Journal of Health Research
eISSN 2586-940X

Volume 37 | Issue 3 Article 5

2023

Health Risk Assessment from Organophosphate Insecticides Residues in Commonly Consumed Vegetables of Local Markets, Northern Thailand

Warangkana Naksen

surat hongsibsong

Zhen-Lin Xu

Natthapol Kosashunhanan

Tanyaporn Kerdnoi

Tippawan Prapamontol

Vanvimol Patarasiriwong

ORIGINAL STUDY

Health Risk Assessment from Organophosphate Insecticides Residues in Commonly Consumed Vegetables of Local Markets, Northern Thailand

Warangkana Naksen [a], Surat Hongsibsong [b,*], Zhen-Lin Xu [c], Natthapol Kosashunhanan [b], Tanyaporn Kerdnoi [d], Tippawan Prapamontol [d], Vanvimol Patarasiriwong [e]

[a] Faculty of Public Health, Chiang Mai University, Chiang Mai 50200, Thailand
[b] School of Health Sciences Research, Research Institute for Health Sciences, Chiang Mai University, Chiang Mai 50200, Thailand
[c] Guangdong Provincial Key Laboratory of Food Quality and Safety, College of Food Science, South China Agricultural University, Guangzhou 510642, China
[d] Environment and Health Research Unit, Research Institute for Health Sciences, Chiang Mai University, Chiang Mai 50200, Thailand
[e] Department of Environmental Quality Promotion, Ministry of Natural Resources and Environment, Bangkok 10400, Thailand

Abstract

Background: **Commonly consumed vegetables from local markets are considered at a higher risk for organophosphate insecticides (OPs) contamination. This study aimed to investigate the contamination of OP residues in commonly consumed vegetable samples from local markets in 5 provinces, i.e., Nan, Chiang Rai, Lamphun, Lampang, and Chiang Mai provinces located in Northern Thailand. A risk assessment of OPs among adult Thai population was also evaluated by calculating the estimated daily intake (EDI) and the acceptable daily intake (ADI).**

Method: **The multi cross-sectional studies were conducted in 5 provinces. Twelve vegetables were sampled from main local markets in every district of sampling sides. All samples have analyzed the level of 19 OP residues using GC-FPD. The estimated daily intake and health risk of OP residues were estimated by using hazard index.**

Results: **Twelve OP residues were detected in 42.8% of the samples, and 16.0% had residue levels that exceeded EU maximum residue levels. The most frequently detected OP residue in the vegetable samples was chlorpyrifos. The percentage of OP contaminated samples slightly increased throughout sampling time, 40.6%, 43.4%, and 47.9%, respectively. Hazard indices (HIs) were calculated based on data from National Health Examination Survey Office and the result showed that HIs of diazinon residue in Pakchoi and dimethoate residue in cucumber were over 1, hence posing a serious threat to human health.**

Conclusion: **The unacceptable levels of OP residues were observed in all types of vegetables collected from local markets, leading to hazardous exposure among consumers.**

Keywords: Health risk assessment, Organophosphate insecticides residues, Local markets, Northern Thailand

1. Introduction

In Thailand, the consumption of organic products is increasing because of the awareness of the dangers of contaminated foodstuffs [1]. However, farming in Thailand is still using chemical pesticides to protect crops [2]. In 2018, more than 18,000 tonnes of insecticides were imported into the country [3]. This number implies the high use of insecticides which might cause the accumulation in the environment and contamination in foods sold in domestic markets.

Organophosphate (OP) insecticides are insecticides with highly toxic and the most widely used insecticides available. OPs are considered a vital component of modern farming, playing a significant role in maintaining high agricultural productivity. OPs are widely used in Thailand because of their high efficiency against the target pest. However, unsuspecting use, uncontrolled and

Received 14 July 2021; revised 29 August 2021; accepted 30 August 2021.
Available online 31 August 2022

* Corresponding author.
E-mail address: surat.hongsibsong@cmu.ac.th (S. Hongsibsong).

https://doi.org/10.56808/2586-940X.1009

overuse of OPs may cause high levels of toxic residue. According to a previous study in Payao Province in Northern Thailand in 2014, OP residues such as diazinon, chlorpyrifos, and profenofos were detected in 12 commonly consumed vegetable samples [4]. The study in central Thailand in 2016 also found the residues of insecticides, including OPs, a synthetic pyrethroid, and carbamate in fruit and vegetable samples from local markets and supermarkets [5]. Another study in 2017 reported the OP residues in vegetable samples from the local fresh market and supermarket of Chiang Mai Province, northern Thailand. This study showed that the most commonly detected residue was chlorpyrifos in both fruits and vegetable samples [6].

OPs can inhibit the activity of cholinesterase, leading to overstimulation of neurotransmission. Exposure to OPs was associated with a range of adverse health effects, including neurological function disorder [7] and the male reproductive system [8]. Some studies reported that OPs exposure could be affected by childhood IQ [9] and brain anomalies [10]. Moreover, they can affect the endocrine system and prostate cancer [11]. The health effects of OPs can occur on occupationally exposed people, like farmworkers and applicators, and non-occupationally exposed people, as consumers. Although organic agriculture is currently being supported and controlled by the Thai government, chemical insecticide like OPs is still widely used in Thailand [6]. It might pose a health risk to Thai people. Therefore, information about the contamination status of OP residues is needed for food safety policy.

This study aimed to observe the contamination of OP residues in common consumable vegetable samples from local markets of 5 provinces in Northern Thailand and assess the health risk associated with OP residue in the vegetable.

2. Materials and methods

2.1. Study design, study area, and samples collection

This study investigated OP residues in sampling common consumable vegetables from local markets in 5 provinces, i.e., Nan, Chiang Rai, Lamphun, Lampang, and Chiang Mai province located in Northern Thailand during 2014–2018.

Twelve vegetables including broccoli (Brassica oleracea var. Italica), cabbage (Brassica oleracea var. capitate), cauliflower (Brassica oleracea var. botrytis), chili (Capsicum Annuum), Chinese cabbage (Brassica Rapa subsp. Pekinensis), coriander (Coriandrum sativum L), cucumber (Cucumis sativus), green onion (Allium fistulosum), kale (Brassica oleracea var. alboglabra), morning glory (Ipomoea Aquatica Forsk. Var. reptan), pakchoi (Brassica rapa L.), and yard long bean (Vigna unguiculata L.), which are commonly consumed in Northern Thailand, were sampled from main local markets in every district of sampling sides. The sampling pattern and number are shown in Table 1. All samples have analyzed the level of 19 OP residues using GC-FPD.

2.2. Ethical issue

The study was approved by the Human Experimentation Committee (HEC) of Research Institute for Health Sciences, Chiang Mai University (Certificated number 64/2013, 56/2014-6.

2.3. Preparation of samples for organophosphate pesticide analyzing

One kilogram of each type of vegetable was purchased from the market (from one seller was one sample). All samples without washing were chopped into small pieces, and then the final processed sample weighed 500 g were randomly taken for analysis. The processing procedure was performed according to Codex protocol 21 [12] within a day after sampling. All samples were transferred in an icebox to the Laboratory of Environmental and Occupational Health Sciences, Research Institute for Health Sciences, Chiang Mai University, and then kept in a freezer −20 °C before analysis.

2.4. Reagents and chemicals

OP standards with certified purity ranging from 97% to 99% including azinphos-ethyl, azinphos-methyl, chlorpyrifos, diazinon, dicrotophos, dimethoate, EPN, ethion, fenitrothion, malathion, methamidophos, methidathion, mevinphos, monocrotophos, parathion-methyl, pirimiphos-methyl, prothiofos, profenofos, triazophos, and triphenylphosphate as internal standard were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany).

All organic solvents, i.e., acetone, ethyl acetate, acetonitrile, hexane, and toluene, were analytical grade, purchased from J.T. Baker (PA, USA). Sodium chloride (NaCl) and sodium sulfate (Na_2SO_4) were obtained from Merck (Darmstadt, Germany).

2.5. Sample extraction and clean-up

The extraction and clean-up method used was followed the method from a previous study [4]. An aliquot of 5 g of sample was placed in a 50-mL centrifuge tube. Then, 250 μL of 5 μg/mL triphenylphosphate was added as an internal standard. Fifteen mL of acetonitrile were added twice, subsequently centrifuged for 5 min at 2500 rpm. The supernatant was transferred to a cleaned 50-mL centrifuge tube with the addition of 6 g of MgSO4 and 3 g of NaCl and then centrifuged again for 5 min at 2500 rpm for clean-up. The extract solution was filtered through Na2SO4 and evaporated to complete dryness using a vacuum rotary evaporator (Buchi, Switzerland) with a water bath at 30–35 °C and then reconstituted with 1 mL of ethyl acetate three times. One mL of ethyl acetate phase was pipetted to dispersive solid-phase extraction tube and centrifuged for 3 min at 2000 rpm. Finally, the upper supernatant was filtered by a 0.2-μm syringe filter and then inject into GC-FPD for analysis.

2.6. Analysis of organophosphate pesticide residue in vegetable samples

Sample preparation and extraction steps have followed the method of Sapbumrer and Hongsibsong [4]. The GC analysis consisted of an Agilent 7890B equipped with a flame photometric detector (GC-FPD), a capillary column (DB-5MS, 0.25 mm I.D.,30 m length, 0.25 1 m film thickness (J&W column; Agilent Technologies, USA), and a computerized data handling system (Open Lab CDS; Agilent Technologies, USA).

The temperature was 220 °C for the injection port (splitless mode). Temperature programming of the oven was as follows: initial temperature of 100 °C, first ramp at 15 °C/min to 180 °C (2 min), second ramp at 5 °C/min to 245 °C (10 min), and final temperature maintained at 280 °C for 4 min. The carrier gas was helium 99.999%.

2.7. Analytical quality assurance

Pooled vegetables were prepared and spiked with OP standard solutions for establishing the standard curve at concentrations 0, 20, 40, 80, 160, and 320 μg/L. Quality control samples at three concentration levels at low (20 μg/L), medium (40 μg/L), and high (160 μg/L) OP concentrations were prepared by spiking working standard solutions into pooled vegetables for recovery, accuracy, and precision testing. All samples for the calibration curve and quality control were processed and analyzed with the same method as described above.

Limit of detection (LOD) and Limit of quantitation (LOQ) were defined as 3 and 10 times of standard

Table 1. Sampling sites, periods and number of vegetable samples in this study.

Sampling periods	Sampling provinces	Number of samples in each sampling period	Number of residue detection in each sampling period (%)	Number of residue exceeded MRLs in each sampling period (%)	Number of samples in each sampling province	Number of residue detection in each sampling province (%)	Number of residue exceeded MRLs in each sampling period (%)
2014–2015	Chiang Rai	438	178 (40.6%)	63 (15.5%)	243	111 (45.7%)	46 (18.9%)
	Nan				195	67 (34.4%)	22 (11.3%)
2015–2016	Lampang	581	252 (43.4%)	137 (23.6%)	190	141 (36.1%)	63 (33.2%)
	Lamphun				391	111 (58.4%)	74 (18.9%)
2018	Chiang Mai	117	56 (47.9%)	20 (3.4%)	117	56 (47.9%)	20 (17.1%)
Total					1136 (100%)	486 (44.5%)	225 (19.8%)

deviation of the lowest concentrations, respectively [12]. Recoveries were calculated by comparing the concentrations of the extracted compounds with the actual concentration [13]. The method's precision was evaluated using different batches of quality control samples at 3 levels of the calibration curve. The method's precision was reported through the relative standard deviations (%RSD) with the acceptable range at below 20%.

For every 15 samples, quality control samples prepared at 80 ng/mL were prepared and passed through the entire analytical procedure.

2.8. Health risk assessment

Health risk assessments were done based on OPs residues detected in a vegetable sample from the local market of study areas. The arithmetic mean of OPs residue in each vegetable was used to evaluate the risk from intake in the adult population. The weight 60 kg body weight was used as the body-weight of the adult Thai population and 155.2 g vegetables/person as average daily vegetable consumption from the national data survey by the National Health Examination Survey Office (NHESO) [14]. The acceptable daily intake (ADI) was used from literature studies and estimated daily intake (EDI) was calculated for the pesticide residues and daily vegetables consumable.

The results of the hazard index (HI) were considered as a risk to human health; the HI more than 1 is considered as not safe for human health [15]:

HI = EDI/ADI
where,

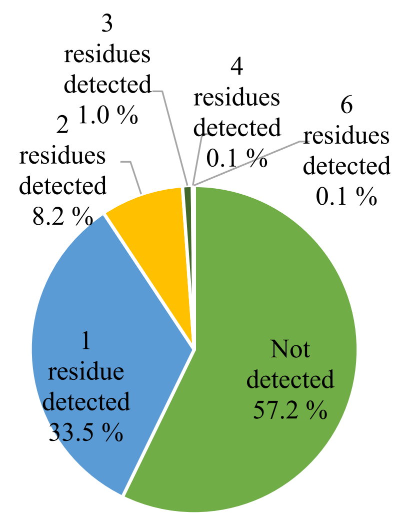

Fig. 1. The percent of vegetable samples relative to the number of detected OP residues (n = 1136).

EDI = estimated daily intake (mg/kg bw/day)
: [(concentration of detected pesticide x food consumption)/body weight of an adult]
ADI = Acceptable daily intake (mg/kg bw/day) based on International Programme on Chemical Safety (IPCS)

2.9. Statistical analysis

The statistical analysis was conducted using SPSS software. Descriptive statistics, including frequency (n), percentage (%), arithmetic mean, and standard deviation (SD), were used to explain the characteristics of samples. The levels of detected OP residues

Table 2. The recovery, LOD, LOQ, and %RSD of OP residues analysis.

Pesticide	% Recovery (n = 3)			Precision: % RSD (n = 3)	LOD (μg/kg)	LOQ (μg/kg)	R^2
	20 μg/kg	40 μg/kg	160 μg/kg				
Azinphos-ethyl	101.16	94.29	99.54	1.88	2.00	6.00	0.99989
Azinphos-methyl	90.78	86.60	91.99	5.87	1.00	2.00	0.99958
Chlorpyrifos	100.78	94.10	100.55	2.08	1.00	2.00	0.99996
Diazinon	103.15	95.85	98.19	2.06	1.00	9.00	0.99999
Dicrotophos	102.89	85.13	88.75	3.70	1.00	3.00	0.99977
Dimethoate	91.31	84.45	91.25	3.83	3.00	4.00	0.99985
EPN	101.41	92.86	97.84	1.98	1.00	2.00	0.99998
Ethion	106.53	92.13	99.12	1.88	1.00	2.00	0.99986
Fenitrothion	103.12	92.82	96.85	1.95	1.00	3.00	0.99997
Malathion	105.90	90.82	97.79	1.70	1.00	2.00	0.99988
Methamidophos	104.01	86.75	93.05	3.45	1.00	2.00	0.99963
Methidathion	102.40	89.83	98.68	3.03	1.00	2.00	0.99993
Mevinphos	105.09	92.65	95.85	3.67	1.50	5.00	0.99992
Monocrotophos	105.24	82.47	89.65	5.46	1.40	2.00	0.99937
Parathion-methyl	102.38	89.70	96.08	2.06	1.00	2.00	0.99992
Pirimiphos-methyl	98.70	89.81	96.00	2.67	1.00	2.00	0.99998
Profenofos	102.50	94.07	98.97	1.91	1.00	3.00	0.99999
Prothiophos	102.91	92.66	99.31	1.80	1.00	5.00	0.99986
Triazophos	98.36	94.89	95.63	1.71	2.00	5.00	0.99988

Table 3. The frequencies of detections, exceeding MRL and lists of OP residues exceeding MRL in vegetable samples of each sampling at different sampling sites and periods in 5 provinces of Northern Thailand.

November 2014–January 2015 (Chiang Rai and Nan)			November 2015 to January 2016 (Lampang and Lamphun)			January 2018 (Chiang Mai)		
N	% detection	OPs detected (% detection,% exceeded MRL)	N	% detection	OPs detected (% detection,% exceeded MRL)	N	% detection	OPs detected (% detection,% exceeded MRL)
21	38.1	Diazinon (24%, 19%), Chlorpyrifos (19%, 5%), Ethion (10%, 5%)	1	0.0	–	2	0.0	–
35	22.9	Chlorpyrifos (8%, 3%), Diazinon (14%, 0%), Ethion (3%, 3%)	33	60.6	Chlorpyrifos (42%, 36%), Diazinon (15%, 9%), Dimethoate (3%, 3%), Ethion (6%, 3%) Pirimiphos-methyl (6%, 3%), Profenofos (36%, 33%),	7	28.6	Chlorpyrifos (14%, 0%) Diazinon (14%, 0%),

in samples will be compared with established European commission regulation-maximum residue level [16], which set MRLs covering all types of vegetables in this study.

3. Results

This study was conducted in 5 provinces in the northern part of Thailand. Twelve types of commonly consumed vegetable samples were randomly purchased from local markets in those provinces. One thousand one hundred thirty-six samples were analyzed for OP residues between 2014 and 2018 (Table 1) at three different cross-sectional samplings. Nineteen OP residues were analyzed in vegetable samples using GC-FPD with the same analytical method, validated for recovery, linearity, precision, and accuracy (Table 2). Limit of detection (LOD) and the limit of quantification (LOQ) for all OP residues ranged from 1.0 to 3.0 and 2.0–6.0 μg/kg, respectively. For the three levels of spiked standards in pooled samples, the recovery values ranged from 82.47% to 106.53%. The calibration curves for all the pesticides analyzed were linear with $R^2 = 0.999$.

Four hundred and eighty-six (42.8%) out of 1136 samples were detected at least 1 OP residue. Approximately thirty percent were detected 1 OP residue. The highest number of detected residues in one sample is 6 (Fig. 1). The percent of OP contaminated samples slightly increased throughout sampling time, 40.6%, 43.4%, and 47.9%, respectively. The maximum and minimum contamination of OP residues were found in Lamphun (58.4%) and Nan (34.3%), respectively. The data was shown in Table 1.

Twelve out of 19 OP residues, including chlorpyrifos, diazinon, dimethoate, ethion, fenitrothion, malathion, methidathion, parathion-methyl, pirimiphos-methyl, profenophos, prothiophos, and triazophos, were detected in samples of this study (Table 3). OP residues were detected in samples of vegetables from every sampling market. The most frequently detected OP residues in the vegetable samples among all sampling sites were chlorpyrifos, diazinon, profenophos, dimethoate, and ethion. These residues were also the most frequently detected residues above the MRLs. However, only dimethoate shows a significant decrease of the median concentrations across the sampling times (data not shown).

Table 4 shows the results of OP contamination among 12 commonly consumed vegetables in three samplings. Coriander was the crop with the highest percentages of residue-detected samples (74%),

Table 4. The frequencies of detections, exceeding MRL and lists of OP residues exceeding MRL in vegetable samples of each sampling at different sampling sites and periods in 5 provinces of Northern Thailand during 2014–2018 (Con.).

November 2014–January 2015 (Chiang Rai and Nan)			November 2015 to January 2016 (Lampang and Lamphun)			January 2018 (Chiang Mai)		
N	% detection	OPs detected (% detection, % exceeded MRL)	N	% detection	OPs detected (% detection, % exceeded MRL)	N	% detection	OPs detected (% detection, % exceeded MRL)
29	13.8	Chlorpyrifos (10%, 0%), Ethion (3%, 0%)	44	29.6	Chlorpyrifos (18%,11%), Diazinon (20%,16%), Profenofos (7%, 4%)	4	25.0	Dimethoate (25%, 0%), Triazophos (25%, 0%)
0	NA	NA	84	34.52	Chlorpyrifos (23%, 13%), Diazinon (11%, 6%), Dimethoate (4%, 4%), Ethion (1%, 1%) Profenofos (12%, 8%),	16	68.8	Chlorpyrifos (44%, 19%), Diazinon (6%, 0%), Dimethoate (6%, 6%), Parathion-methyl (12%, 6%), Prothiophos (6%, 0%), Triazophos (0%, 0%)
47	46.8	Chlorpyrifos (21%, 9%), Diazinon (4%, 0%), Dimethoate (2%, 2%), Malathion (2%, 0%), Parathion-methyl (19%, 13%), Triazophos (4%, 0%)	48	22.9	Chlorpyrifos (8%, 6%), Diazinon (6%, 4%), Dimethoate (8%, 8%), Triazophos (0%, 0%)	5	20.0	Dimethoate (20%, 20%)

while morning glory was the lowest residue-detected crop (21%). However, coriander was the crop with the lowest percentages of MRL-exceeded samples (5%), while chili was the highest MRL-exceeded crop (25%) (data not shown). In each type of vegetable, there was more than one OP-residue detected. Chlorpyrifos was the most frequently detected residue in three sampling periods. Other 11 detected OP residues had a low percentage of detection. Only the concentration of dimethoate contamination was significantly decreased while other residues slightly changed but not significantly. There was one sample that detected the varying concentration of ethion in the second sampling batch. Chlorpyrifos and ethion were detected in all 12 types of vegetables, followed by diazinon, dimethoate, and malathion.

The vegetable samples in this study were mainly leafy vegetables except chili, cucumber and yard long bean. However, there was no obvious difference of OP contamination among vegetable characteristics (Fig. 2).

3.1. Assessment of health risks from OP residues

Table 5 showed the results of assessing the degree of risk that may arise from the residue of organophosphate pesticides in commonly consumed vegetables and widely available vegetables in fresh markets. The risk assessment was calculated using EDI and ADI. EDI compared to the average daily showed the estimated daily intake values of the OPs residues and health risk index in the commonly consumed vegetable samples. They are showed EDI and HI had no risk to health except diazinon residue in Pakchoi and dimethoate residue in cucumber with HI over 1 (see Table 6).

4. Discussions

This study shows the results of contamination status of OP residues in commonly consumed vegetable samples in Northern Thailand during 2014–2018 from three cross-sectional samplings. We sampled similar types of vegetables in each sampling time.

In our study, 19 OP residues were analyzed with good sensitivity using a multi-residue analysis method validated by evaluating recovery, linearity, and precision. Twelve out of 19 OP residues, including chlorpyrifos, diazinon, dimethoate, ethion, malathion, methidathion, parathion-methyl, pirimiphos-methyl, profenophos, prothiophos, and triazophos, were detected in samples of this study. These pesticides were in the lists of Thailand imported pesticides in 2018 except methidathion and parathion-methyl, which were in the watched and banned lists, respectively. Other residues which were not detected in this study were in the watched list, including dicrotophos and EPN and banned lists including azinphos-ethyl (banned in 2000), azinphos-methyl (banned in 2000), methamidophos (banned in 2003), mevinphos (banned in 2000), monocrotophos (banned in 2000), parathion ethyl (banned in 1988), and parathion methyl (banned in 2004) [17]. However, chlorpyrifos will be decided for banning in the next year. This result may indicate that the success of

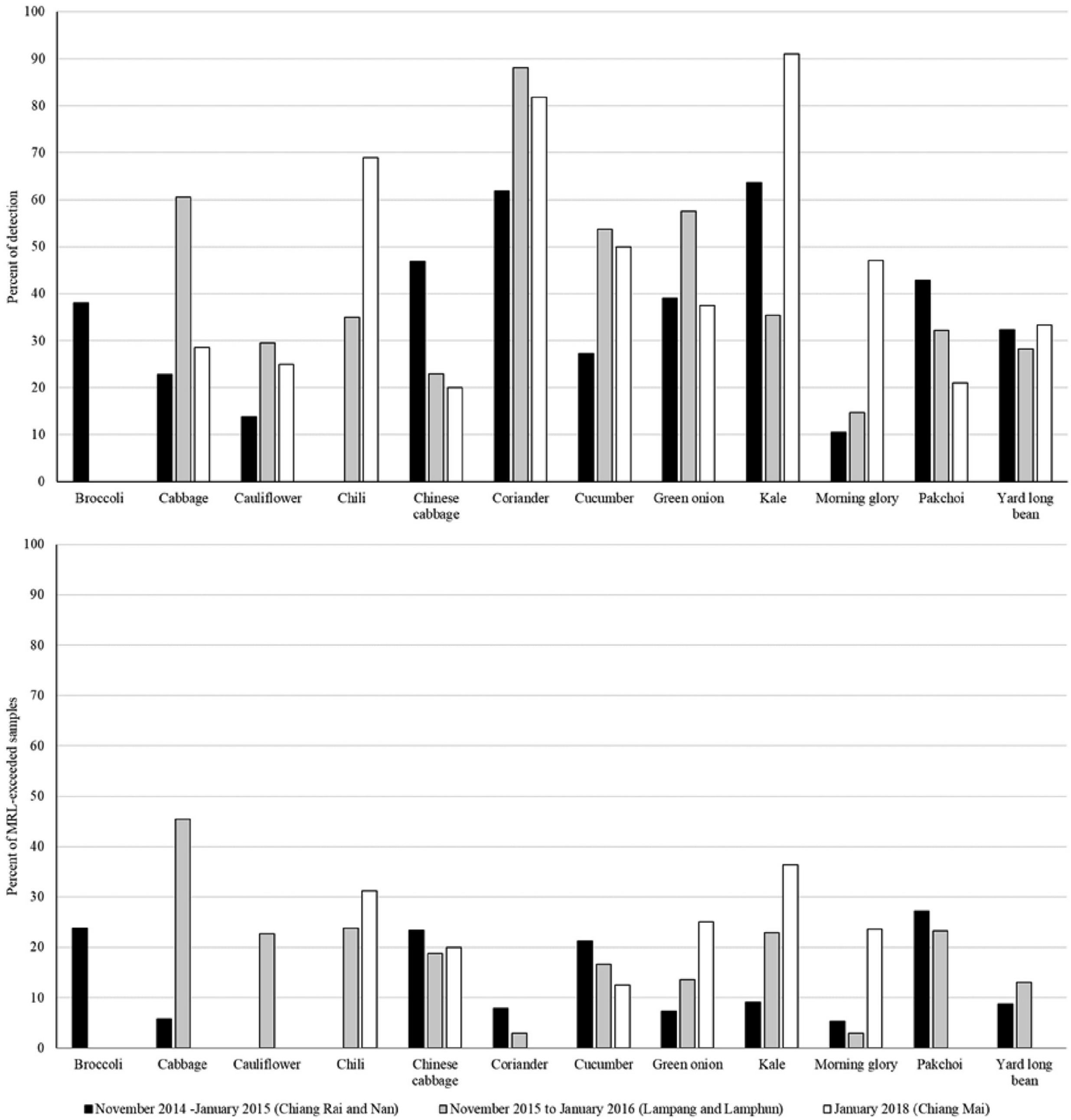

Fig. 2. Percentages of vegetable samples detected OP residues (upper) and above MRLs (lower) in three cross-sectional sampling in 5 provinces of Upper Northern Thailand categorized by vegetable types.

the pesticide ban policy, which was set by the legal body, such as the Department of Agriculture (DOA), in Thailand [18].

OP residues were detected in all 12 types of vegetables over the year 2014–2018 except broccoli. However, only six out of twelve, including chili, Chinese cabbage, cucumber, green onion, kale, and morning glory, were detected OP residues that exceeded the MRLs. The percentages of contamination exceeded the MRLs in coriander, cucumber, Pakchoi, and yard long bean was decreased across the different periods while those in green onion, kale, and morning glory were increased. This result shows the different trends of OP use in the different crops. The percentage of OP residue detection in each type of vegetable sample in this study was similar to previously reported residue [19]. These mean OPs were used in crops production for a long time, and still, residue in vegetables sold in market and consumers always had in their body. However, the unacceptable levels of OP residues

Table 5. Estimate daily intake (EDI) and health risk index (HI) assessment from organophosphate pesticide residue in vegetables.

Organophosphate pesticide	ADI* (mg/kg bw/ day)	Broccoli		Cabbage		Cauliflower		Chili		Chinese cabbage		Coriander	
		EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI	HI
Chlorpyrifos	0.00100	0.00007	0.0711	0.00028	0.2809	0.00060	0.6030	0.00068	0.6759	0.00019	0.1939	0.00026	0.2612
Diazinon	0.00200	0.00005	0.02328	0.00014	0.06835	0.00051	0.25401	0.00006	0.02990	0.00022	0.11213	0.00032	0.15766
Dimethoate	0.00200	–	–	0.00058	0.28764	0.00001	0.00362	0.00006	0.03174	0.00009	0.04300	0.00002	0.00802
Ethion	0.00200	0.00003	0.01358	0.00025	0.12470	0.00001	0.00517	0.00137	0.68547	0.00135	0.67737	0.00022	0.10872
Fenitrothion	0.00500	–	–	–	–	–	–	–	–	–	–	–	–
Malathion	0.02000	–	–	–	–	–	–	–	–	0.00001	0.00065	0.00002	0.00078
Methidathion	0.00100	–	–	–	–	–	–	–	–	–	–	–	–
Parathion-methyl	0.02000	–	–	–	–	–	–	0.00003	0.00147	0.00028	0.01401	0.00002	0.00084
Pirimiphos-methyl	0.03000	–	–	0.00002	0.00078	–	–	–	–	–	–	–	–
Profenophos	0.00300	–	–	0.00039	0.13045	0.00022	0.07315	0.00041	0.13761	–	–	0.00060	0.19935
Prothiophos	0.10000	–	–	–	–	–	–	–	–	–	–	–	–
Triazophos	0.00100	–	–	0.00001	0.00517	–	–	–	–	0.00002	0.02344	–	–

* based on International Programme on Chemical Safety (IPCS).

found in this study imply unsafe consumption for consumers. This result is also related to other previous studies that reported the high exposure to OP insecticides among farmers and consumers living in Northern Thailand [6,20]. From the result, in every sampling batch which from different provinces, there were samples which had OP residues exceeded MRLs. This might be caused by the intensive use of OPs in all farming areas.

Considering the contaminated level of OP residues detected in vegetable samples, the ranges of median concentrations in each type of vegetables were broad. Cabbage had found the highest median concentration of chlorpyrifos and the highest percentage of MRL-exceeded samples. The high median concentrations of other commonly found OP residues such as diazinon, dimethoate, and ethion were found in cabbage, cauliflower, and Chinese cabbage. Although the highest OP residue detection was found in coriander, the low number of samples was detected residue above MRLs because of the high MRLs set for coriander (Table 6).

According to sampling sites and sampling period, a significant difference was not found. This may imply that OP residue use in every area in Northern Thailand was not varied. All detected OP residues are currently permitted for agricultural use. Reasonably, there was no change of OP use observed over the period. Although there is a plan for banning chlorpyrifos soon, it is still available in the commercial market.

According to sampling sites and sampling period, the significant difference was not found. This may imply that the patterns of OP residue use in every area in Northern Thailand were not varied. All detected OP residues are currently permitted for

Table 6. Estimate daily intake (EDI) and health risk index (HI) assessment from organophosphate pesticide residue in vegetables (Con.).

Organophosphate pesticide	ADI* (mg/kg bw/ day)	Cucumber		Green onion		Kale		Morning glory		Pakchoi		Yardlong bean	
		EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI	HI	EDI	HI
Chlorpyrifos	0.001	0.00004	0.0425	0.00014	0.1439	0.00003	0.0311	0.00030	0.3047	0.00093	0.9291	0.00013	0.1288
Diazinon	0.002	0.00004	0.01950	0.00002	0.00854	0.00008	0.03783	–	–	0.00223	1.11718	–	–
Dimethoate	0.002	0.00416	2.07839	–	–	0.00006	0.03076	0.00001	0.00336	0.00076	0.37934	0.00001	0.00349
Ethion	0.002	0.00001	0.00336	0.00002	0.00993	0.00001	0.00254	0.00003	0.01617	0.00006	0.02949	0.00001	0.00569
Fenitrothion	0.005	–	–	0.00004	0.00787	–	–	–	–	–	–	–	–
Malathion	0.020	–	–	0.00019	0.00947	–	–	–	–	–	–	–	–
Methidathion	0.001	0.00003	0.02641	–	–	0.00004	0.04496	–	–	–	–	–	–
Parathion-methyl	0.020	0.00003	0.00166	0.00014	0.00686	–	–	–	–	0.00002	0.00101	–	–
Pirimiphos-methyl	0.030	0.00001	0.00023	0.00005	0.00165	0.00004	0.00138	–	–	0.00001	0.00035	–	–
Profenophos	0.003	–	–	0.00001	0.00384	0.00006	0.01919	0.00012	0.04154	0.00219	0.73082	–	–
Prothiophos	0.100	–	–	–	–	–	–	–	–	0.00001	0.00015	–	–
Triazophos	0.001	0.00001	0.00556	0.00001	0.01081	0.00003	0.02851	0.00001	0.00789	0.00023	0.23228	–	–

* based on International Programme on Chemical Safety (IPCS).

agricultural use. Reasonably, there was no change of OP use observed over the period of time. Although there is a plan for banning chlorpyrifos in the near future, it is still available in the commercial market.

Among the 19 OPs, there were the unacceptable health risk due to diazinon and dimethoate highly contaminated Pakchoi and cucumber samples, respectively [5]. Both diazinon and dimethoate can pose health risk to human even at low levels, and the continuous intake could cause health hazards in the long term [21]. However, EDIs which were calculated based on consumption rate and average body weight of population showed that individual EDIs were mostly lower than ADIs, thus no risk by calculation should be concerned. However, we calculated EDIs and HIs individually. The cumulative risk of pesticides should be more studied.

The limitation of this study is the different sampling sites and periods. This led to the difficulty of OP contamination comparison among sites and time. However, in all sampling areas, the types of vegetables selected were similar, and all sampling sites were in Northern Thailand. They shared similar patterns of crop cultivation. Food consumption is also one of the weak points of this study, and we had no data specific to people of the Northern part of Thailand. Thus, we carefully analyzed and discussed the results considering the mentioned limitation.

5. Conclusions

This study reported the contamination status of OP residues in commonly consumed vegetables monitored in local markets in Northern Thailand. The results show similar patterns of OP use among 5 provinces and different years. The unacceptable level of OP residues was observed in all types of vegetable samples collected from local markets, leading to hazardous exposure among consumers in these areas.

Residues of diazinon and dimethoate detected in the vegetable samples showed a potential threat to human health in the adult Thai population. It is recommended to continue monitoring of OPs residue in commonly consumed vegetables in local markets. The result from this study showed the high rate of detection of OP residues in vegetables samples although estimated health risks of consumption of vegetables contaminated with OP residues were mostly in acceptable range. We suggested that consumers should always wash fruits and vegetables before eating or cooking appropriately. At the same time, the policy on agricultural pesticide management and practical education program for farmers and consumers should be implemented to protect people's health.

Data availability

All data were included in this paper.

Funding statement

This study was supported by Department of Environmental Quality Promotion, Ministry of Natural Resources and Environment, Thailand, and Chiang Mai University.

Conflict of interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

Acknowledgments

Authors would like to thank Department of Environmental Quality Promotion, Ministry of Natural Resources and Environment, Thailand, and Chiang Mai University for supporting.

References

[1] Sangkumchaliang P, Huang WC. Consumers' perceptions and attitudes of organic food products in Northern Thailand. Int Food Agribus Manag Rev 2012;15(1):87−102.
[2] Yanakittkul P, Aungvaravong C. A model of farmers intentions towards organic farming: a case study on rice farming in Thailand. Heliyon 2020;6(1). e03039.
[3] DOA (Department of Agriculture). Amount of imported agricultural hazardous substances during 2013-2019 [cite 2020 December 20] Available from: http://service.nic.go.th/em-dashboard.php?id=5.
[4] Sapbamrer R, Hongsibsong S. Organophosphorus pesticide residues in vegetables from farms, markets, and a supermarket around kwan phayao lake of northern Thailand. Arch Environ Contam Toxicol 2014;67:60−7.
[5] Wanwimolruk S, Phopin K, Boonpangrak S, Prachayasittikul V. Food safety in Thailand 4: comparison of pesticide residues found in three commonly consumed vegetables purchased from local markets and supermarkets in Thailand. PeerJ 2016;4. Article ID e2432.
[6] Silipunyo T, Hongsibsong S, Phalaraksh C, Laoyang S, Kerdnoi T, Patarasiriwong V, et al. Determination of organophosphate pesticides residues in fruits, vegetables and health risk assessment among consumers in Chiang Mai province, northern Thailand. Res J Environ Toxicol 2016;11:20−7.
[7] Steenland K, Jenkins B, Ames RG, O'Malley M, Chrislip D, Russo J. Chronic neurological sequelae to organophosphate pesticide poisoning. Am J Publ Health 1994;84(5):731−6.
[8] Mehrpour O, Karrari P, Zamani N, Tsatsakis AM, Abdollahi M. Occupational exposure to pesticides and consequences on male semen and fertility: a review. Toxicol Lett 2014;230:146−56.
[9] Bouchard MF, Chevrier J, Harley KG, Kogut K, Vedar M, Calderon N, et al. Prenatal exposure to organophosphate pesticides and IQ in 7-year-old children. Environ Health Perspect 2011;119:1189−95.

[10] Rauh VA, Perera FP, Horton MK, Whyatt RM, Bansal R, Hao X, et al. Brain anomalies in children exposed prenatally to a common organophosphate pesticide. Proc Natl Acad Sci USA 2012;109:7871–6.
[11] Fritschi L, Glass DC, Tabrizi JS, Leavy JE, Ambrosini GL. Occupational risk factors for prostate cancer and benign prostatic hyperplasia: a case-control study in western Australia. Occup Environ Med 2007;64:60–5.
[12] Codex. Recommended methods of sampling for the determination of pesticide residues for compliance with MRLs. CAC/GL 1999;33.
[13] AOAC. Official methods of analysis of AOAC international. nineteenth ed. Gaithersburg, Md: AOAC International; 2012.
[14] Suntudrob J, Jongmevasna W, Payanan T, Srikote R, Wittayanan W. Monitoring of pesticide residues in domestic vegetables in Thailand during 2015. Asia Pac J Sci Technol 2018;23(4). Article ID.: APST-23-04-03.
[15] Hossain MS, Fakhruddin ANM, Chowdhury AZM, Rahman MA, Khorshed AM. Health risk assessment of selected pesticide residues in locally produced vegetables of Bangladesh. Int Food Res J 2015;22(1):110–5.
[16] European Commission. EU pesticide database [cite 2020 December 20] Available from: https://ec.europa.eu/food/plant/pesticides/eu-pesticides-database/public/?event=pesticide.residue.selection&language=EN.
[17] DOA. List of imported hazardous substances in 2019 (in thai). Bangkok, Thailand: Department of Agriculture; 2019.
[18] Panuwet P, Siriwong W, Prapamontol T, Ryan PB, Fiedler N, Robson MG, et al. Agricultural pesticide management in Thailand: situation and population health risk. Environ Sci Pol 2012;17:72–81.
[19] Prapamontol T, Hongsibsong S, Naksen W, Kerdnoi T, Kawichai S, Polyiem W, et al. Multiple pesticide residues found in vegetables and fruits from rural and urban markets in upper northern Thailand. Chiang Mai Univ J Nat Sci 2021;20(1):1–12.
[20] Naksen W, Prapamontol T, Mangklabruks A, Chantara S, Thavornyutikarn P, Srinual N, et al. Associations of maternal organophosphate pesticide exposure and pon1 activity with birth outcomes in sawasdee birth cohort, Thailand. Environ Res 2015;142:288–96.
[21] Salih EMA. Toxic effect of dimethoate and diazinon on the biochemical and hematological parameters in male rabbits. Jordan J Biol Sci 2010;3(2):77–82.