

Rapid Screening of Pesticide Residues in Organic-labeled and Conventional Vegetables in Southern Luzon, Philippines and Its Implication on Food Safety

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Pesticide residues on vegetables pose a major concern with regard to food safety. The Rapid Bioassay for Pesticide Residues (RBPR) is a spectrophotometric method used to rapidly screen residues of acetylcholinesterase-inhibiting pesticides – namely, organophosphates (OP) and carbamates (CM) in vegetable samples. In this study, RBPR was utilized to detect pesticide residues in organic-labeled and conventional vegetables from various markets and stalls in selected areas in Southern Luzon, Philippines from January 2016–March 2018. Samples (n = 443) that resulted in > 20% inhibition in RBPR, the value considered as a permissible error, were considered positive for OP and/or CM residues. Results showed that 70 (15.8%) out of 443 samples were positive based on RBPR. Vegetable stalls from Laguna and Quezon, and Laguna public markets got the highest positivity of samples. Based on the farming method, 18.3% (n = 262) were positive in the conventional samples, as compared to 12.2% (n = 181) in the organic-labeled samples. Positivity rate was observed to be highest in bitter melon, *pechay*, and tomato. A few samples (n = 6) were then analyzed by gas chromatography–mass spectrometry (GC-MS) as a confirmatory test. One organic-labeled sample was found to have exceeded the existing MRL based on GC-MS analysis. Overall, the study showed the presence of pesticide residues in commonly consumed conventional and even organic-labeled vegetables in Southern Luzon. The positivity of many organic-labeled samples is of serious concern as it may indicate fraud or mislabel and non-compliance to principles of organic farming. This also highlights the critical need for extensive intervention strategies to limit the potential health risk to consumers. It is advised that pesticide residues be monitored on a regular basis and that farmers be educated about better pesticide safety procedures, particularly the importance of adhering to prescribed pre-harvest intervals (PHI).

Keywords: conventional, food safety, organic-labeled, pesticide residues, rapid bioassay of pesticide residues

INTRODUCTION

In the agriculture-based Philippines, many farmers rely on pesticides to ensure high yield and quality of produce. A warm climate and humid environment favor the

occurrence of pest and disease infestations which propel farmers to apply pesticides on their crops. Vegetables, which are considered high-value crops, are important sources of invaluable nutrients and are often produced through intensive pesticide use (Ulrichs *et al.* 2011; Magcale-Macandog *et al.* 2016). Despite the benefits of

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consuming vegetables, the presence of pesticide residues poses food safety concerns, especially when eaten raw (Reiler *et al.* 2015).

Injudicious use of pesticides like overdosing, dipping of produce in insecticide solution, use of spray cocktails, neglecting label recommendations, and use on non-appropriate crops are some practices done by some farmers in the country (Bajet and Tejada 1995; Ulrichs *et al.* 2011). When harvesting, some farmers tend to ignore PHI, which is the waiting time between the last pesticide application and the harvest of treated crops (Latif *et al.* 2011). These practices often lead to a higher concentration of pesticide residues in food, health hazards to the pesticide applicator, and contamination of the environment (Ulrichs *et al.* 2011; Perez *et al.* 2015). Moreover, exceedance on the maximum residue limits (MRLs) has been a global concern in the past decade (Latif *et al.* 2011; Srivastava *et al.* 2011; Bempah *et al.* 2012; Sinha *et al.* 2012; Chowdhury *et al.* 2013; Wanwimolruk *et al.* 2015; Philippe *et al.* 2021). An MRL is the highest level of pesticide residue that is legally tolerated in or on food or feed when pesticides are applied correctly in accordance with good agricultural practices (GAPs) (FAO/WHO 2018). While MRLs are not to be perceived as levels of safety concern (Winter and Jara 2015) and exceedance of MRLs is generally understood to be of rare health significance (Winter 1992), some proponents are claiming harmful health effects of pesticides even in minute amounts (Hayes *et al.* 2006).

This variety of problems has led some consumers to purchase organic vegetables in order to avoid pesticide residues (WFM 2005). The growing demand for safer food in the country has led to the appropriation of Republic Act No. 10068 or the Organic Act of 2010, which is a law to “promote, propagate, develop further, and implement the practice of organic agriculture in the Philippines” (Official Gazette 2010). Organically grown vegetables sound healthier; however, there are still risks for organic-labeled commodities as there is a possibility of non-compliance (Bajet *et al.* 2016).

A rapid detection method will be useful in screening and detecting pesticide residues in large samples of agricultural commodities. The RBPR was developed in Taiwan and is currently used as a screening test in markets and farms. The RBPR is a cheap and simple test to detect OP and CM groups of pesticides on fruits and vegetables in as fast as 10 minutes (Kao *et al.* 2010). The method works by the inhibition of the enzyme acetylcholinesterase due to OP and CM pesticides, which will result in a colorless solution. On the other hand, the absence of the OP and CM pesticides will allow the enzyme to catalyze acetylcholine to form acetic acid and choline. The generated choline reacts with the Ellman’s reagent 5,5'-dithiobis-(2-

nitrobenzoic acid) (DTNB) to form a yellow 5-thio-2-nitrobenzoic acid. This color reaction can be measured quickly using a spectrophotometer. The method can thus be used as a screening tool in various vegetables and other commodities to determine their present risks and safety due to pesticide residues. RBPR can also be a basis for decisions prior to consumption or market acceptance. While there are modern instrumental methods to analyze pesticide residues like that of GC and high-performance liquid chromatography (HPLC), they are expensive and will require several days to obtain results, and thus, cannot be used for screening a large number of vegetable samples.

This study aimed to screen organic-labeled and conventional vegetables in various markets and stalls in Southern Luzon, Philippines, particularly in Laguna and Quezon provinces, and in Metro Manila for OP and CM pesticide residues using RBPR. Laguna and Quezon are active producers of lowland vegetables, wherein the bulk of the vegetables produced in these areas are supplied to Metro Manila and nearby provinces. Metro Manila, on the other hand, has supermarkets where high-end produce and organic products are sold. Furthermore, the study aimed to explain the results of the analyses with respect to its potential impact on food safety.

MATERIALS AND METHODS

Chemicals

The complete set of chemicals for RBPR was purchased from Si Cheng Biotechnology Co., Taiwan. The kit consists of phosphate buffer pH 7.5, acetylcholinesterase enzyme, DTNB, acetylcholine iodide (ATCI). Sodium bromide and ethanol were purchased from Scharlau Chemicals. Bromine water was prepared using sodium bromide.

Vegetable Sampling

Conventional and organic-labeled vegetables were randomly collected from January 2016–March 2018. The vegetables collected were eggplant [*Solanum melongena*], tomato [*Solanum lycopersicum*], lettuce [Romaine, green ice, iceberg, *Lactuca sativa* ssp.], pechay [*Brassica rapa* ssp.], Taiwan pechay [*chingkang*, *Brassica rapa* var *chinensis*], bitter melon [*Momordica charantia*], cucumber [*Cucumis sativus*], snap beans [*Phaseolus vulgaris*], cabbage [*Brassica oleracea*], Chinese cabbage [*wombok*, *Brassica rapa* var *pekinensis*], yardlong beans [*Vigna unguiculata* ssp. *sesquipedalis*], *etc.* (Table 1). The vegetables gathered were from town markets, trading posts, supermarkets, and outlet stalls in Laguna, Quezon, and Metro Manila (Table 2). Vegetable samples were classified as organic-labeled if it is collected from

Table 1. Distribution of samples according to the type of farming method.

Crop	Type		Total
	Conventional no. of samples	Organic-labeled no. of samples	
Bell pepper	1	6	7
Bitter melon	47	9	56
Cabbage	8	15	23
Celery	0	2	2
Chili	9	0	9
Cucumber	5	13	18
Eggplant	44	16	60
French bean	0	7	7
Leek	0	1	1
Lettuce	10	42	52
Mustard	0	1	1
Okra	16	3	19
Pechay	27	10	37
Sitao	27	5	32
Snap bean	22	6	28
Taiwan pechay	11	9	20
Tomato	34	25	59
Wombok	1	10	11
Zucchini	0	1	1
Total	262	181	443

[1] a supermarket and indicated in the label as organic; [2] farm or farm outlet, which is considered organic; [3] organic farmer association group and outlets; [4] labeled with information denoting the non-use of pesticides such as “no chemicals, pesticide-free”; or [5] as declared by the farmer/seller. Unlabeled produce was assumed to be produced through conventional farming methods and, thus, labeled as conventional. The weight of the samples collected ranged from 0.25–1.0 kg depending on the size and surface area of the vegetable.

Sample Preparation

For leafy vegetables, four pieces of around 2.5-cm leaf disks were obtained from four randomly selected leaves using a cylindrical stainless steel cutter with a diameter of 2.4 cm. The leaf discs, approximately equivalent to 1 g, were further cut into small pieces using a cutter knife. The cut samples were transferred to small test tubes; two replicates were analyzed using RBPR without bromine water and another two replicates using RBPR with bromine water. The purpose of the bromine water was to

differentiate OP insecticides that have a phosphorus-sulfur double bond (P=S) and a phosphorus-oxygen double bond (P=O) moiety. The addition of the reagent will cause an oxidation reaction that can be measured as five to 10 times increased in % inhibition.

For non-leafy vegetables, the surface of each vegetable sample was peeled and cut into small pieces using a cutter knife. Then, around 1 g of the samples were transferred to small test tubes; two replicates were analyzed using RBPR without bromine water and another two replicates using RBPR with bromine water.

All materials used for sample preparation were wiped with cotton soaked in 95% ethanol in between samples to prevent possible cross-contamination.

Sample Extraction

For the RBPR test without bromine water, 1.0 mL of 95% ethanol was added to each of the tubes, mixed for 20–30 s using a vortex mixer, and were left to stand for 3 min. The extracts were then decanted into clean tubes. For RBPR

with bromine water, 2.0 mL of 95% ethanol and 200- μ L bromine water were added to each of the tubes, mixed for 20–30 s using a vortex mixer, and were left to stand for 30 min to allow dissipation of the excess bromine solution. The extracts were then decanted into new tubes.

Enzyme Inhibition Assay

In a cuvette, 3.0 mL of phosphate buffer, 20 μ L of acetylcholinesterase, and 20- μ L sample extract were added and mixed immediately. After 2.5 min, 100- μ L DTNB (5,5-dithio-bis-(2-nitrobenzoic acid) was added to the mixture and at exactly 3.0 min after the initial mixing with the sample extract, 20- μ L ATCI was added. The mixture was mixed immediately and read in a spectrophotometer (Shimadzu UV Visible Spectrophotometer Model 1240) set at 412 nm. The assay was done at room temperature. Using the built-in RBPR program, results were automatically expressed as % inhibition. Manual computation to calculate % inhibition was done using the following formula:

$$\% \text{ inhibition} = \frac{\text{absorbance (control)} - \text{absorbance (sample)}}{\text{absorbance (control)}} * 100 \quad (1)$$

Data Analysis

Samples with % inhibition greater than 20% were considered positive according to Tejada *et al.* (1998). Considering that residues may not be uniformly distributed on the vegetables, a conservative measure was applied.

Quantification of Pesticide Residues by GC-MS

Six randomly selected vegetables were analyzed by GC-MS at the BPI-NPAL using a validated multi-residue method. The method covers the analysis of eleven organophosphate pesticides (mevinphos, dimethoate, diazinon, isazophos, methyl parathion, fenitrothion, malathion, chlorpyrifos, phenthoate, profenofos, and triazophos). Six pyrethroids were included in the analysis (lambda-cyhalothrin, permethrin, cyfluthrin, cypermethrin, fenvalerate, and deltamethrin) and eight organochlorines (lindane, aldrin, heptachlor, alpha and beta endosulfan, endosulfan sulfate, heptachlor epoxide, and 4,4-DDE). Organochlorines are not registered for use on vegetables and are banned in the Philippines but are part of the validated multi-residue method of BPI-NPAL.

RESULTS AND DISCUSSION

Analysis of Pesticide Residue Using RBPR

A total of 443 vegetable samples were collected from selected local markets, supermarkets, trading posts, vegetable stalls, and a consolidator in Metro Manila and

the provinces of Laguna and Quezon in the Philippines. Samples collected include local *pechay*, Taiwan *pechay*, Chinese cabbage or *wombok*, eggplant, lettuce (Romaine, green ice, and iceberg), tomato, snap beans, yardlong beans, French beans, bitter melon, cucumber, and pepper (bell pepper and hot pepper), among others (Table 1). Samples from a sampling site do not necessarily mean that they originated from the same area. For instance, one organic vegetable consolidator in Metro Manila obtains its vegetables from farm cooperators in Benguet and Rizal provinces, and trading posts in Quezon are sourcing its vegetables from nearby towns and even from provinces in Northern Luzon. The samples were collected from January 2016–March 2018 and were classified as conventional and organic-labeled with a total of 262 and 181 vegetable samples, respectively.

Results based on the location of the selected sampling sites are presented in Table 2. A positive RBPR result means that enzyme inhibition is greater than 20%, which indicates the presence of either OP, CM, or a combination of both. Overall, results showed that 70 out of 443 samples were positive (15.8%).

Based on the type of sampling sites for each location, vegetable stalls from Laguna and Quezon, and Laguna public markets came up with the highest percentages of positive samples at 27.3, 25.0, and 20.8%, respectively (Table 2). Higher positivity may be due to higher conventionally grown vegetable samples in these sites. Vegetable stalls in Laguna were commonly located beside the road where farms are in the vicinity. Samples were observed to be recently harvested and, thus, may have higher pesticide residues. As for Quezon vegetable stalls, these are a group of stalls located in a more urban setting. Vegetables were also likely recently harvested as well, and what is worse is that many of these samples were organic-labeled. Meanwhile, vegetables coming from public markets mostly were sourced from conventional farms and these farms are usually not supervised nor monitored. This then may lead to injudicious use of pesticides, resulting in higher residues on vegetables. Metro Manila stores, on the other hand, had a lower percentage of positive samples (9.6–10.0%) as many of them have a greater proportion of organic-labeled samples, especially the stalls and supermarkets in this study. In general, it was found out that Laguna (21.6%, $n = 176$) has the highest positivity rate, followed by Quezon (16.7%, $n = 84$) and Metro Manila (9.8%, $n = 183$). It should be noted that the sampling sites are limited and may not reflect the entirety of the region.

Table 3 summarizes the RBPR results based on the farming method. As expected, conventional samples had higher positivity of 18.3% ($n = 262$) compared to the organic-labeled samples, which was 12.2% ($n = 181$). Conventional positive samples may possibly indicate that

Table 2. Percent positivity and distribution of samples according to location and types of sampling site.

Location	Type of sampling site	No. of samples	No. of positive samples ^a	Positivity (%)
Metro Manila	Supermarkets	70	7	10.0%
	Consolidator	73	7	9.6%
	Stalls	40	4	10.0%
	Subtotal	183	18	9.8%
Laguna	Public markets	154	32	20.8%
	Stalls	22	6	27.3%
	Subtotal	176	38	21.6%
Quezon	Trading posts	72	11	15.3%
	Stalls	12	3	25.0%
	Subtotal	84	14	16.7%
Total		443	70	15.8%

^aSamples are regarded as positive when percent inhibition is > 20%

Table 3. Percent positivity of samples according to farming method.

Type	Total samples	No. of positive samples ^a	Positivity (%)
Organic	181	22	12.2
Conventional	262	48	18.3
Total	443	70	15.8

^aSamples are regarded as positive when percent inhibition is > 20%

pesticide label recommendations in terms of application rate, application interval or frequency, and/or the PHI were not followed. PHI is the minimum amount of time (in days) between the last pesticide application and when a crop can be harvested. This is unique for each pesticide-crop combination, which may last for 1–28 d. PHI ensures that the crop will meet the established pesticide residue tolerances (Fouche *et al.* 2000). The non-compliance of some farmers to the PHI may be attributed to the early harvesting due to several factors such as lack of knowledge on the concept of pesticide residues and PHI, high market price, high market demand (Darko and Akoto 2008), incoming typhoon, severe pest infestation/outbreak, among others.

Meanwhile, the percentage of positive organic-labeled samples (12.2%) can be attributed to at least one of the following: [1] non-compliance of some organic vegetable growers to the non-synthetic pesticide use principle of organic farming, [2] inadequate/lack of pesticide residue analysis for organic produce as part of the monitoring of compliance to organic authenticity, [3] drift of synthetic pesticides from nearby conventionally managed fields, [4] and mislabeling or false claims of some producers/

sellers of organic vegetables (Benbrook *et al.* 2021). The relatively high number of positive samples may be of serious concern, especially since this is contrary to what the Organic Agriculture Act of 2010 is advocating. According to the law, mislabeling is punishable by imprisonment and/or a fine of not more than PHP 50,000.00 (Official Gazette 2010).

Based on the type of vegetable and regardless of farming method, bitter melon has the greatest positivity rate of 33.9% (n = 56) followed by *pechay* (29.7%, n = 37), tomato (22.0%, n = 59), lettuce (19.2%, n = 52), and eggplant (13.3%, n = 60) (Figure 1). This is in exception to celery, which has only two samples and has a 50% positivity rate. Some commodities are much fewer in the number of samples because of their non-availability in the sampling sites. The results were found similar to the study done by Tipa *et al.* (1997). According to the study, conventional vegetables such as eggplant, tomato, and *pechay* were also found to be positive by RBPR, with tomato being the most frequently positive among the 11.3% positive samples (n = 168) detected. Moreover, the results of the pesticide residue monitoring conducted by the BPI-NPAL from 2013–2015 as reported by Magcale-

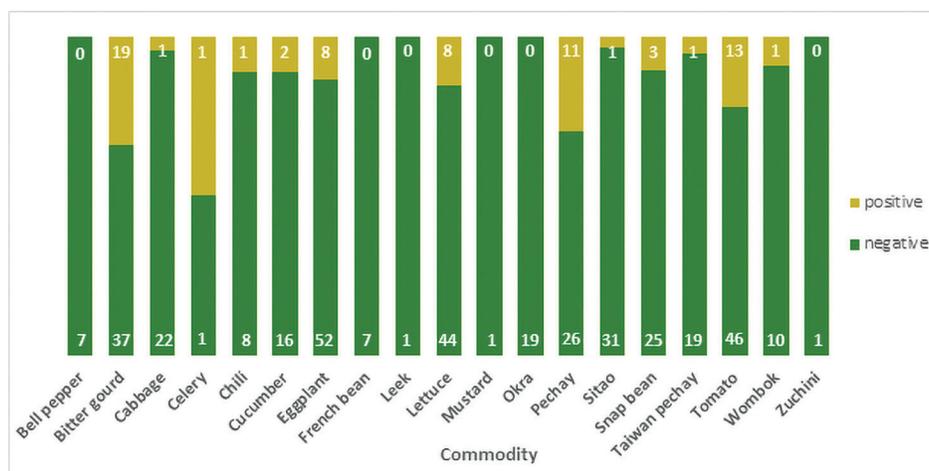


Figure 1. Relative distribution of positive and negative samples per commodity based on RBPR results. The respective amount of positive and negative samples per commodity is labeled on the top and bottom portion of the bars, respectively.

Macandog *et al.* (2016) showed that the OPs were indeed one of the most common pesticides in vegetables (tomato, eggplant, *pechay*, bitter gourd). The organophosphate insecticides detected were profenofos, chlorpyrifos, dimethoate, malathion, and phenthoate. Other pyrethroid insecticides were also detected as well.

In the presence of bromine water, the method can differentiate OPs from other OPs and CMs due to enhanced detection of phosphorothioates, which are OPs characterized by a sulfur-phosphorus double bond (Vale and Lotti 2015). Examples of these are malathion, chlorpyrifos, fenthion, fenitrothion, and diazinon. Bromine water converts these compounds to their corresponding oxo-analogs (Figure 2), which inhibits acetylcholinesterase more effectively, resulting in increased sensitivity for these compounds. This effect is shown by the higher percent inhibition in the result with bromine water as compared to the treatment without bromine water. Of the positive samples analyzed, 16 out of 70 (22.9%) resulted in a significant increase in % inhibition in RBPR with bromine water, indicating the presence of a phosphorothioate.

Based on the recommendations of Tejada *et al.* (1998), samples with < 20% inhibition can be allowed to be sold and traded in the market, while samples with % inhibition between 20–50% can also be allowed to be traded given

that the commodities are washed first. However, for samples with > 50% inhibition, the commodities may be withheld, and farmers or traders must be advised to follow the recommended interval between the last pesticide application and the harvest of crops to ensure that the pesticide residues are within the acceptable limits. From the results of the study, 85.7% (n = 70) of the positive samples had 20–50% inhibition while 14.3% (n = 70) of the positive samples had > 50% inhibition. Vegetable samples that can be safely traded (samples with < 20% inhibition) include different upland and lowland crops such as cabbage, lettuce, snap bean, *wombok*, eggplant, *pechay*, tomato, and bitter gourd. Meanwhile, vegetables that resulted in > 50% inhibition were mostly lowland crops such as tomato, bitter gourd, and *pechay*.

Confirmatory Analysis of Pesticide Residues by GC-MS

As shown in Table 4, negative samples tested by the RBPR (sample nos. 4 and 6) were confirmed by GC-MS to be negative to OP as shown by the < LOQ (limit of quantification) results (0.01 mg/kg). Among the four positive samples tested by the RBPR, organic-labeled *pechay* (sample no. 3) was found to have the highest % inhibition at 92.3% using RBPR with bromine. This very high result as compared to 8.6% inhibition using RBPR without bromine indicates the presence of a phosphorothioate. Chlorpyrifos, a phosphorothioate, was

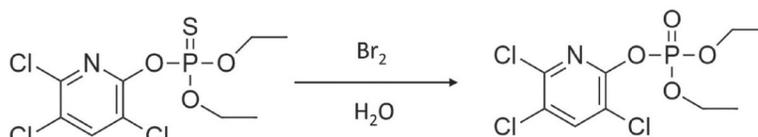


Figure 2. Conversion of chlorpyrifos to its oxo-analog.

Table 4. Comparative results of RBPR and GC-MS analysis of selected vegetable samples.

Sample number	Sample type	Vegetable	% inhibition, without bromine	% inhibition, with bromine	RBPR ^a result	GC-MS ^b result (mg/kg)
1	Conventional	Snap bean	9.8	27.3	Positive	< LOQ ^c
2	Conventional	Eggplant	18.4	24.0	Positive	< LOQ
3	Organic-labeled	<i>Pechay</i>	8.6	92.3	Positive	Profenofos = 0.45, Chlorpyrifos = 0.27
4	Organic-labeled	Eggplant	15.1	19.4	Negative	Lambda-cyhalothrin = 0.01, Cypermethrin = 0.03
5	Organic-labeled	Yardlong bean	38.6	31.3	Positive	Cypermethrin = 0.08
6	Organic-labeled	Yardlong bean	0.4	1.1	Negative	< LOQ

^aRapid bioassay of pesticide residues

^bGas chromatography–mass spectrometry

^cLimit of quantification = 0.01 mg/kg

Table 5. Samples with detected pesticide residues using GC-MS and its corresponding existing MRLs.

Sample	GC-MS result (mg/kg)	CAC MRL ^a (mg/kg)	EU MRL ^b (mg/kg)	USDA ^c MRL (mg/kg)
<i>Pechay</i>	Profenofos = 0.45	_d	0.01	–
	Chlorpyrifos = 0.27	1	0.01	1
Eggplant	Lambda-cyhalothrin = 0.01	0.3	0.3	0.2
	Cypermethrin = 0.03	0.03	0.5	–
Yardlong bean	Cypermethrin = 0.08	0.7	0.7	–

^aCodex Alimentarius Commission maximum residue limit (<https://www.fao.org/fao-who-codexalimentarius/codex-texts/dbs/pestres/pesticides/en/>)

^bEuropean Union maximum residue limit (<https://ec.europa.eu/food/plant/pesticides/eu-pesticides-database/mrls/?event=search.pr>)

^cUnited States Department of Agriculture maximum residue limit (<https://www.fas.usda.gov/maximum-residue-limits-mrl-database>)

^dMeans no available maximum residue limit

found to be present at 0.27 mg/kg, while profenofos was at 0.45 mg/kg. Organic-labeled yardlong bean (sample no. 5), conventional snap bean (sample no. 1), and conventional eggplant (sample no. 2) were also found to be positive using RBPR, but GC-MS results did not indicate the presence of OPs at concentrations above 0.01 mg/kg, which is the LOQ of the method. Samples may contain other pesticide active ingredients not covered by the GC-MS multi-residue method (covers only OPs, organochlorines, and pyrethroids). On the other hand, the pyrethroid insecticides cypermethrin and lambda-cyhalothrin were detected in organic eggplant (sample no. 4) at 0.03 mg/kg and 0.01, mg/kg, respectively, while cypermethrin was found in organic-labeled yardlong bean (sample no. 5) at 0.08 mg/kg.

Table 5 presents the residue concentrations of the positive samples as detected by the GC-MS as compared with their respective MRLs. For the residues in *pechay*, 0.45

mg/kg profenofos detected exceeded the European Union MRL (EU MRL), whereas 0.27 mg/kg chlorpyrifos detected exceeded the EU MRL but was below the 1 mg/kg CODEX MRL and United States MRL (US MRL) for a related vegetable Chinese cabbage. In addition, the 0.03 mg/kg cypermethrin and 0.01 mg/kg lambda-cyhalothrin detected in eggplant did not exceed the CODEX, EU, and US MRL. In the case of the detected cypermethrin for yardlong bean (0.08 mg/kg), it did not exceed the CODEX and EU MRL. The toxicity of acetylcholinesterase-inhibiting compounds such as OPs has been documented and it may pose a potential risk to the metabolic, neurological, and endocrine systems (Kumar *et al.* 2016).

Implication on Food Safety

The result of this study shows the presence of pesticide residues in conventional and organic-labeled vegetables

from the monitoring conducted. One organic-labeled vegetable even exceeded the existing MRLs indicated for chlorpyrifos and profenofos. According to Hughner *et al.* (2007) and Stolz *et al.* (2011), consumers prefer to buy organically grown vegetables and fruits due to the absence of pesticide residues and other potentially unwanted chemicals. The presence of pesticide residues in organic-labeled samples poses a bigger concern since the use of synthetic pesticides is prohibited for organic farming. To the best of our knowledge, these positive results of organic-labeled vegetables were the first documented scientific evidence in the Philippines of the presence of pesticide residues in vegetables being marketed as “organic.”

Pesticide contamination in organic food cannot be absolutely avoided; however, it should at most contain trace amounts only due to unintentional contamination and should not exceed the MRLs as compared to conventional food. This was observed by Baker *et al.* (2002) when they conducted a broad study on the pesticide residue monitoring of organically grown crops, conventional, and integrated pest management crops (IPM) or IPM grown crops. They hypothesized that the organically grown crops would result in few incidences of pesticide residues as compared to the IPM grown and conventionally grown crops. The study concluded that pesticide was present in some organic samples, but it was three times lower than in conventional production.

Moreover, according to the US Department of Agriculture (DA) National Organic Program, the standard for unavoidable contamination for organically grown crops is 5% of the relevant US Environmental Protection Agency (US EPA) limit. Based on the results of this study, the positive organic-labeled *pechay* with profenofos and chlorpyrifos residue exceeded the 5% set limit, which means that the organic-labeled *pechay* contains a high amount of pesticide residues. Since organic vegetables retail at a higher price compared to the conventional counterpart, there may be cases of claiming conventionally produced crops as organic, for instance in the US (Global News 2019). Non-compliance and mislabeling incidents must be addressed by the organic community and government agencies and, possibly, develop effective systems for investigating and correcting them.

Detection of pesticides with the same mode of action is also a cause of concern, with the mixing of multiple pesticides previously reported (Claeys *et al.* 2011; BPI-NPAL, as reported by Magcale-Macandog *et al.* 2016). The use of pesticides with the same mode of action for a long period of time may hasten the buildup of pest resistance; therefore, farmer education on good pesticide management anchored to GAP must be done. Ultimately, improper agricultural application of pesticides can have

major environmental and health effects. Hence, there is a need to enhance farmer education on the use of pesticides and observed pesticide residue levels on vegetables and fruits. Compliance with national food safety regulations should be monitored on a regular basis.

CONCLUSION

Through RBPR, large samples of conventionally grown vegetables – as well as the organic-labeled vegetables collected in various sites in Southern Luzon, Philippines – were screened and determined to be contaminated with pesticide residues. One organic-labeled sample was found to exceed the set MRL based on GC-MS analysis. Positivity in many organic-labeled samples is of significant concern as it may indicate non-compliance to organic principles of farming or even mislabeling. To the best of our knowledge, this is the first scientific report to indicate the presence of pesticide residues in organic-labeled samples in the country. Therefore, continuous pesticide residue monitoring in agricultural commodities is highly recommended. Also, intensive farmer education with a focus on good pesticide management and pesticide label recommendations should be implemented.

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REFERENCES

- BAJET CM, TEJADA AW. 1995. Pesticide Residues in the Philippines: an Analytical Perspective. *Trends in Analytical Chemistry* 14(9): 430–434.
- BAJET CM, MANUBEN JJ, SARMIENTO J, CRUZ EJ. 2016. Pesticide residues on vegetables using rapid detection tools: an update. *Philippine Entomologist (Philippines)*.

- BAKER BP, BENBROOK CM, III EG, BENBROOK KL. 2002. Pesticide residues in conventional, integrated pest management (IPM)-grown and organic foods: insights from three US data sets. *Food Additives & Contaminants* 19(5): 427–446.
- BEMPAH CK, BUAH-KWOFIE A, ENIMIL E, BLEWU B, AGYEI-MARTEY G. 2012. Residues of organochlorine pesticides in vegetables marketed in Greater Accra Region of Ghana. *Food Control* 25(2): 537–542.
- BENBROOK C, KEGLEY S, BAKER B. 2021. Organic Farming Lessens Reliance on Pesticides and Promotes Public Health by Lowering Dietary Risks. *Agronomy* 11(7): 1266.
- CHOWDHURY MAZ, FAKHRUDDIN ANM, ISLAM N, MONIRUZZAMAN M, GAN SH, ALAM K. 2013. Detection of the residues of nineteen pesticides in fresh vegetable samples using gas chromatography–mass spectrometry. *Food Control* 34(2): 457–465.
- CLAEYS WL, SCHMIT JF, BRAGARD C, MAGHUIN-ROGISTER G, PUSSEMIER L, SCHIFFERS B. 2011. Exposure of several Belgian consumer groups to pesticide residues through fresh fruit and vegetable consumption. *Food Control* 22(3–4): 508–516.
- DARKO G, AKOTO O. 2008. Dietary intake of organophosphorus pesticide residues through vegetables from Kumasi, Ghana. *Food and Chemical Toxicology* 46(12): 3703–3706.
- [FAO/WHO] Food and Agriculture Organization of the United Nations/ World Health Organization. 2018. Maximum Residue Limits. Retrieved on 01 Dec 2017 from <http://www.fao.org/fao-who-codexalimentarius/codex-texts/maximum-residue-limits/en/>
- FOUCHE C, MOLINAR R, CANEVARI M, JOSHEL C, MULLEN B, WEBER J. 2000. Pesticides for Specialty Crops [PDF]. Retrieved on 01 Aug 2018 from <https://anrcatalog.ucanr.edu/pdf/7253.pdf>
- HAYES TB, CASE P, CHUI S, CHUNG D, HAEFFLE C, HASTON K, LEE M, MAI VP, MARJUOA Y, PARKER J, TSUI M. 2006. Pesticide mixtures, endocrine disruption, and amphibian declines: are we underestimating the impact? *Environmental Health Perspectives* 114(Suppl 1): 40–50.
- GLOBAL NEWS. 2019. Head of America’s largest organic food fraud scheme sentenced to 10 years. Retrieved on 18 Sep 2021 from https://globalnews.ca/news/5778147/organic-food-fraud-scheme/?fbclid=IwAR2CjZlxUqqNB_tUw8VzAIHVv7C512MLp-S9tEDaJZZHWSQ0xxa9q8p-XWrg
- HUGHNER RS, MCDONAGH P, PROTHERO A, SCHULTZ CJ, STANTON J. 2007. Who are organic food consumers? A compilation and review of why people purchase organic food. *Journal of Consumer Behaviour: an International Research Review* 6(2–3): 94–110.
- KAO CH, HSIEH YS, CHIANG MY, HUANG YB. 2010. Residues Control by Using Rapid Bioassay of Pesticide Residues (RBPR) for Market Inspection and Farm Education. In: *Technology on Reducing Post-harvest Losses and Maintaining Quality of Fruits and Vegetables*. Huang CC, Yang CM, Ou SK, Chen JJ eds. Proceedings of 2010 AARDO Workshop; Taiwan Agricultural Research Institute, Wufeng, Taichung Hsien, Taiwan, ROC. p. 72–82.
- KUMAR S, KAUSHIK G, FRANCISCO VCJ. 2016. Scenario of organophosphate pollution and toxicity in India: a review. *Environ Sci Pollut Res* 23(10).
- LATIF Y, SHERAZI STH, BHANGER MI. 2011. Assessment of pesticide residues in commonly used vegetables in Hyderabad, Pakistan. *Ecotoxicology and Environmental Safety* 74(2011): 2299–2303.
- MAGCALE-MACANDOG D, PARAISO PM, SALVACION A, ESTADOLA R, QUINONES S, SILAPANIM and BRIONES R. 2016. An Overview of Agricultural Pollution in the Philippines: the Crops Sector. International Bank for Reconstruction and Development, The World Bank, Washington, DC.
- OFFICIAL GAZETTE. 2010. Republic Act No. 10068. Retrieved from <https://www.officialgazette.gov.ph/2010/04/06/republic-act-no-10068/>
- PEREZ IC, GOOC CM, CABILI JR, RICO MJ, EBASAN MS, ZARAGOSA MJ, LACUNA ML. 2015. Pesticide use among farmers in Mindanao, Southern Philippines. *Advances in Environmental Sciences* 7(1): 90–108.
- PHILIPPE V, NEVEEN A, MARWA A, BASEL AYA. 2021. Occurrence of pesticide residues in fruits and vegetables for the Eastern Mediterranean Region and potential impact on public health. *Food Control* 119: 107457.
- REILER E, JÓRS E, BÉLUM J, HUICI O, CAERO MMA, CEDERGREEN N. 2015. The influence of tomato processing on residues of organochlorine and organophosphate insecticides and their associated dietary risk. *Science of the Total Environment* 527–528(15): 262–269.
- SINHA SN, VASUDEV K, VISHNU VARDHANA RAO M. 2012. Quantification of organophosphate insecticides and herbicides in vegetable samples using the “Quick Easy Cheap Effective Rugged and Safe” (QuEChERS) method and a high-performance liquid

- chromatography–electrospray ionisation–mass spectrometry (LC–MS/MS) technique. *Food Chemistry* 132(3): 1574–1584.
- SRIVASTAVA AK, TRIVEDI P, SRIVASTAVA MK, LOHANI M, SRIVASTAVA LP. 2011. Monitoring of pesticide residues in market basket samples of vegetables from Lucknow City, India: QuEChERS method. *Environmental Monitoring and Assessment* 176(1–4): 465–472.
- STOLZ H, STOLZE M, JANNSEN M, HAMM U. 2011. Preferences and determinants for organic, conventional and conventional-plus products – the case of occasional organic consumers. *Food Quality and Preference* 22(8): 772–779.
- TEJADA AW, VARCA LM, CALUMPANG SMF, BAJET CM. 1998. Enzyme Inhibition and Other Rapid Techniques for Pesticide Residue Detection. In: Seeking agricultural produce free of pesticide residues. Kennedy IR, Sherritt JH, Johnson GI, Highley E eds. *Proceedings of an International Workshop; 17–19 Feb 1998; Yogyakarta, Indonesia.*
- TIPA EV, TEJADA AW, BARRIL CR, MERCA FE, QUINTANA BB. 1997. Rapid bioassay of pesticide residues in vegetable samples from Laguna, Philippines. *Philippine Agriculturist*. Retrieved on 10 Dec 2017 from <http://agris.fao.org/agris-search/search.do?recordID=PH1999100539>
- ULRICHS C, BURLEIGH JR, MEWIS I. 2011. The effects of IPM and farmer practices on yield and pesticide residues of *pakchoi* (*Brassica rapa* L. cv *pakchoi*) in Central Luzon, Philippines. *Tropics* 19(3): 113–122.
- VALE A, LOTTI M. 2015. Organophosphorus and carbamate insecticide poisoning. *Handbook of Clinical Neurology* 131: 149–168.
- WANWIMOLRUK S, KANCHANAMAYOON O, PHOPIN K, PRACHAYASITTIKUL V. 2015. Food safety in Thailand 2: pesticide residues found in Chinese kale (*Brassica oleracea*), a commonly consumed vegetable in Asian countries. *Science of the Total Environment* 532: 447–455.
- [WFM] Whole Foods Market. 2005. *Whole Foods Market Organic Trend Tracker*. Austin, TX.
- WINTERCK. 1992. Pesticide tolerances and their relevance as safety standards. *Regulatory Toxicology and Pharmacology* 15: 137–150.
- WINTER CK, JARA EA. 2015. Pesticide food safety standards as companions to tolerances and maximum residue limits. *Journal of Integrative Agriculture* 14(11): 2358–2364.