulation of chemicals on treated crops.

According to the recommendations given by the EU intervention in 2016 (Director General of Health and Food Safety, 2016), Department of Agriculture has implemented programs for recommending suitable pesticides for leafy vegetables. However, thorough monitoring and awareness programs are needed to be implemented in all sectors involve in exporting of fruits and vegetables to the EU market. A comprehensive exporter registration system (including pack-houses and farms) also has been implemented by the Department of Agriculture. Conducting continuous monitoring on exporter facilities will reduce the export violations.

CONCLUSION

According to the results, 26% of tested samples are contaminated with pesticide residues. Among them 65% samples exceeded the Maximum Residue Limit of the

European Union (EU-MRL). Contamination of leafy vegetables was relatively higher than that of the other food categories analyzed in this study. Farmer practices in snake guard and bitter guard fields where they targeted EU market are conforming to Good Agriculture Practices (GAP). This has resulted low pesticide contaminations. Further attention needs to be paid on leafy vegetables. Pack houses which prepared fruits and vegetables for export need to pay more attention on controlling of pesticide residues while maintaining the traceability.

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