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#### Research article

## Multiple Pesticide Residues Found in Vegetables and Fruits from Rural and Urban Markets in Upper Northern Thailand

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**Abstract** Multiple pesticides are used in vegetable and fruit cultivation worldwide, including in Thailand. A survey series on pesticide residues in vegetable and fruit samples sourced from markets in urban and rural areas of upper northern Thailand during 2007–2013. Sixteen different vegetables (n = 412) and 11 different fruits (n = 301) were analyzed for 43 pesticide residues including 20 organophosphates (OP), 6 synthetic pyrethroids, 12 carbamates, 2 abamectins, imidacloprid, dithiocarbamates, and carbendazim. Out of the 412 vegetable samples, 235 (57%) had pesticide residues and 185 (45%) had pesticide residues that exceeded the maximum residue limits (MRLs). For the fruit samples, 245 (81%) of the 301 samples had pesticide residues and 165 (55%) had pesticide residues that exceeded the MRLs. The vegetable and the fruit samples had multiple synthetic pyrethroid residues and higher levels of residues than OP and other pesticides. Among the OP pesticides, chlorpyrifos was the most frequently detected pesticide. Residue detection in the rural samples was higher than that in the urban samples. The present study found very high numbers of samples to be above the MRLs: 45% of the vegetable samples and 55% of the fruit samples. Therefore, multi-residue methods are proposed as a regular monitoring system to ensure coverage of the multiple pesticides that are commonly used in agriculture and secure the national food safety policy.

Key words; Pesticide residues; Vegetables and fruits; MRL; Food safety

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Thailand is a tropical country located in Southeast Asia and cultivates several kinds of vegetables and fruits for both domestic consumption and export to places such as Europe (Skretteberg et al., 2015; Poulsen et al., 2017). Multiple pesticides have been reported to be used in vegetable and fruit cultivation in Thailand (Schreinemachers et al., 2011; Schreinemachers and Tipragsa, 2012; Riwthong et al., 2015) although several sectors have tried to promote reduction of pesticide use (Posri et al., 2006; Chalermphol et al., 2014). The agricultural area in the northern region holds approximately 4 million hectares, or about 22.1%, of the agricultural area of the country. The northern region was ranked the second after the northeastern region (45.4%). In addition, out of the total 5.6 million agricultural farm holdings in the country, the northern region had its farmers using pesticides the most (70%), and ranked second after the central region (73%) (National Statistical Office, 2003). Globally, most of the investigation on multiple pesticide use and residues have been conducted in food sources (Boobis et al., 2008; Prapamontol et al., 2010; Hjorth et al., 2011; Jardim and Caldas, 2012; Nougadère et al., 2012; Arias et al., 2014; Panseri et al., 2014; Szpyrka et al., 2015; Wanwimolruk et al., 2019), and some have been carried out in environmental samples (Sangchan et al., 2012; Pose-Juan et al., 2015). Biological assessment of exposure to pesticides has been reported from various populations in northern Thailand such as small-scale farmers (Panuwet et al., 2008; Hongsibsong et al., 2019), school children (Panuwet et al., 2009), farmers and consumers (Thiphom et al., 2014; Wongta et al., 2018), and nursing farmers (Naksen et al., 2014). Urinary pesticide metabolites found among small-scale farmers were metabolites of organophosphate (OP) pesticides, for example, dialkylphosphates (DAPs) (Hongsibsong et al., 2019); ethylene bisdithiocarbamates, e.g., ethylenethiourea (ETU); malathion; 2,4-D; and alachlor (Panuwet et al., 2008). Whereas the specific urinary metabolites detected in school children for example para-nitrophenol (PNP); 3,5,6-trichloro-2-pyridinol (TPCY, metaboliteof chlorpyrifos); and 3-phenoxybenzoicacid (3-PBA, a common metabolite of pyrethroids, PYR) (Hongsibsong et al., 2019; Panuwet et al., 2009). Recently, 3-phenoxybenzoic acid metabolite of PYRs was found in serum samples from farmers and consumers (Thiphom et al., 2014). Furthermore, organochlorine pesticides, persistent compounds, were frequently detected in serum and breast milk (Stuetz et al., 2001); OPs; non-persistent compounds, were also detected in maternal serum and breast milk samples (Naksen et al., 2014). These biological metabolites of the pesticides detected are evidence of the exposure to OP and PYR pesticides. Furthermore, there has been a dramatic increase in the import of pesticides into Thailand during 2002-2016 (Department of Agriculture, Office of Agricultural Economics, Thailand). It is appeared that herbicides, insecticides, and fungicides are among the major pesticides used in Thailand and that the total quantity of pesticides imported into Thailand increased about fourfold, from 2002 to 2016.

Due to the limitation of available analytical resources, data on pesticide residues in vegetables and fruits in Thailand are scarce. To our knowledge, there are no reports from Thailand of multiple pesticide residues in vegetables and fruits. Here we report the series' survey of multiple pesticide residues in 16 different vegetables (n = 412) and 11 different fruits (n = 301) collected from rural and urban markets from four provinces engaged in intensive agriculture in upper northern Thailand during 2007–2013.

# **MATERIALS AND METHODS**

#### Study sites

Vegetable and fruit samples were bought from rural and urban markets in four provinces engaged in intensive agriculture in upper northern Thailand, including Chiang Mai (18°50'14"N 98°58'14"E) (the site it was most conducted), Chiang Rai

(19°54'N 99°49'E), Lampang (18°18'N 99°30'E), and Phayao (19°9'55"N 99°54'13"E) provinces, as shown in Figure 1.



**Figure 1.** A map of four provinces including Chiang Mai, Chiang Rai, Lampang, and Phayao which were the study sites in this study in year 2010.

# Collecting and processing vegetable and fruit samples

Sixteen different vegetables and 11 different fruits (Table 1) that are commonly consumed were collected from urban and rural markets during 2007–2013.

| Table 1. Sixteen vegetables and | eleven fruits sampled from various | markets during 2007–2013. |
|---------------------------------|------------------------------------|---------------------------|
|                                 |                                    | · · · · · · · · · · · ·   |

|      |  | Common name (Thai name)       | Scientific name  |
|------|--|-------------------------------|--|
| Vege | etables  |                               |  |
| 1    | *  | Baby corn (Khoa Pod On)       | Zea mays   |
| 2    |  | Broccoli                      | Brassica oleracea  |
| 3    |  | Cabbage (Ka Lum)              | Brassica oleracea L. var. capitata L.                            |
| 4    |  | Cauliflower (Ka Lum Dok)      | Brassica oleracea L. var. botrytis L                             |
| 5    |  | Chili pepper (Prik Chi Fah)   | Capsicum annuum  |
| 6    |  | Chinese cabbage (Pak Kad Kow) | Brassica pekinensis  |
| 7    | The second secon | Pakchoi (Pak Kwang Tung)      | Brassica chinensis Jusl var parachinensis (Bailey)<br>Tsen & Lee |
| 8    | Ś  | Chinese kale (Ka Nah)         | Brassica alboglabra  |
| 9    | 0  | Cucumber (Tang Kwa)           | Cucumis sativus  |

#### Table 1. Continued

|     |  | Common name (Thai name)                        | Scientific name         |
|-----|--|--|-------------------------|
| Ve  | getables   |  |                         |
| 10  | 12   | Ginger (Khing)                                 | Zingiber officinale     |
| 11  | AL.  | Bird's chili, Thai chili (Prik Ki Noo)         | Capsicum frutescens     |
| 12  | 15   | Pepper (Prik Tai)                              | Piper nigrum            |
| 13  | DE E   | Sugar pea, Sweet pea, Garden pea (Tua Lan Tao) | Pisum sativum           |
| 14  |  | Tomato (Ma Khua Thet)                          | Lycopersicon esculentum |
| 15  |  | Water spinach, Water morning glory (Pak Bung)  | Ipomcea aquatica        |
| 16  |  | Yard long bean (Tua Fak Yao)                   | Vigna unguiculata       |
| Fru | ıits   |  |                         |
| 17  |  | Apple  | Malus domestica         |
| 18  |  | Asian pear (Sa Lhi)                            | Pyrus pyrifolia         |
| 19  |  | Dragon fruit (Keaw Mang Kon)                   | Hylocereus undatus      |
| 20  | 28   | Guava (Fa Rang)                                | Psidium guajava         |
| 21  |  | Grape (A Ngoon)                                | Vitis vinifera          |
| 22  | -  | Lychee (Lin Jee)                               | Litchi chinensis        |
| 23  | age of the second secon | Mango (Ma Muang)                               | Mangifera indica        |
| 24  | ٨  | Rose apple (Chom Phoo)                         | Syzygium samarangense   |
| 25  |  | Strawberry                                     | Fragaria ananassa       |
| 26  |  | Tangerine (Som Kheaw Wan)                      | Citrus Reticulata       |
| 27  |  | Watermelon (Tang Mo)                           | Citrullus lanatus       |

## **Sample collection**

During 2007–2013, Chiang Mai urban (local and supermarkets) and rural areas were major sites of vegetable and fruit sample collection for pesticide residue analysis in the present study. However, since Chiang Mai province was a major source of tangerine cultivation in the country, contamination of tangerine with pesticide residues

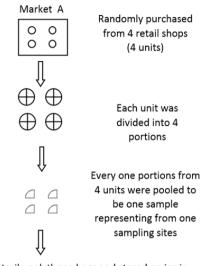
was a public health concern (Prapamontol et al, 2010). Therefore, in 2009, 100 fresh tangerine samples were collected from rural markets in Chiang Mai for multiple pesticide residue analysis. Besides the studies on Chiang Mai province, in 2010, the study sites were extended to three more provinces of upper northern Thailand practicing intensive agriculture. Therefore, in 2010, vegetable and fruit samples were collected from rural markets in Chiang Mai, Chiang Rai, Lampang, and Phayao provinces. Furthermore, in 2013, vegetables and some medicinal plants, that is, ginger and pepper, were analyzed for two groups of fungicides, namely carbendazim and dithiocarbamates. The details of the sample collection in each of the years, sites, pesticide residues, and number of samples are shown in Table 2.

The methods adopted for sampling the vegetables and the fruits were based on Codex recommended methods of sampling for the determination of pesticide residues for compliance with MRLs (CODEX, 1999).

For sampling vegetables and fruits from different markets, each type of vegetable and fruit was purchased from four or more retail shops (4 units) in each market. Approximately, 1 kg of each vegetable or fruit type was purchased from one retail shop. Each unit was divided into four portions. One portion was put in sterile polythene bags with label and stored on ice in a Styrofoam box during transportation to the toxicology laboratory of the Environment and Health Research Unit, Research Institute for Health Sciences, Chiang Mai University. All the labeled samples were stored at  $-20^{\circ}$ C until analysis. The diagram of the sampling protocol is presented in Figure 2. All samples were not washed and cleaned according to CODEX (1999) Edible parts of all the samples were chopped into small pieces and homogenized before the extraction procedure.

| Year                | 2007                                 | 2008                                 | 2009  | 2010  | 2013                              |
|---------------------|--------------------------------------|--------------------------------------|---|---|-----------------------------------|
| Sampling site       |                                      |                                      |   |   |                                   |
| -Areas              | Urban                                | Urban                                | Rural   | Rural   | Urban                             |
| -Provinces (n)      | Chiang Mai<br>(150)                  | Chiang Mai<br>(150)                  | Chiang Mai<br>(156)   | Chiang Mai (53)<br>Chiang Rai (39)<br>Lampang (41)<br>Phayao (38) | Chiang Mai (86)                   |
| Types (n)           | -Vegetables<br>(100)<br>-Fruits (50) | -Vegetables<br>(100)<br>-Fruits (50) | -Vegetables (35)<br>-Fruits (21)<br>-Tangerines (100)   | -Vegetables (91)<br>-Fruits (80)                                  | -Vegetables (86)                  |
| Analyzed pesticides | OP pesticides                        | OP pesticides                        | - OP pesticides<br>-Carbamates<br>-Synthetic<br>pyrethroids<br>-Carbendazim<br>-Imidacloprid<br>-Abamectins | OP pesticides   | -Carbendazim<br>-Dithiocarbamates |

 Table 2. Details of vegetable and fruit sample collection during 2007–2013.



Samples were kept in sterile polythene bags and stored on ice in Styrofoam box during transportation

# **Figure 2.** A sampling protocol for the different types of vegetables and fruits from one market.

## **Chemicals and standards**

All the organic solvents were of analytical grade. Acetone, acetonitrile, ethyl acetate, and toluene were purchased from J.T. Baker (Radner, Pennsylvania, USA). Sodium chloride, sodium sulfate, potassium dihydrogen phosphate, and dipotassium hydrogen phosphate were purchased from Merck (Darmstadt, Germany). The graphite carbon black (GCB) was purchased from Agilent Technologies (USA). The standard pesticides: 20 organophosphate (OP) pesticides, 10 carbamates, and 5 synthetic pyrthroids, and bifenthrin, carbendazim, macozeb, abamectin, and imidacloprid were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Triphenylphosphate was purchased from Fluka (Buchs, Germany).

#### Methods

The pesticides selected for the determination of the presence of pesticide residues in the samples were based on the data on pesticides imported into Thailand (OAE, 2013). An aliquot of sample was analyzed using internal standard and matrixmatched standard calibration curve. Quality assurance and control was performed using spiked matrix-matched aliquot (in duplicate) as a sample. Pesticide analysis methods for OPs (Polyiem et al., 2018), carbamates (Tuan et al., 2009), synthetic PYRs (Pakvilai abamectins (Diserens and Henzelin, 1999), carbendazim, et al., 2015), dithicarbamates, and imidacloprid were performed according to methods previously described. The methods employed in this study have limit of detection (LOD) and limit of quantitation (LOQ) below the MRLs set by Codex Alimentarius. In case Codex MRLs were not available, MRLs from Thailand (Thai Agricultural Commodity and Food Standard TACFS 9002-2008), (https://law.moj.gov.tw/ENG/), Taiwan Korea (http://www.foodsafetykorea.go.kr/residue/prd/), Japan (http://db.ffcr.or.jp/front/), and the EU (Draft EU MRLs in view of the first establishment of Annex II to Regulation (EC) N.396/2005) were adopted. Each batch of sample analysis had at least an aliquot of spiked pooled matrix added as the internal quality control sample.

**Organophosphate pesticides**. OP pesticides have been widely used in vegetable and fruit cultivation. The method used for detecting 20 OP pesticide residues was as described previously (Polyiem et al., 2018). Twenty OP pesticides analyzed were azinphos-ethyl, azinphos-methyl, chlorpyrifos, diazinon, dicrotophos, dimethoate, EPN,

ethion, fenitrothion, malathion, methamidophos, methidathion, mevinphos, monocrotophos, parathion-methyl, pirimiphos-ethyl, pirimiphos-methyl, profenofos, prothiophos, and triazophos. Briefly, 5 g of well-homogenized vegetable and fruit samples was extracted with 5 mL acetonitrile and cleaned using 500 mg/5 mL graphite carbon black. The ethyl acetate extract (1  $\mu$ L) was analyzed by gas chromatography–flame photometric detection (Hewlett Packard 6890 Series, Agilent Technologies, CA, USA).

**Cabamate pesticides.** The method used for detecting the 10 carbamate pesticides was adopted from the Taiwan Agricultural Chemicals and Toxic Substances Research Institute, Taichung, Taiwan (Tuan ed al., 2009). Ten carbamate pesticides assayed were 3-hydroxy carbofuran, aldicarb, aldicarb sulfone, aldicarb sulfoxide, Baygon, carbaryl, carbofuran, methiocarb, methomyl, and oxamyl. Briefly, 10 g of the homogenized sample was extracted with acetonitrile and then cleaned. The clean acetone extract was subjected to post-column derivatization with o-phthaladehyde (OPA) and analyzed using high performance liquid chromatography–fluorescence detection (Shimadzu SCL-10A VP, Shimadzu, Japan).

**Synthetic pyrethroids pesticides.** Since PYR pesticides have enatiometric isomers (Corcellas et al., 2015), the present study used the method reported by Pakvilai et al. (Pakvilai et al., 2015), and the summation of the area peaks from each PYR pesticide was employed for the calculation of the concentration. Six synthetic PYR pesticides including cyfluthrin, cypermethrin, deltamethrin, fenvalerate, lambda cyhalothrin, and permethrin were analyzed. In brief, 5 g of the homogenized sample was extracted with 10 mL of dichloromethane. The filtered extracts were cleaned using 250 mg graphitized carbon cartridge solid phase extraction. The clean extract was analyzed using gas chromatography–electron capture detection (Hewlett Packard 6890 Series, Agilent Technologies, CA, USA).

**Carbendazim.** Analysis of carbendazim in vegetable samples was carried out by employing the method reported by Phansawan et al. (2015). In brief, 5 g of the homogenized vegetable sample was extracted with ethyl acetate in acidic condition. The extract was cleaned by filtering through a SAX/PSA dual-layer cartridge. The clean extract was dissolved in HPLC mobile phase (methanol:water, 25:75, v/v) and analyzed using HPLC-variable UV detection (Agilent 1100, Agilent Technologies, CA, USA).

**Dithiocarbamates.** The method used for analysis of DTCs in vegetables such as cucumber, ginger, and pepper was a method reported by Phansawan et al. (2014). Briefly, 5 g of homogenized fresh vegetable samples was transferred into the extraction tube, followed by the addition of 4.0 mL of freshly prepared 0.5% SnCl<sub>2</sub> made in 5 M hydrochloric acid. Then, into each tube was added 1.0 mL of isooctane. The samples were heated in an ultrasonic water bath at 80°C for 40 min with continuous stirring and then allowed to cool down to room temperature (25°C). During cooling, the tubes were periodically shaken by hand to assist with the CS<sub>2</sub> sipping into the isooctane layer, and 1  $\mu$ L of the isooctane layer was injected into a gas chromatography–flame photometric detector (Hewlett Packard 6890 Series, Agilent Technologies, CA, USA).

**Imidacloprid.** The analysis of imidacloprid was carried out by using a method modified from the method reported previously (Ferrer and Thurman, 2007). In brief, 10 g of the homogenized sample was extracted by using acetonitrile (1% acetic acid), and the extract was analyzed using HPLC-UV detection (Agilent 1100, Agilent Technologies, CA, USA).

**Abamectins.** Analysis of abamectins in fruits was performed using the method reported by Discerns and Henzelin with slight modifications (Diserens and Henzelin, 1999). In brief, 10 g of the homogenized sample was extracted using methanol and cleaned by solid phase extraction (Vertipak C18 SPE, Vertical, Bangkok, Thailand). The extract was derivatized with reagent 1 (acetonitrile and trifluoroactetic acid, 2:1) and reagent 2 (acetonitrile and 1-methylimidazole, 1:1), and analyzed by HPLC-Fluorescence detection (Agilent 1100, Agilent Technologies, CA, USA).

# RESULTS

This study reported pesticide residues in vegetables and fruits from four provinces in northern Thailand during 2007–2013. A total of 713 vegetable and fruit samples were analyzed for 43 multi-classes of pesticides including insecticides (20 OPs, 10 carbamates, 6 PYRs, 2 abamectins, and imidacloprid), and fungicides (carbendazim and dithiocarbamate). However, organochlorine pesticides were not included in this study because they have been banned in agriculture by the Thai government since 1983 (IPM, 2015).

## Multi-pesticide residues found in vegetable and fruit samples

Table 3 summarizes the incidence of pesticide residues found in vegetables and fruits from rural and urban markets. Individual pesticide residues detected and their concentrations (mg/kg) in vegetables and fruits can be found in Supplement A.

 Table 3. Summary of pesticide residues found in vegetables and fruits from rural and urban markets in upper northern Thailand during 2007-2013.

| Types      | Number of pesticide<br>residues detected | Urban markets | Rural markets |
|------------|--|---------------|---------------|
| Vegetables | Single & multiple                        | 42.6%         | 63.5%         |
|            | Multiple                                 | 16.8%         | 29.4%         |
|            |  | N = 286       | N = 126       |
| Fruits     | Single & multiple                        | 74%           | 83.3%         |
|            | Multiple                                 | 38%           | 67.2%         |
|            |  | N = 100       | N = 201       |

It was appeared that vegetable and fruit samples bought from rural markets had pesticide residues with higher frequency of detection, in both 'single & multiple' and 'multiple', than vegetable and fruit samples from urban markets.

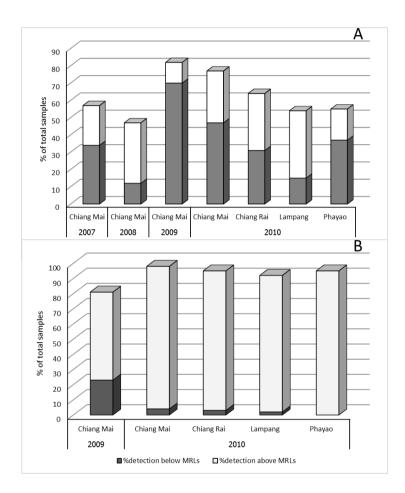
The fruit samples were found to have more residue-detected samples than the vegetable samples. The vegetable and the fruit samples with the highest percentages of contamination were, respectively, Chinese kale (74%) and tangerine (98%). There were 21 different pesticides detected in the tangerine samples. Baby corn was the only type of sample in this study which was not found to have pesticide contamination.

#### Frequently detected pesticides

All the pesticides and their percentages of detection are shown in Supplement B. Among the 20 OPs analyzed in the vegetable samples, chlorpyrifos was the most-detected OP. Twenty percent of the samples from urban markets and 18.9% of the samples from rural markets had chlorpyrifos levels that exceeded the MRL. Among 10 carbamates, aldicarb and carbaryl were found in the fruits (tangerines), and exceeded the MRLs by 1% and 2%, respectively. Synthetic PYRs were the most frequently detected residue in the both vegetables and fruits samples from rural markets and all six compounds exceeded the MRLs by values between 2.9% and 65.5%. Abamectin was found only in the tangerine samples, and only abamectin 1a compound was found to exceed the MRL in 13% of the analyzed samples. Cabendazim and imidacloprid were also found in the tangerine samples, but their contents did not exceed the MRL. However, in the vegetable samples from urban markets, the carbendazim residues detected were found to exceed the MRL by 15.6%. Dithiocarbamate fungicide was also detected, and found to exceed the MRL by 5.6%. In summary, among the 43 compounds of pesticides investigated in the present report, 27 (62.8%) were detected in the vegetable and fruit samples. According to the present report, the top 10 pesticide residues that exceeded the MRL show as following: (1) lambda-cyhalothrin (65.5%), (2) deltamethrin (56.7%), (3) cyfluthrin (34.5%), (4) fenvalerate (29.8%), (5) cypermethrin (21.6%), (6) chlorpyrifos (19.6%), (7) profenophos (5.4%), (8) methamidophos (5.2%), (9) ethion (4.8%), and (10) permethrin (2.9%).

## Comparison of pesticide residues found in four provinces

In 2010, 171 vegetable and fruit samples from 4 provinces including Chiang Mai, Chiang Rai, Lampang, and Phayao were analyzed for multi-pesticide residues. The number of OP pesticide-contaminated samples sourced from Chiang Mai was the highest (Figure 3A). At the same time, the numbers of pyrethriod pesticide-contaminated samples obtained from the four provinces were not markedly different (Figure 3B).



**Figure 3.** Percentages of OP pesticide (A) and pyrethroid (B) residue-detected samples (both below and above MRLs) in Chiang Mai Province during 2007–2010, and the percentages in the four provinces in 2010.

# DISCUSSION

The present report shows that multiple pesticide residues were detected in vegetable and fruit samples with high frequency. The frequencies of pesticide residues detected were higher in samples purchased from rural markets than in samples from urban markets. Similar reports showed higher pesticide residue concentrations in vegetables collected from the farms than bought from the markets and supermarkets (Sapbamrer and Hongsibsong, 2014; Kumari and John, 2019). This phenomenon might be explained from recent or just after cultivated produce as most of common consume

vegetable and fruits growing in the rural areas. Furthermore, lower residue detection in vegetables sold urban markets and supermarkets may be explained by pre-processing step which include washing and removal of unwanted parts. Overall, pesticide residues were more frequently detected in fruits than in vegetables (Table 3). In addition, among samples of vegetables (n = 412) and fruits (n = 301), 45% of vegetable and 55% of fruit samples found pesticide residues exceeded their MRLs (Table 4). Among 43 pesticides analyzed, residues of pyrethroid pesticides detected in samples from four provinces in 2010 almost all exceeded the MRLs (Figure 3B) compared with residues of OP pesticides (Figure 3A). Chlorpyrifos, an OP pesticide, was the most detected OP in the present report and among the top 10 in the list of active ingredients imported into Thailand during 2007-2014 (OAE, 2013). Chlorpyrifos is a common OP pesticide detected in vegetables and fruits in Thailand. It has been reported to be found in vegetables from markets around Kwan Phayao Lake, Phayao Province, upper northern Thailand (Sapbamrer and Hongsibsong, 2014), Chinese kale (purchased from markets in Nakhon Pathom Province, central Thailand (Wanwimolruk et al., 2015), and mangosteen bought from markets in Bangkok, central and eastern provinces of Thailand (Phopin et al., 2017) were also found to contain chlorpyrifos residues.

MRL is the maximum concentration of a pesticide residue (expressed as mg/kg), recommended by the Codex Alimentarius Commission to be legally permitted in or in food commodities and animal feeds. MRLs are based on good agricultural practices (GAP) data and foods derived from commodities that comply with the respective MRLs and are intended to be toxicologically acceptable (<u>http://www.fao.org/waicent/faostat/</u><u>Pest-Residue/pest-e.htm</u>). In order to protect consumers' health, residues of pesticides in any foods must not above or exceed their MRLs.

This report is derived from survey series of pesticide residues for 5 years in 2007, 2008, 2009, 2010 and 2013. To our best knowledge, this is the longest survey duration ever reported from Thailand. Furthermore, this study covered several different pesticide groups used in Thailand namely organophosphates, carbamates, synthetic pyrethroids, carbendazim, dithiocarbamates, imidaclopid, abamectins which constitute 43 pesticides in total. This report also covered vegetables and fruits that common consume in northern as well as other parts of Thailand. Zaidon et al. (2016) reported a review on pesticides occurrence in fruits and vegetables in Malaysia from 2007 to 2014 covered 10 pesticides and 30% of pesticide residues detected exceeded their MRLs.) Poulsen et al. (2017) also reported the results from the Danish monitoring programme for pesticide residues from 2004-2011.

This survey was conducted in the academic institution with some limitation of funding for some time over a long period of time (2007-2013). In order to protect consumers' health, national institution (s) would be able to secure funding over years' plan such as Thai Ministry of Public Health by regular monitoring of pesticide residues in foods and Ministry of Agriculture and Cooperatives by enforcement monitoring of good agricultural practices (GAP).

# CONCLUSION

A total of 713 samples of vegetables and fruits collected from urban and rural markets in northern Thailand during 2007–2013 were analyzed for 43 pesticide residues. This study found that 67% of the vegetable and fruit samples had pesticide residues and 43% had residues that exceeded the MRLs set by Codex and others, namely the EU, Japan, Taiwan, Korea, and Thailand.

The fruit samples were found to have percentage of pesticide detection higher than the vegetable samples. Of the 301 fruit samples, 81% contained pesticide residues and 55% exceeded the MRL while in the 412 vegetable samples, 57% had pesticide residues and 45% exceeded the MRL. The samples collected from rural markets had percentage detection of pesticide residues higher than the samples collected from urban markets. The top 10 of the pesticide residues detected in the present report include 6 synthetic pyrethroids and 4 organophosphate pesticides. Lambda-cyhalothrin was the most frequently detected synthetic pyrethroid and chlorpyrifos was the most frequently detected organophosphate pesticide.

**Table 4.** Summary of pesticide residue detection in vegetable and fruit samples from urban and rural markets in upper northern Thailand during 2007-2013.

| Site               | Total samples           | Detection in<br>sample<br>(% of total<br>samples) | Exceeding<br>MRL<br>(% of total<br>samples) | Type of<br>pesticide | No. of<br>analyzed<br>samples | Detection in<br>sample<br>(% of<br>analyzed<br>samples) | Exceeding<br>MRL<br>(% of<br>analyzed<br>samples) | Detected pesticides  |
|--------------------|-------------------------|---|---|----------------------|-------------------------------|---|---|--|
| All                | Total (N = 713)         | 480 (67%)   | 308 (43%)                                   |                      |                               |   |   |  |
|                    | Vegetables<br>(n = 412) | 235 (57%)   | 185 (45%)                                   |                      |                               |   |   |  |
|                    | Fruits $(n = 301)$      | 245 (81%)   | 165 (55%)                                   |                      |                               |   |   |  |
| Urban<br>(n = 386) | Total (n = 386)         | 199 (52%)   | 96 (25%)                                    |                      |                               |   |   |  |
|                    | Vegetables<br>(n = 286) | 125 (44%)   | 58 (20%)                                    | OP pesticides        | 200                           | 94 (47%)  | 50 (25%)  | azinophos-methyl, chlorpyrifos, diazinon,<br>dicrotophos, dimethoate, EPN, ethion, malathion,<br>methamidophos, mevinphos, monocrotophos,<br>parathion-methyl, pirimiphos-ethyl, pirimiphos-<br>methyl, profenophos, triazophos (n = 16) |
|                    |                         |   |   | carbamates           | Not analyzed                  |   |   | -  |
|                    |                         |   |   | pyrethroids          | Not analyzed                  |   |   | -  |
|                    |                         |   |   | others               | 86                            | 31 (36%)  | 8 (9%)  | carbendazim, mancozeb (n=2)  |
|                    | Fruits (n = 100)        | 74 (74%)  | 38 (38%)                                    | OP pesticides        | 100                           | 74 (74%)  | 38 (38%)  | chlorpyrifos, diazinon, dimethoate, ethion,<br>fenitrothion, malathion, methamidophos,<br>methidathion, monocrotophos, parathion-methyl,<br>pirimiphos-ethyl, pirimiphos-methyl, profenophos,<br>prothiophos, triazophos (n = 15)        |
|                    |                         |   |   | carbamates           | Not analyzed                  |   |   | -  |
|                    |                         |   |   | pyrethroids          | Not analyzed                  |   |   | -  |
|                    |                         |   |   | others               | Not analyzed                  |   |   | -  |

Table 4. Continued.

| Site          | Total samples           | Detection in<br>sample<br>(% of total<br>samples) | Exceeding<br>MRL<br>(% of total<br>samples) | Type of<br>pesticide | No. of<br>analyzed<br>samples | Detection in<br>sample<br>(% of<br>analyzed<br>samples) | Exceeding<br>MRL<br>(% of<br>analyzed<br>samples) | Detected pesticides  |
|---------------|-------------------------|---|---|----------------------|-------------------------------|---|---|--|
| Rural (n=327) | Total (n = 327)         | 281 (86%)   | 212 (65%)                                   | ·                    |                               |   |   |  |
|               | Vegetables<br>(n = 126) | 110 (87%)   | 85 (67%)                                    | OP pesticides        | 126                           | 110 (87%)   | 29 (26%)  | chlorpyrifos, diazinon, dicrotophos, dimethoate,<br>parathion-methyl, ethion, malathion,<br>methamidophos, monocrotophos, parathion-<br>methyl, profenophos, triazophos (n = 12) |
|               |                         |   |   | carbamates           | Not analyzed                  |   |   | -  |
|               |                         |   |   | pyrethroids          | 91                            | 85 (93%)  | 85 (93%)  | cyfluthrin, cypermethrin, deltamethrin, fenvalerate,<br>lamda-cyhalothrin, permethrin (n = 6)  |
|               |                         |   |   | others               | Not analyzed                  |   |   | -  |
|               | Fruits ( n = 201)       | 171 (85%)   | 127(63%)                                    | OP pesticides        | 201                           | 171 (85%)   | 51 (25%)  | chlorpyrifos, diazinon, dicrotophos, dimethoate,<br>ethion, malathion, methamidophos,<br>monocrotophos, profenophos, prothiophos,<br>triazophos (n = 11)                         |
|               |                         |   |   | carbamates           | 100                           | 1 (1%)  | 1 (1%)  | abamectin 1a, aldicarb, carbaryl,  |
|               |                         |   |   | pyrethroids          | 180                           | 159 (88%)   | 127 (70%)   | cyfluthrin, cypermethrin, deltamethrin, fenvalerate,<br>lamda-cyhalothrin (n = 6)  |
|               |                         |   |   | others               | 100                           | 87 (87%)  | 4 (4%)  | carbendazim, imidacloprid  |

This study result may add to the existing evidence of pesticide residues which could be able to estimate adverse effect on public health for policy maker. Finally, multi-residue methods are proposed as a regular monitoring system to enhance food safety policy both internal consumption and exportation.

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| Samples      |    |                                   |   | Urban   |                                      |    |                                   |   | Rural   |   |
|--------------|----|-----------------------------------|---|---|--------------------------------------|----|-----------------------------------|---|---|---|
|              | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected multi-<br>residues | Detected<br>pesticides;<br>range or mean <sup>b</sup><br>(mg/kg)  | No. exceeding<br>MRL                 | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected multi-<br>residues | Detected OP<br>pesticides;<br>range or mean <sup>b</sup><br>(mg/kg)   | No. exceeding<br>MRL                              |
| Baby Corn    | 20 | 0                                 | 0   | none  | 0                                    | -  | -                                 | -   | -   | -   |
| Broccoli     | 20 | 12                                | 4   | chlorpyrifos (0.005–0.030)<br>dicrotophos (0.014)<br>diazinon (0.014)<br>ethion (0.002)<br>mevinphos (0.030–0.011)<br>pirimiphos-ethyl (0.008–0.018)<br>pirimiphos-methyl (0.005) | 2<br>0<br>0<br>0<br>0<br>0<br>0<br>0 | -  | -                                 | -   | -   | -   |
| Cabbage      | 20 | 5                                 | 3   | diazinon (0.009–0.402)<br>azinophos-methyl (0.088)  | 0<br>0                               | 24 | 11                                | 3   | chlorpyrifos (0.002–0.020)<br>dicrotophos (0.010)<br>ethion (0.001)<br>methamidophos (0.015–0.017)<br>monocrotophos (0.044)<br>cyfluthrin (0.047–0.656)<br>cypermethrin (0.028–0.321)<br>deltamethrin (0.079–4.854)<br>fenvalerate (0.131–2.374)<br>lamda-cyhalothrin (0.063–0.762)<br>permethrin (0.002–0.063) | 1<br>0<br>2<br>0<br>9<br>2<br>13<br>12<br>14<br>0 |
| Cauliflower  | 20 | 10                                | 8   | chlorpyrifos (0.002–0.232)<br>dimethoate (0.004)<br>ethion (0.001)<br>methamidophos (0.005–0.639)<br>pirimiphos-methyl (0.013)  | 7<br>0<br>0<br>4<br>0                | 9  | 3                                 | 0   | chlorpyrifos (0.001–0.016)<br>dicrotophos (0.027)   | 1<br>0  |
|              | 4  | 3                                 | NA  | carbendazim (0.013–0.020)   | 1                                    |    |                                   |   | cyfluthrin (0.028–0.276)<br>cypermethrin (0.031–0.402)<br>deltamethrin (0.436–2.002)<br>fenvalerate (0.129–0.867)<br>lamda-cyhalothrin (0.185–0.757)<br>permethrin (0.074–0.261)  | 6<br>1<br>6<br>3<br>8                             |
| Chili pepper | 4  | 3                                 | NA  | carbendazim (0.038-0.040)   | 0                                    | -  | -                                 | -   | -   | -   |

**Supplement A.** Pesticide residues detected in vegetable and fruit samples in upper northern Thailand during 2007–2013.

| Samples            |    |                                   |  | Urban   |                            |    |                                   |  | Rural  |   |
|--------------------|----|-----------------------------------|--|---|----------------------------|----|-----------------------------------|--|--|---|
|                    | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)   | No. exceeding<br>MRL       | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected OP<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)   | No. exceeding<br>MRL  |
| Chinese<br>cabbage | 20 | 7                                 | 2  | chlorpyrifos (0.009–0.043)<br>diazinon (0.001)<br>pirimiphos-methyl (0.011–0.034)<br>profenophos (0.025)  | 5<br>0<br>0<br>0           | 15 | 12                                | 9  | chlorpyrifos (0.001–1.596)<br>dicrotophos (0.001–0.139)<br>diazinon (0.001–0.002)<br>dimethoate (0.002–0.159)<br>parathion-methyl (0.002)<br>profenophos (0.001–0.055)<br>triazophos (0.001)<br>cyfluthrin (0.077–0.544)<br>cypermethrin (0.051–7.728)<br>deltamethrin (0.298–1.052)<br>fenvalerate (0.131–0.547)<br>lamda-cyhalothrin 0.167–0.862<br>permethrin (0.035–0.160)           | 3<br>0<br>0<br>0<br>0<br>1<br>0<br>5<br>4<br>6<br>4<br>8<br>0 |
| Pakchoi            | -  | -                                 | -  | -   | -                          | 14 | 13                                | 10   | chlorpyrifos (0.001–1.596)<br>dicrotophos (0.001–0.775)<br>methamidophos (0.001–0.012)<br>monocrotophos (0.001–0.313)<br>parathion-methyl (0.020)<br>profenophos (0.002–0.256)<br>triazophos (0.006)<br>cyfluthrin (0.060–0.525)<br>cypermethrin (0.041–8.389)<br>deltamethrin (0.404–4.753)<br>fenvalerate (0.165–0.743)<br>lamda-cyhalothrin (0.103–1.072)<br>permethrin (0.007–0.120) | 4<br>1<br>2<br>0<br>1<br>0<br>4<br>3<br>9<br>5<br>10<br>0     |
| Chinese kale       | 20 | 13                                | 11   | chlorpyrifos (0.007–1.394)<br>diazinon (0.144)<br>dicrotophos (0.003–0.188)<br>ethion (0.001–0.020)<br>monocrotophos (0.003)<br>profenophos (0.013–0.130) | 1<br>9<br>1<br>1<br>0<br>0 | 18 | 15                                | 9  | chlorpyrifos (0.002–0.120)<br>dicrotophos (0.020–0.973)<br>ethion (0.001)<br>malathion (0.002)<br>methamidophos (0.002)<br>profenophos (0.160–0.251)   | 3<br>2<br>0<br>0<br>0<br>2                                    |

| Samples      |    |                                   |   | Urban   |                       |    |                                   |   | Rural  |                            |
|--------------|----|-----------------------------------|---|---|-----------------------|----|-----------------------------------|---|--|----------------------------|
|              | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected multi-<br>residues | Detected<br>pesticides;<br>range or mean <sup>b</sup><br>(mg/kg)  | No. exceeding<br>MRL  | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected multi-<br>residues | Detected OP<br>pesticides;<br>range or mean <sup>b</sup><br>(mg/kg)  | No. exceeding<br>MRL       |
| Chinese kale | 4  | 2                                 | NA  | carbendazim (0.034–0.119)   | 0                     |    |                                   |   | cyfluthrin (0.014–0.470)<br>cypermethrin (0.044–0.594)<br>deltamethrin (0.319–9.972)<br>fenvalerate (0.028–2.583)<br>lamda-cyhalothrin (0.038–0.472)<br>permethrin (0.015–0.210) | 6<br>4<br>7<br>6<br>5<br>0 |
| Cucumber     | 20 | 13                                | 6   | chlorpyrifos (0.002–0.106)<br>dimethoate (0.010)<br>malathion (0.006)<br>pirimiphos-ethyl (0.006–0.022)<br>pirimiphos-methyl (0.006–0.007)<br>profenophos (0.093) | 6<br>0<br>0<br>0<br>1 | 19 | 12                                | 2   | chlorpyrifos (0.003–0.116)<br>dicrotophos (0.010–0.034)<br>ethion (0.001)  | 4<br>0<br>0                |
|              | 24 | 5                                 | 0   | carbendazim (0.013–0.020)<br>mancozeb (0.12–0.27)   | 0                     |    |                                   |   | cyfluthrin (0.096–0.619)<br>cypermethrin (0.078–0.228)<br>deltamethrin (0.434–0.681)<br>fenvalerate (0.092–0.553)<br>lamda-cyhalothrin (0.211–0.976)<br>permethrin (0.021–0.130) | 6<br>1<br>4<br>2<br>4<br>0 |
| Ginger       | 24 | 2                                 | NA  | carbendazim (0.009–0.011)   | 0                     | -  | -                                 | -   | -  | -                          |
| Bird's chili | 4  | 3                                 | NA  | carbendazim (0.011–0.081)   | 1                     | -  | -                                 | -   | -  | -                          |
| Pepper       | 14 | 7                                 | NA  | mancozeb (0.14-30.6)  | 3                     | -  | -                                 | -   | -  | -                          |
| Sweet pea    | 20 | 10                                | 3   | chlorpyrifos (0.005–0.649)<br>dimethoate (0.002–0.024)<br>methamidophos (0.015–0.032)<br>pirimiphos-methyl (0.005–0.006)  | 4<br>0<br>3<br>0      | 3  | 2                                 | 0   | ethion (0.001)   | 0                          |
| Tomato       | 4  | 3                                 | NA  | carbendazim (0.014–0.064)   | 1                     |    |                                   |   |  |                            |

| Samples       |    |                                   |  | Urban   |                      |    |                                   |  | Rural  |                      |
|---------------|----|-----------------------------------|--|---|----------------------|----|-----------------------------------|--|--|----------------------|
|               | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg) | No. exceeding<br>MRL | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected OP<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg) | No. exceeding<br>MRL |
| Water         | 20 | 9                                 | 6  | chlorpyrifos (0.002-0.003)  | 0                    | 8  | 4                                 | 0  | chlorpyrifos (0.002-0.103)   | 2                    |
| morning glory |    |                                   |  | dicrotophos (0.004-0.403)   | 2                    |    |                                   |  | dicrotophos (0.072)  | 0                    |
|               |    |                                   |  | ethion (0.001–0.002)  | 0                    |    |                                   |  |  |                      |
|               |    |                                   |  | monocrotophos (0.030–0.089)   | 2                    |    |                                   |  |  |                      |
|               |    |                                   |  | dimethoate (0.013–0.019)<br>pirimiphos-methyl (0.025)               | 0<br>0               |    |                                   |  |  |                      |
|               |    |                                   |  | pirimphos-methyr (0.023)  | 0                    |    |                                   |  | cyfluthrin (0.055–0.429)   | 2                    |
|               |    |                                   |  |   |                      |    |                                   |  | cypermethrin (0.073–1.182)   | 4                    |
|               |    |                                   |  |   |                      |    |                                   |  | deltamethrin (0.424–1.030)   | 4                    |
|               |    |                                   |  |   |                      |    |                                   |  | fenvalerate (0.046–0.906)  | 2                    |
|               |    |                                   |  |   |                      |    |                                   |  | lamda-cyhalothrin (0.150–1.054)  | 5                    |
|               |    |                                   |  |   |                      |    |                                   |  | permethrin (0.017-0.248)   | 0                    |
| Yard long     | 20 | 12                                | 5  | chlorpyrifos (0.002-0.033)  | 3                    | 16 | 8                                 | 4  | chlorpyrifos (0.003-0.018)   | 1                    |
| bean          |    |                                   |  | dicrotophos (0.104)   | 0                    |    |                                   |  | dicrotophos (0.001-0.060)  | 0                    |
|               |    |                                   |  | dimethoate (0.004–0.082)  | 1                    |    |                                   |  | ethion (0.001)   | 0                    |
|               |    |                                   |  | EPN (0.004-0.616)   | 4                    |    |                                   |  | profenophos (0.014)  | 0                    |
|               |    |                                   |  | malathion (0.002)   | 0                    |    |                                   |  |  |                      |
|               |    |                                   |  | parathion-methyl (0.002)  | 0                    |    |                                   |  |  |                      |
|               |    |                                   |  | profenophos (0.024–0.025)   | 0                    |    |                                   |  |  |                      |
|               | 4  | 2                                 | ΝΑ   | triazophos (0.043)  | 0                    |    |                                   |  | $\alpha$   | C                    |
|               | 4  | 3                                 | NA   | carbendazim (0.008-0.334)   | 1                    |    |                                   |  | cyfluthrin $(0.096-0.619)$   | 6                    |
|               |    |                                   |  |   |                      |    |                                   |  | cypermethrin (0.078–0.228)<br>deltamethrin (0.434–0.681)               | 1<br>4               |
|               |    |                                   |  |   |                      |    |                                   |  | fenvalerate (0.092–0.553)  | 4<br>2               |
|               |    |                                   |  |   |                      |    |                                   |  | lamda-cyhalothrin (0.211–0.976)  | 4                    |
|               |    |                                   |  |   |                      |    |                                   |  | permethrin (0.021–0.130)   | 4<br>0               |

| Samples |     |                                   |  | Urban   |                      |     |                                   |  | Rural  |                                  |
|---------|-----|-----------------------------------|--|---|----------------------|-----|-----------------------------------|--|--|----------------------------------|
|         | Z   | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)   | No. exceeding<br>MRL | z   | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected OP<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)   | No. exceeding<br>MRL             |
| Fruits  | 100 | 74<br>(74%)                       | 38   | chlorpyrifos<br>diazinon<br>dimethoate<br>ethion<br>fenitrothion<br>malathion<br>methamidophos<br>methidathion<br>monocrotophos<br>parathion-methyl<br>pirimiphos-methyl<br>pirimiphos-methyl<br>profenophos<br>prothiophos<br>triazophos |                      | 201 | 170<br>(84%)                      | 135  | aldicarb<br>carbaryl<br>chlorpyrifos<br>cyfluthrin<br>cypermethrin<br>deltamethrin<br>diazinon<br>dicrotophos<br>dimethoate<br>ethion<br>fenvalerate<br>imidacloprid<br>abamectin 1a<br>lamda-cyhalothrin<br>malathion<br>methamidophos<br>monocrotophos<br>permethrin<br>profenophos<br>prothiophos<br>triazophos |                                  |
| Apple   | 20  | 12                                | 1  | chlorpyrifos (0.003-0.161)<br>ethion (0.001)<br>fenitrothion (0.004-0.007)  | 1<br>0<br>0          | 19  | 7                                 | 5  | chlorpyrifos (0.001–0.018)<br>ethion (0.001)<br>cyfluthrin (0.041–0.147)<br>cypermethrin (0.010–0.290)<br>deltamethrin (0.008–1.177)<br>fenvalerate (0.014–0.196)<br>lamda-cyhalothrin (0.024–0.385)<br>permethrin (0.002–2.271)   | 2<br>0<br>1<br>8<br>0<br>10<br>5 |

| Samples      |    |                                   |  | Urban  | Rural  |    |                                   |  |  |  |  |  |  |  |
|--------------|----|-----------------------------------|--|--|--|----|-----------------------------------|--|--|--|--|--|--|--|
|              | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)  | No. exceeding<br>MRL                           | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected OP<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)   | No. exceeding<br>MRL                                     |  |  |  |  |
| Asian pear   | -  | -                                 | -  | -  | -  | 12 | 11                                | 2  | chlorpyrifos (0.001–0.038)<br>dicrotophos (0.001)<br>ethion (0.001–0.003)<br>cyfluthrin (0.013–0.513)<br>cypermethrin (0.008–0.151)<br>deltamethrin (0.023–0.397)<br>fenvalerate (0.050–0.765)<br>lamda-cyhalothrin (0.024–0.395)<br>permethrin (0.011–0.340)  | 7<br>0<br>3<br>0<br>7<br>5<br>8<br>0                     |  |  |  |  |
| Dragon fruit | 10 | 3                                 | 0  | fenitrothion (0.004-0.011)   | 0  | 5  | 2                                 | 0  | ethion (0.001–0.002)   | 0  |  |  |  |  |
| Guava        | 20 | 20                                | 8  | chlorpyrifos (0.003-0.057)<br>dimethoate (0.003-0.005)<br>ethion (0.002-0.092)<br>malathion (0.001-0.010)<br>parathion-methyl (0.002-0.004)<br>profenophos (0.156)<br>prothiophos (0.007-0.0303)   | 8<br>0<br>0<br>1<br>0                          | 12 | 9                                 | 5  | chlorpyrifos (0.002–0.027)<br>diazinon (0.003)<br>dicrotophos (0.001)<br>ethion (0.001–0.224)<br>methamidophos (0.006–0.008)<br>prothiophos (0.005)<br>cyfluthrin (0.011–2.79)<br>cypermethrin (0.021–0.642)<br>deltamethrin (0.105–0.540)<br>fenvalerate (0.027–0.272)<br>lamda-cyhalothrin (0.015–0.324)<br>permethrin (0.015–0.492) | 1<br>0<br>0<br>1<br>0<br>0<br>4<br>2<br>6<br>1<br>8<br>0 |  |  |  |  |
| Grape        | 10 | 10                                | 8  | chlorpyrifos (0.003–0.038)<br>dimethoate(0.002–0.120)<br>ethion (0.001)<br>malathion (0.005)<br>methamidophos (0.025–0.030)<br>methidathion (0.002)<br>monocrotophos (0.008)<br>pirimiphos-ethyl (0.001)<br>pirimiphos-methyl (0.012)<br>prothiophos (0.005–0.373) | 2<br>3<br>0<br>2<br>0<br>0<br>0<br>0<br>0<br>2 | 2  | 2                                 | 2  | chlorpyrifos (0.001–0.006)<br>ethion (0.003)<br>monocrotophos (0.001)  | 0<br>0<br>0  |  |  |  |  |

| Samples    |    |                                   |   | Urban   | Rural                |    |                                   |   |  |                      |  |  |  |  |
|------------|----|-----------------------------------|---|---|----------------------|----|-----------------------------------|---|--|----------------------|--|--|--|--|
|            | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected multi-<br>residues | Detected<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg) | No. exceeding<br>MRL | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected multi-<br>residues | Detected OP<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg) | No. exceeding<br>MRL |  |  |  |  |
| Lychee     | -  | -                                 | -   | -   | -                    | 5  | 5                                 | 0   | chlorpyrifos (0.001-0.002)   | 0                    |  |  |  |  |
| -          |    |                                   |   |   |                      |    |                                   |   | ethion (0.001-0.498)   | 1                    |  |  |  |  |
| lango      | -  | -                                 | -   | -   | -                    | 14 | 6                                 | 1   | chlorpyrifos (0.001–0.022)   | 2                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | ethion (0.001)   | 0                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | triazophos (0.001)   | 0                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | cyfluthrin (0.010–0.700)   | 3                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | cypermethrin (0.006–0.155)   | 0                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | deltamethrin (0.030–0.382)   | 6                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | fenvalerate (0.003–1.024)  | 4                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | lamda-cyhalothrin (0.030–0.186)  | 6                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | permethrin (0.007-0.491)   | 0                    |  |  |  |  |
| Rose apple | -  | -                                 | -   | -   | -                    | 10 | 6                                 | 4   | diazinon (0.001)   | 0                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | dicrotophos (0.001)  | 0                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | dimethoate (0.005-0.007)   | 0                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | ethion (0.001–0.096)   | 0                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | cyfluthrin (0.020–0.131)   | 1                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | cypermethrin (0.051–1.595)   | 3                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | deltamethrin (0.020-2.739)   | 6                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | fenvalerate (0.050–3.744)  | 3                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | lamda-cyhalothrin (0.043–0.325)  | 6                    |  |  |  |  |
|            |    |                                   |   |   |                      |    |                                   |   | permethrin (0.019-0.200)   | 0                    |  |  |  |  |
| Strawberry | 10 | 9                                 | 2   | chlorpyrifos (0.003)  | 0                    | -  | -                                 | -   | -  | -                    |  |  |  |  |
|            |    |                                   |   | diazinon (0.001)  | 0                    |    |                                   |   |  |                      |  |  |  |  |
|            |    |                                   |   | ethion (0.002–0.467)  | 1                    |    |                                   |   |  |                      |  |  |  |  |
|            |    |                                   |   | malathion (0.002)   | 0                    |    |                                   |   |  |                      |  |  |  |  |
|            |    |                                   |   | traizophos (0.001-0.490)  | 1                    |    |                                   |   |  |                      |  |  |  |  |

| Samples    |    |                                   |  | Urban  |   | Rural |                                   |  |  |   |  |  |  |  |  |
|------------|----|-----------------------------------|--|--|---|-------|-----------------------------------|--|--|---|--|--|--|--|--|
|            | z  | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)  | No. exceeding<br>MRL                      | z     | No. of<br>contaminated<br>samples | No. of samples<br>detected<br>multi-residues | Detected OP<br>pesticides;<br>range or<br>mean <sup>b</sup><br>(mg/kg)   | No. exceeding<br>MRL  |  |  |  |  |  |
| Tangerine  | 20 | 19                                | 19   | chlorpyrifos (0.003-0.315)<br>diazinon (0.001)<br>dimethoate (0.017-0.131)<br>ethion (0.002-0.467)<br>malathion (0.002-0.068)<br>methamidophos (0.023-0.069)<br>pirimiphos-methyl (0.005-0.019)<br>profenophos (0.008-0.303)<br>triazophos (0.001-0.490) | 9<br>0<br>1<br>1<br>1<br>8<br>0<br>0<br>1 | 22    | 22                                | 20   | chlorpyrifos (0.002–0.150)<br>diazinon (0.001–0.014)<br>dicrotophos (0.121)<br>dimethoate (0.005–0.157)<br>ethion (0.002–1.130)<br>malathion (0.001–0.288)<br>methamidophos (0.009–0.052)<br>monocrotophos (0.063)<br>prothiophos (0.001–0.002)<br>profenophos (0.017–1.196)<br>triazophos (0.151–0.325)<br>cyfluthrin (0.031–0.272)<br>cypermethrin (0.106–5.398)<br>deltamethrin (0.228–0.788)<br>fenvalerate (0.112–0.942)<br>lamda-cyhalothrin (0.026–0.971)<br>permethrin (0.012–0.172) | 16<br>0<br>2<br>11<br>2<br>12<br>1<br>0<br>10<br>2<br>6<br>12<br>10<br>4<br>12<br>0 |  |  |  |  |  |
|            |    |                                   |  |  |   | 100   | 100                               | 96   | chlorpyrifos (0.002–0.211)<br>dimethoate (0.018–0.629)<br>ethion (0.003–2.887)<br>malathion (0.002–0.360)<br>methamidophos (0.001–0.118)<br>profenophos (0.002–0.211)<br>cyfluthrin (0.15–86.1)<br>cypermethrin (0.01–3.95)<br>fenvalerate (0.01–0.47)<br>lamda-cyhalothrin (0.01–0.80)<br>permethrin (0.01–0.80)<br>aldicarb (0.45)<br>carbaryl (0.14–0.31)<br>imidacloprid (0.003–0.048)<br>abamectin 1a (0.003–0.035)   | 0<br>0<br>21<br>0<br>0<br>2<br>25<br>1<br>18<br>6<br>8<br>1<br>0<br>0<br>13         |  |  |  |  |  |
| Watermelon | 10 | 1                                 | 0  | chlorpyrifos (0.032)   | 1   | -     | -                                 | -  | -  | -   |  |  |  |  |  |

| Pesticide            | MRL*<br>(mg/kg) | Urba | n          | -          |          |               | 1             | Rural            | Rural |            |            |          |               |               |                  |
|----------------------|-----------------|------|------------|------------|----------|---------------|---------------|------------------|-------|------------|------------|----------|---------------|---------------|------------------|
|                      |                 | z    | N detected | % detected | exceeded | %<br>exceeded | Mean<br>mg/kg | Min–Max<br>mg/kg | z     | N detected | % detected | exceeded | %<br>exceeded | Mean<br>mg/kg | Min-Max<br>mg/kg |
| Organophosphate      | •               |      | •          |            | -        |               | •             |                  |       |            |            |          |               |               | -                |
| Azinphos-ethyl       | 0.05-1 Co       | 300  | ND         | ND         | ND       | ND            | ND            | ND               | 337   | ND         | ND         | ND       | ND            | ND            | ND               |
| Azinphos-methyl      | 0.5-1 Eu        | 300  | 1          | 0.3        | 0        | 0.0           | 0.088         | 0.088-0.088      | 337   | ND         | ND         | ND       | ND            | ND            | ND               |
| Chlopyriphos         | 0.01-2 Co       | 300  | 107        | 35.7       | 61       | 20.3          | 0.076         | 0.002-1.394      | 337   | 92         | 40.5       | 43       | 18.9          | 0.052         | 0.001-1.596      |
| Diazinon             | 0.05-0.5 Co     | 300  | 10         | 3.3        | 4        | 1.3           | 0.094         | 0.0005-0.402     | 337   | 12         | 5.3        | 0        | 0.0           | 0.004         | 0.002-0.014      |
| Dicrotophos          | 0.5 Ta          | 300  | 21         | 7.0        | 0        | 0.0           | 0.056         | 0.003-0.403      | 337   | 37         | 16.3       | 3        | 1.3           | 0.095         | 0.001-0.973      |
| Dimethoate           | 0.05-1 Co       | 300  | 28         | 9.3        | 7        | 2.3           | 0.032         | 0.001-0.131      | 337   | 11         | 4.8        | 3        | 1.3           | 0.041         | 0.001-0.159      |
| EPN                  | 0.02–0.1 Ja     | 300  | 8          | 2.7        | 4        | 1.3           | 0.151         | 0.004-0.616      | 337   | ND         | ND         | ND       | ND            | ND            | ND               |
| Ethion               | 0.1-2 Eu        | 300  | 41         | 13.7       | 10       | 3.3           | 0.144         | 0.001-1.658      | 337   | 76         | 33.5       | 14       | 6.2           | 0.101         | 0.0002-1.279     |
| Fenitrothion         | 0.5 Co          | 300  | 8          | 2.7        | 0        | 0.0           | 0.027         | 0.004-0.161      | 337   | ND         | ND         | ND       | ND            | ND            | ND               |
| Malathion            | 0.2-8 Co        | 300  | 17         | 5.7        | 2        | 0.7           | 0.072         | 0.002-0.776      | 337   | 9          | 4.0        | 1        | 0.4           | 0.083         | 0.001-0.288      |
| Methamidophos        | 0.01–0.3 Ja     | 300  | 20         | 6.7        | 17       | 5.7           | 0.069         | 0.005-0.639      | 337   | 21         | 9.3        | 11       | 4.8           | 0.017         | 0.002-0.052      |
| Methidathion         | 0.05-1 Co       | 300  | 2          | 0.7        | 0        | 0.0           | 0.002         | 0.002-0.002      | 337   | ND         | ND         | ND       | ND            | ND            | ND               |
| Mevinphos            | 0.05–0.5 Ja     | 300  | 2          | 0.7        | 0        | 0.0           | 0.020         | 0.011-0.030      | 337   | ND         | ND         | ND       | ND            | ND            | ND               |
| Monocrotophos        | 0.05–1 Ja       | 300  | 6          | 2.0        | 2        | 0.7           | 0.031         | 0.003-0.089      | 337   | 4          | 1.8        | 3        | 1.3           | 0.095         | 0.001-0.313      |
| Parathion-methyl     | 0.05-0.5 Co     | 300  | 3          | 1.0        | 0        | 0.0           | 0.002         | 0.001-0.004      | 337   | 2          | 0.9        | 0        | 0.0           | 0.011         | 0.002-0.020      |
| Pirimiphos-ethyl     | NA              | 300  | 8          | 2.7        | 0        | 0.0           | 0.011         | 0.001-0.022      | 337   | 2          | 0.9        | 0        | 0.0           | 0.001         | 0.001-0.001      |
| Pirimiphos-methyl    | 0.05–5 Ja       | 300  | 15         | 5.0        | 0        | 0.0           | 0.011         | 0.002-0.034      | 337   | ND         | ND         | ND       | ND            | ND            | ND               |
| Profenophos          | 0.05–0.2 Ja     | 300  | 23         | 7.7        | 14       | 4.7           | 0.097         | 0.008-0.303      | 337   | 26         | 11.5       | 14       | 6.2           | 0.152         | 0.001-1.196      |
| Prothiofos           | 0.2–2 Ja        | 300  | 11         | 3.7        | 2        | 0.7           | 0.068         | 0.005-0.373      | 337   | 5          | 2.2        | 0        | 0.0           | 0.003         | 0.001-0.005      |
| Triazophos           | 0.1 Co          | 300  | 5          | 1.7        | 1        | 0.3           | 0.110         | 0.001-0.490      | 337   | 5          | 2.2        | 2        | 0.9           | 0.097         | 0.001-0.325      |
| Carbamates           |                 |      |            |            |          |               |               |                  |       |            |            |          |               |               |                  |
| 3-Hydroxy carbofuran | NA              | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |
| Aldicarb             | 0.3 Ja          | -    | -          | -          | -        | -             | -             | -                | 100   | 1          | 1.1        | 1        | 1.1           | 0.45          | 0.45             |
| Aldicarb sulfone     | NA              | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |
| Aldicarb sulfoxide   | NA              | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |
| Baygon               | NA              | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |
| Carbaryl             | 1.5 Th          | -    | -          | -          | -        | -             | -             | -                | 100   | 2          | 2.2        | 2        | 0             | NA            | 0.14-0.31        |
| Carbofuran           | 0.5 Ko          | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |
| Methiocarb           | NA              | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |
| Methomyl             | 1 Th            | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |
| Oxamyl               | 5 Ko            | -    | -          | -          | -        | -             | -             | -                | 100   | ND         | ND         | ND       | ND            | ND            | ND               |

**Supplement B.** Multiple pesticide residues detected in samples collected from urban and rural markets in upper northern Thailand during 2007-2013.

| Pesticide          | MRL*                 | Urba     | n          |            |          |            |            | Rural            |     |            |            |          |            |            |                  |
|--------------------|----------------------|----------|------------|------------|----------|------------|------------|------------------|-----|------------|------------|----------|------------|------------|------------------|
|                    | (mg/kg)              | z        | N detected | % detected | exceeded | % exceeded | Mean mg/kg | Min-Max<br>mg/kg | z   | N detected | % detected | exceeded | % exceeded | Mean mg/kg | Min-Max<br>mg/kg |
| Pyrethroids        |                      | <b>!</b> |            |            |          |            |            |                  |     | -1         |            |          | <b>_</b>   |            | •                |
| Cyfluthrin         | 0.1-2 Co             | -        | -          | -          | -        | -          | -          | -                | 171 | 106        | 62.0       | 59       | 34.5       | 0.19       | 0.01-2.79        |
| Cypermethrin       | 0.2-2 Co             | -        | -          | -          | -        | -          | -          | -                | 171 | 135        | 79.0       | 37       | 21.6       | 0.42       | 0.01-8.39        |
| Deltamethrin       | 0.02-2 Co            | -        | -          | -          | -        | -          | -          | -                | 171 | 99         | 57.9       | 97       | 56.7       | 0.92       | 0.008-17.45      |
| Fenvalerate        | 0.2-2 Co             | -        | -          | -          | -        | -          | -          | -                | 171 | 104        | 61.0       | 51       | 29.8       | 0.42       | 0.0025-3.74      |
| Lambda-cyhalothrin | 0.05-0.5 Co          | -        | -          | -          | -        | -          | -          | -                | 171 | 127        | 74.3       | 112      | 65.5       | 0.28       | 0.02-1.07        |
| Permethrin         | 0.5-5 Co             | -        | -          | -          | -        | -          | -          | -                | 171 | 114        | 66.7       | 5        | 2.9        | 0.14       | 0.002-2.27       |
| Other insecticides |                      |          |            |            |          |            |            |                  |     |            |            |          |            |            |                  |
| Abamectin 1a       | 0.01 Th              | -        | -          | -          | -        | -          | -          | -                | 100 | 40         | 44.0       | 13       | 13         | NA         | 0.003-0.035      |
| Abamectin 1b       | 0.01 Th              | -        | -          | -          | -        | -          | -          | -                | 100 | ND         | ND         | ND       | ND         | ND         | ND               |
| Imidacloprid       | 0.5 Ko               | -        | -          | -          | -        | -          | -          | -                | 100 | 79         | 86.8       | 0        | 0          | NA         | 0.003-0.048      |
| Fungicides         |                      |          |            |            |          |            |            |                  |     |            |            |          |            |            |                  |
| Carbendazim        | 0.05-0.5 Co<br>(7 K) | 32       | 21         | 65.6       | 5        | 15.6       | 0.047      | 0.008-0.334      | 100 | 27         | 29.7       | 0        | 0          | NA         | 0.006-0.077      |
| Dithiocarbamates   | 2.0 Th               | 54       | 10         | 18.5       | 3        | 5.6        | 4.18       | 0.120-30.64      | -   | -          | -          | -        | -          | -          | -                |
| (Mancozeb)         |                      |          |            |            |          |            |            |                  |     |            |            |          |            |            |                  |