RESIDUES OF ORGANOPHOSPHATE PESTICIDES USED IN VEGETABLE CULTIVATION IN AMBIENT AIR, SURFACE WATER AND SOIL IN BUENG NIAM SUBDISTRICT, KHON KAEN, THAILAND

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Abstract. Agricultural pesticide utilization is one of the important problems in rural and urban crop-cultivated areas, with the majority of pollutants dispersing via ambient air, water and other natural pathways. This study was therefore conducted in a specially selected village which is known to be a leading vegetable growing area in Khon Kaen Province. The aim of the study was to assess pesticide residues, and measure the seasonal fluctuations in organophosphate concentrations during 2010 in the environment of a risk area. Samples from selected sites were collected in two phases: Phase I was in summer (during March to May) and Phase II was in winter (during October to December). A total of 150 samples were analyzed using gas chromatography with flame photometric detection. The results showed that dicrotophos, chlorpyrifos, profenofos and ethion were found at the highest concentrations in soil and at the lowest concentrations in ambient air (p<0.001). The highest mean concentration of a pesticide in ambient air samples was 0.2580 ± 0.2686 mg/m^3 for chlorpyrifos in summer and 0.1003 ± 0.0449 mg/m^3 for chlorpyrifos in winter. In surface water samples, the highest mean concentration of a pesticide was 1.3757 ± 0.5014 mg/l for dicrotophos in summer and 0.3629 ± 0.4338 mg/l for ethion in winter. The highest mean concentration of a pesticide in soil samples was 42.2893 ± 39.0711 mg/kg ethion in summer and 90.0000 ± 24.1644 mg/kg of ethion in winter.

Keywords: organophosphate, residues, ambient air, water, soil, Thailand

INTRODUCTION

Thailand is a major food supplier for the world's population. Forty-two percent of the Thai working population are farmers. Cash crops are major agricultural products being exported (Taylor, 1996). However, nowadays Thai farmers still have many economic concerns about the control of weeds, pathogens, and insect pests. Because of the high pesticide application rates commonly used in Thai agriculture, pesticide residues have been widely detected in soils, surface

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and ground waters, and in agricultural products. Alarmingly, they have also been detected in humans, such as in the breast milk of female farmers (Baun *et al,* 1998; Thapinta and Hudak, 2000; Stuetz *et al,* 2001), and pesticide exposure has been related to elevated cancer risks and reproductive dysfunctions in agricultural workers. Apart from the risks to human health, the use of pesticides has been associated with several other concerns such as the death of farm animals and the alteration of the local environment. Organophosphate pesticides are a group of pesticides widely used in Thailand, and these can be efficiently absorbed by inhalation, ingestion, and skin penetration. The occurrence of poisoning depends on the rate at which the pesticide is absorbed (Jaipieam *et al*, 2009).

In recent years, several national environmental agencies have been involved in the development of regulations to eliminate or severely restrict the use and production of a number of pesticides (Aranzazu *et al*, 2011). Despite these actions, pesticides continue to be present and cause adverse effects on humans and the environment. Monitoring the presence of pesticides in different environmental compartments has proved to be a useful tool to quantify the amount of pesticides entering the environment and to assess ambient levels for the evaluation of trends and potential problems (Muir *et al*, 2004; Donal *et al*, 2005). Recently, different countries have undertaken, or are currently undertaking, campaigns to regulate and control use of pesticides, with various degrees of intensity and success (Wiersma, 2004). Although numerous local and national monitoring studies have been conducted around the world and provided nationwide patterns of pesticide presence and distribution (Roberts *et al*, 1998; Andreu and Pico, 2004), there are still a number of gaps in the information regarding pesticides.

Several previous studies have shown that Thai rice/vegetable farmers grow vegetables on community farms near residential areas and that this could cause pesticide residues to spread easily into the places where people live (Siriwong *et al*, 2007; Kongtip *et al*, 2009). People in these areas should be appropriately informed about the risks and given adequate knowledge about the quality of the ambient air, surface water and soil and effective measures for protecting their health. The aim of this study which was conducted in Khon Kaen, Thailand was to assess pesticides residues and measure the seasonal fluctuations in organophosphate concentrations during 2010 in the environment of a risk area. The results of this study were expected to increase knowledge about the quality of ambient air, surface water and soil and to improve health protection during seasonal peaks of pesticide residues.

MATERIALS AND METHODS

Study area

Khon Kaen Province was selected on the basis of previous studies which showed that people in this part of Thailand have a high risk of pesticide exposure, especially from fields in their rice/vegetable farms (Chaigarun and Nathapindhu, 2006). The village of Bueng Chim in the Bueng Niam Sub-district was deliberately selected for study because, according to information from the Khon Kaen Provincial Agricultural Office, it is known to be the leading vegetable growing area in the province. This area is located in the Mueang District of Khon Kaen Province which is in the northeast of Thailand. A map of the study area and sample sites is

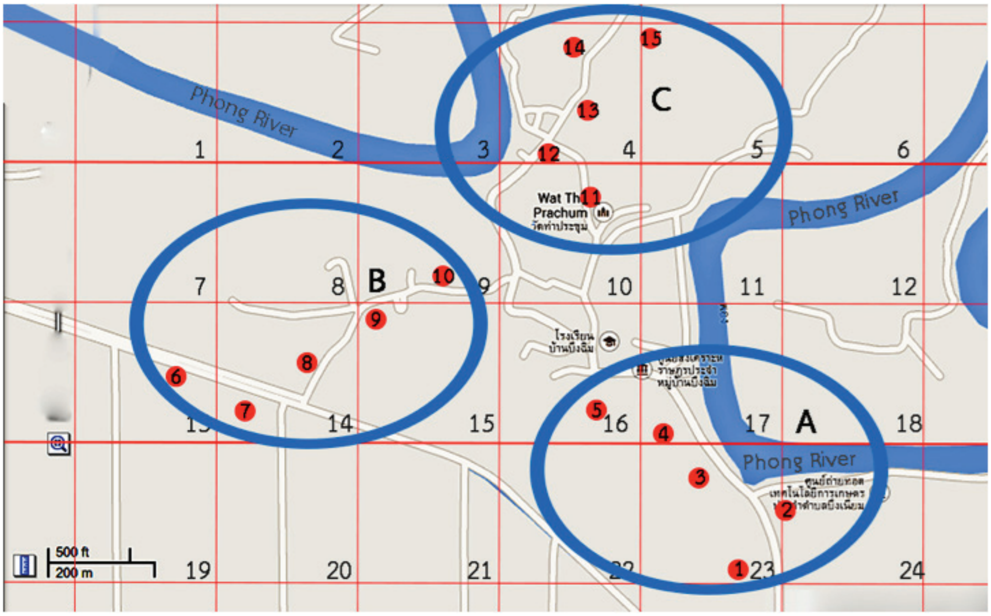

Fig 1–Grid reference system for the Bueng Chim sampling site.

shown in Fig 1. As the map shows, the village is located along the Phong River which is an important water resource for daily usage and cultivation among surrounding villages and districts. Farmers of the village usually grow rice twice a year and plant mixed vegetables several times per year.

Site identification

The sample site selection process began by identifying the vegetable growing areas of Bueng Chim Village where organophosphate pesticide applications typically occur. The map was developed and applied using a global positioning system (GPS). By means of grid sampling, samples were taken at regularly spaced intervals. An initial location or time was chosen at random, and then the remaining sampling locations were defined so that all locations were sampled at regular intervals over a given area. Using random systematic sampling, an initial sampling location was chosen at random and the remaining sampling sites were specified so that they were located according to a regular pattern. In this study, the sample sites were separated into three zones, which were designated as zones A, B and C, with the use of a grid reference system. The entire site was divided into 24 grid zones. The zones and grid numbers are shown in Fig 1.

Methodology

The total concentrations of organophosphate pesticide residues in the environment were monitored by collecting 150 environmental samples at 15 sample collection stations (five in each of zones A, B and C). Environmental sampling took place in two phases. Phase I was performed in summer (during March-May, 2010), and Phase II was performed in winter (during October-December, 2010).

Air sampling. The data were collected outdoors at various air sampling stations. Sampling was conducted according to the NIOSH Method 5600 for OP pesticides (NIOSH, 1994). OVS (OSHA versatile sampler) tubes containing XAD-2 sorbent were attached to a pump using flexible tubing. The sampling assembly was positioned with the intake downward or in a horizontal position in an unobstructed area, at least 30 m to air flow (Harner *et al,* 2006) and at 1 to 2 m above ground level. After the XAD-2 cartridge was correctly inserted and positioned, the elapsed time meter was activated, and the start time was recorded. The pumps were checked during the sampling process, and any abnormal conditions discovered were recorded on the sampling form. Ambient temperatures and barometric pressures were measured and recorded periodically during the sampling procedure. At the end of the desired sampling period, the cartridge was wrapped in its original aluminum foil packaging and placed in a sealed, labeled container for transport under a blue ice pack (<4ºC), and then taken to the laboratory. At least one field blank was returned to the laboratory with each group of samples and then treated exactly like a sample except that no air was drawn through the cartridge. It was stipulated that extraction must occur within seven days of sampling and that analysis must be done within 40 days of extraction. All samples were injected into the autosampler of a gas chromatograph with flame photometric detection (GC-FPD) (Hela *et al,* 2000). The appropriate dilution factor was applied for all calculations. Pesticide concentrations in the samples were compared with an internal standard.

Surface water sampling. Water samples were collected in 1-liter bottles with Teflon-lined caps, which were rinsed with samples immediately prior to collection. The bottles were prepared by cleaning with soap and rinsing five times with tap water. Prior to each sample's collection, the tap was kept open for five minutes to wash out any contaminants in the pipes and to rinse out the bottles. Samples were stored on ice for up to 12 hours, filtered through baked, stored in the dark at 4ºC, and were analyzed within seven days following sample collection (Lawrence, 1996). The samples were analyzed for organophosphates using gas chromatography (GC).

Soil sampling. Soil samples were collected in the same sampling stations as the air samples. Representative soil samples were best obtained by using a core sampling tool. The use of a proper sampling tool was essential for sampling to depths below 15 cm. Soil cores were collected at 0-15 cm depths in 15 to 20 sites that represented soil samples in vegetable area. The benchmark process was further extended by establishing a couple of benchmark areas in different areas which allowed customization of fertilizer rates. By identifying a primary benchmark area and a secondary benchmark area, and perhaps even a tertiary benchmark area, a fine-tuned fertility management strategy could be achieved even without variable rate technology.

The best time to identify different soil characteristics was through crop development. At the beginning of the growing season, differences in crop establishment and intensity could be seen, making it easier to choose a representative location. Other ways of selecting potential benchmark sites included the use of productivity, yield, aerial or topographic maps.

RESULTS

Seasonal variation in types of vegetables and pesticides

There was some seasonal variation in the type of vegetables planted in the study

Table 1
Comparison of concentrations of organophosphate pesticides in ambient air, water and soil samples during summer.

	Mean (SD)	F^a	*p*-value
Dicrotophos		5.004	0.016
Ambient air (mg/m^3)	0.0925 (0.1520)		
Water (mg/l)	1.3757 (0.5014)		
Soil (mg/kg)	18.3964 (17.8635)		
Chlorpyrifos		22.918	<0.001
Ambient air (mg/m^3)	0.2580 (0.2686)		
Water (mg/l)	0.2920 (0.2163)		
Soil (mg/kg)	32.1927 (22.9684)		
Profenofos		25.500	<0.001
Ambient air (mg/m^3)	0.0347 (0.0413)		
Water (mg/l)	0.9520 (0.4982)		
Soil (mg/kg)	41.8080 (28.2392)		
Ethion		7.828	0.002
Ambient air (mg/m^3)	0.0137 (0.0129)		
Water (mg/l)	0.2529 (0.1287)		
Soil (mg/kg)	42.2893 (39.0711)		

[a] ANOVA (*F*-test)

area. Every year Chinese kale, coriander, spring onion and chili were grown in summer, while cabbage, Chinese cabbage and cauliflower were grown in winter. Farmers regularly applied organophosphate pesticides, such as chlorpyrifos, profenofos, ethion and dicrotophos to kill insects.

Comparison of concentrations of organophosphate pesticide residues

Pesticide residues leftover from agriculture not only contaminated crops, but also the environment, such as ambient air, surface water and soil. These findings reinforce the concern about pollution by organophosphates in areas surrounding agriculture areas of pesticide use. In summer, the results showed that pesticides were found at the highest mean concentration in soil and at the lowest mean concentration in ambient air (dicrotophos, p=016, chlorpyrifos, $p<0.001$, profenofos, $p<0.001$ and ethion, $p=0.002$) as shown in Table 1.

In winter, the results similarly showed that chlorpyrifos, profenofos and ethion were found at the highest mean concentration in soil and at the lowest mean concentration in ambient air ($p<0.001$) (Table 2).

Multiple pesticides were detected in all vegetables areas. In ambient air samples, the highest mean concentration in vegetable areas was 0.2580 ± 0.2686 mg/m^3 for chlorpyrifos during March to May and 0.1003 ± 0.0449 mg/m^3 for chlorpyrifos in winter. In surface water samples, the highest mean concentration of pesticides in vegetable areas was 1.3757 ± 0.5014 mg/l for dicrotophos in summer and 0.3629 ± 0.4338 mg/l for ethion in winter. In soil samples, the highest mean concentration of pesticide in vegetable areas was 42.2893 ± 39.0711 mg/kg for ethion in summer and 90.0000 ± 24.1644 mg/kg for ethion in winter.

Table 2
Comparison of concentrations of organophosphate pesticides in ambient air, water and soil samples during winter.

	Mean (SD)	F^a	p-value
Chlorpyrifos		33.505	<0.001
Ambient air (mg/m^3)	0.1003 (0.0449)		
Water (mg/l)	0.1214 (0.1047)		
Soil (mg/kg)	28.5740 (18.7093)		
Profenofos		14.170	<0.001
Ambient air (mg/m^3)	0.0020 (0.0005)		
Water (mg/l)	0.3186 (0.1732)		
Soil (mg/kg)	16.5956 (10.9642)		
Ethion		199.951	<0.001
Ambient air (mg/m^3)	0.0109 (0.0173)		
Water (mg/l)	0.3629 (0.4338)		
Soil (mg/kg)	90.0000 (24.1644)		

[a] ANOVA (*F*-test)

DISCUSSION

The results showed that a wide range of pesticides of different chemical groups was used in crop protection. Chlorpyrifos was the pesticide most frequently applied in many areas. In many cases, mixtures of different types of pesticides were applied to the same crop. In addition, application rates often exceeded manufacturers' recommendations because farmers often believed that mixing several kinds of pesticides was more effective than using only a single kind. Pesticides were applied to cash crops every 4-7 days, and even more frequently during the summer. Recommended pre-harvest intervals were not strictly followed, and early harvesting was encouraged due to incidents of theft. Thus, pesticide contamination could be detected and could cause health problems to the farmers and other people in the area.

Despite the benefits of pesticide use in agricultural production, spray drift of pesticides can be a threat to the sustainability of farming and is a major source of pollution in agricultural areas (Hamilton and Crossley, 2004). The methods of pesticide application are a major factor in the increase of pesticide residues in the environment. Based on the data from this study, there is an urgent need to reduce pesticide drift and improve application equipment and methods. An environmental risk assessment of the pesticides used in Thailand can form an important contribution towards reducing the spread of impacts from the use of pesticides.

A total of 150 samples from 15 areas were studied. Chlorpyrifos, dicrotophos, profenofos and ethion residues were found in the air samples from vegetable fields. The results showed that chlorpyrifos was the most frequently found organophosphate. The concentration of dicrotophos in winter was below the analytic limit of detection for all samples. The concentration of profenofos was significantly higher in summer than in winter. However, there were no differences in chlorpyrifos, dicrotophos and ethion concentrations between the summer and winter seasons.

According to a previous study, organophosphate pesticides were the most commonly used pesticides in conventional farming areas (Visuthismajarn *et al*, 2005). Through personal interviews, it was found that chlorpyrifos, dicrotophos and profenofos were intensively used on conventional farms. Chlorpyrifos was used as an insecticide on farms to kill worms and in the household to kill cockroaches. Dicrotophos and profenofos are often used as an acaricide to kill mites. A previous study on biological monitoring found that farm children living in or near the vegetable farming areas in the Bang Rieng agricultural subdistrict in southern Thailand excreted higher levels of a dialkyl phosphate metabolite (a biomarker of organophosphate exposure) in their urine than reference groups living outside the farm area (Petchuay *et al*, 2006). Jaipieam *et al* (2009) found residues of organophosphate pesticides (chlorpyrifos, dicrotophos and profenofos) in samples taken from 33 vegetable growers compared with 17 reference subjects. Their results showed that median concentrations of organophosphates in the air in farm areas were in the range of 0.022-0.056 mg/m^3, while the air in non-farming areas contained organophosphate concentrations in the range of $<$0.0016-$<$0.005 mg/m^3. Another study in Thailand comparing chlorpyrifos concentrations on vegetable and rice farms found that the chlorpyrifos exposure of 33 vegetable growers (0.0016-0.4537 mg/m^3) was slightly less than the chlorpyrifos exposure of 31 rice farmers (ranging from 0.0216-0.5500 mg/m^3). This might be a result of the relative size of the farms; the average area of an individual vegetable farm is 0.8 hectare, while rice farms were generally in the range of 1.6-3.2 hectares (Kongtip *et al*, 2009).

While many of the newer pesticides currently in use are less persistent than their predecessors, they also contaminate the air and can travel many miles from the target areas; for example, chlorothalonil, chlorpyrifos, metolachlor, terbufos and trifluralin have been detected in Arctic environmental samples (air, fog, water, snow) (Dimitra *et al*, 2008). Regarding modern insecticides, organophosphorus compounds (parathion, malathion, diazinon and chlorpyrifos) have been the most often studied. Locally high concentrations of pesticides in the air were found to be very seasonal and were correlated with patterns of local use (Dimitra *et al*, 2008). The highest concentrations in the air usually occur in the spring and summer months, coinciding with application times and warmer temperatures. However, for some pesticides that are detected, it is not always clear if their concentration and frequency in the air is associated with local use or long-range transport from other sources, or both (Tuduri *et al*, 2006).

Pesticides may be lost from the site of application to surface waters, and this loss can occur in measurable concentrations throughout the year (Neumann *et al*, 2002; Blanchoud *et al*, 2007). There are two main mechanisms for agricultural pesticide losses: (a) diffuse losses from agricultural soils after pesticide applications to crops, and (b) pesticide spills on roads and farmyards washed off by the urban drainage system (point-source losses). In general, for diffuse losses, pesticide concentrations have been found to increase with increasing discharge during rain events in the application season. Spills, on the other hand, may cause extremely high but short-lived concentration peaks independently of the discharge dynamics (Irene, 2010).

Mean organophosphate concentrations in vegetable areas were higher during the winter than the summer. This may be a result of agricultural runoff and leaching from the rainwater. Typically, the closer pesticide application is to a heavy or sustained rainfall, the greater likelihood that some pesticide leaching will occur. During the dry season there is probably high photodegradation due to a higher intensity of sunlight. Through personal interviews, it was found that pesticide application and timing and pesticide characteristics were similar among vegetable growers. It therefore unlikely that these factors could account for the differences in organophosphate concentrations between the summer and winter seasons.

The results of the present study showed that organophosphate (chlorpyrifos, dicrotophos, profenofos and ethion) residues were detected in most of the surface water samples from the agricultural area. People who lived in the agricultural communities could therefore be exposed to organophosphates by ingestion of contaminated water. People in the vegetable farm areas had a greater exposure to organophosphates, which were in high use on the farms. Importantly, there was extreme rainfall in September. During this peak period of rainfall, the increase in ethion and profenofos concentrations in this area was similar to that which occurred in the application period, despite the event occurring two months after the last application.

Regarding soil, insecticides are mainly applied on the plants, but they can also reach the soil. On the other hand, nematicides and herbicides are applied directly to the soil. Organophosphates remain active from a few hours to a few months in soils and crops, but they may leave residues in agricultural products (Dimitra *et al*, 2008). The rate of degradation in soil depends on soil type, soil moisture, adsorption, pH, soil temperature, concentration of pesticide, microbial activity and photodecomposition (Dimitra *et al*, 2008). It is known that a higher organic content of the soil is related to a greater binding of pesticide to soil and thus a greater persistence of pesticides. Also, the higher the soil acidity, the longer it takes for an organophosphate to be degraded. Because different types of organophosphate have different properties, it is clear that each of them should be evaluated on its own merits, and no extrapolation of results can be made from one type of pesticide to another. One pesticide may be easily decomposed, while another may be strongly adsorbed on soil. Some leach out easily and may reach groundwater. In these processes, the soil type and water solubility are of great importance. Furthermore, it should be recognized that not only the parent compound should be considered, but the breakdown products or metabolites also have to be taken into account.

Pesticides are currently regarded as some of the most dangerous environmental contaminants because of their stability, mobility and long-term effects on living organisms (Chaigarun and Nathapindhu, 2006). Pesticides can be absorbed and disseminated by air and rainfall, and their metabolites can likewise accumulate. Pesticide contamination in ambient air, surface water and soil has an adverse effect on people's health in communities located near cultivation areas. Pesticide residues in the air can be spread by the wind, and contaminated surface water and soil can enter nearby streams, which are the primary sources of drinking water for local residents. Children may be exposed to pesticide residues from swimming in the water, and fish and other aquatic organisms may also be contaminated.

In conclusion, the results from this study showed that chlorpyrifos, propinofos and ethion residues were detected in ambient air, surface water and soil samples in both summer and winter, but dicrotophose was only detected in winter. Mean organophosphate concentrations in vegetable areas were higher in winter than in summer. This might be a result of agricultural runoff and leaching in rainwater. Typically, the closer pesticide application is to a heavy or sustained rainfall, the greater is the likelihood that some pesticide leaching will occur. In summer, there was probably high photodegradation of pesticides due to a higher intensity of sunlight. Pesticide application, timing and pesticide characteristics were similar among vegetable growers and did not influence the differences in organophosphate concentrations during the summer and winter seasons.

In general, farmers follow a weekly calendar of spraying with "cocktails" of insecticides which are specially formulated for high-value vegetable crops. Multiple pesticides were detected in all vegetable areas. There were four pesticides detected in total. In these circumstances, control systems and improvement of the work environment are necessary in cultivation areas and surrounding communities. This should include training and promotion in the use of personal protective equipment. Farmers should strictly follow safety guidelines at work in order to prevent occupational diseases resulting from pesticide use. Because pesticides can break down and enter the environment, farmers should read and follow all label instructions and mix only the necessary amount of the product they intend to use. Pesticide drift is commonly thought to occur only when applications are not done properly, and pesticides can drift away from the target areas, harming people or property. Laws and regulations governing pesticide application were written with this kind of illegal, harmful drift in mind.

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