

## **ORGANOPHOSPHATE PESTICIDE RESIDUES IN FOOD COMMODITIES IN SRI LANKA: A REVIEW**

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### **ABSTRACT**

Organophosphates (OP) are the group of pesticides with the highest acute mammalian toxicity that is used in Sri Lanka. There are about nine active ingredients of OP registered in the country with two of them namely; dimethoate and fenthion are in the process of being phased out. The entire OP active ingredients registered in Sri Lanka except for fenamiphos belong to the hazard class II of the classification of the World Health Organization. Of the total importation of pesticides, OP contributes to about 10%, with chlorpyrifos being the most frequently imported OP insecticide. The OP pesticides are recommended to control a variety of insect pests in rice, vegetable and fruit cultivation. Literature reveals that OP are causing most of the intentional poisonings in Sri Lanka. The residues of OP, mostly chlorpyrifos, are reported in many food commodities and also in water sources.

**KEYWORDS:** Pesticides, Organophosphates, Residues, Intentional poisoning

### **INTRODUCTION**

Pesticides play a major role in controlling a variety of pests in agricultural crops in Sri Lanka. There are several hundred recommendations on pesticide usage for different crop-pest combinations (DOA, 2009). There are a range of active ingredients used and herbicides, insecticides and fungicides have 34, 46 and 27 active ingredients (a.i.), respectively, registered for use in Sri Lanka. These a.i. are registered under 525 product names out of which 463 are marketed in the country at present. About 3880 mt of pesticides formulations and 382 mt of technical material were imported for pest control purposes in agriculture during the year 2009 (DOA, 2009).

#### **Pesticides regulations**

All the activities from importation to usage of pesticides in Sri Lanka are regulated by the Office of the Registrar of Pesticides, under the Control of Pesticides Act No. 33 of 1980 and its amendment No. 06 enacted in 1994. This requires that any of the pesticide products in use within the country should be registered under the Act. The pesticide formulations included in the WHO Hazard Class Ia, Ib (Table 1; WHO, 2010) and the organochlorine

pesticides including persistent organic pollutants (POP) are banned in the country under the conditions of a gazette notification (Gazette No.1190/24, 2001). At present, the highest toxic class of pesticide formulations registered for regular use in Sri Lanka is classified under the WHO Hazard Class II.

**Table 1. WHO Recommended Classification of Pesticides by Hazard**

WHO Class	LD50 for the rat(mg/kg body weight)	
	Dermal	Oral
Ia - Extremely hazardous	<5	<50
Ib - Highly hazardous	5-50	50-200
II - Moderately hazardous	50-200	200-2000
III - Slightly hazardous	Over 2000	
U - Unlikely to present acute hazard	5000 or Higher	

(Source: WHO, 2010)

### Inappropriate use

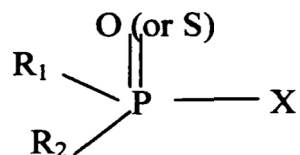
Despite the knowledge on risks of handling pesticides, farmers tend to apply pesticides without proper protective measures. They find the personal protective measures are inconvenient, such as to wear in the hot and humid climate, cost, limited availability and interference with work (Chandrasekara *et al.*, 1989, Van der Hoek *et al.*, 1998). Investigations have shown that the incidences of overexposure are common within spraying situations and overdosing can take place due to not following the label instructions on safety (Chandrasekara *et al.*, 1989; Nugaliyadde *et al.*, 2001). Application of pesticides to harvested fruits and vegetables and harvesting before completing the pre-harvest interval (PHI) also contribute to pesticide exposure (Chandrasekara *et al.*, 1989). The overuse of pesticides can result in high residue levels in commodities and in the immediate environments such as soil, biota and aquatic systems.

### Organophosphate pesticides

The organophosphates (OP) are organic compounds derived from acids containing phosphorus (Ecobichon, 1982). These compounds are used as pesticides, plasticizers, oil additives, lubricants and warfare agents. Among the OP pesticides are insecticides, herbicides, fungicides and others (Eto, 1974). All the OP insecticides are esters, amides or thiol derivatives of the pantavalent phosphorus acid. Despite the specificity as insecticides, many of them are responsible for posing acute and chronic toxicities in humans (Ecobichon, 1982).

The general structure of an OP compound is given in Figure 1. The chemical, physical and biological properties of OP compounds vary according to the R<sub>1</sub>, R<sub>2</sub> and X groups attached to the phosphorus atom. The R<sub>1</sub> and R<sub>2</sub>

are simple alkyl or aryl groups and X (the leaving group) can be aliphatic, aromatic or heterocyclic (IPCS, 1986).



**Figure 1. General Structure of Organophosphorus Compound**

### **Organophosphate mode of action - Acetylcholinesterase (AChE) inhibition**

The OP compounds with easily displaceable X groups are good insecticides or rather good cholinesterase inhibitors (Ecobichon, 1982). The reason behind this is their ability to phosphorylate the acetylcholinesterase (AChE) enzyme in the nervous system of insects generating an insecticidal activity. The AChE is one of the hydrolytic enzymes for neurotransmitter acetylcholine, which is found at the synaptic vesicles of the nerve endings. At an impulse transmission between synapses, acetylcholine released by the vesicle is rapidly hydrolysed by AChE before the second impulse comes. Therefore the inhibition of AChE affects the nervous function which leads to severe and often lethal damage in the organism (Eto, 1974).

### **Occupational exposure to pesticides in Sri Lanka**

Occupational exposure to pesticides in Sri Lanka has been recorded in many instances (Jeyaratnam *et al.*, 1982; Jeyaratnam *et al.*, 1987; De Alwis, 1989; Jeyaratnam *et al.*, 1990; Sivayoganathan *et al.*, 1995; Van der Hoek, 1998; Aponso and Manuweera., 2002; Smit *et al.*, 2003). The majority of the poisonings have been caused by the organophosphates (Jeyaratnam *et al.*, 1982; Eddleston, 2005; Ministry of Healthcare & Nutrition, 2006).

### **Organophosphate insecticides used in Sri Lanka**

Nine active ingredients of OP insecticides are registered under the Control of Pesticides Act No.33 of 1980 and two of them, dimethoate and fenthion, have been cancelled registration with effect from 31<sup>st</sup> December 2010 due to the health hazards posed on humans. The active ingredients of these OP are included in the WHO Hazard class II except for fenamiphos which is classified as class Ib (Table 2).

**Table 2. Organophosphate pesticides registered in Sri Lanka**

<i>Pesticide</i>	<i>WHO Classification of Active Ingredient</i>
Acephate	II
Chlorpyrifos	II
Diazinon	II
Fenamiphos	Ib
Phenthoate	II
Quinalphos	II
Dimethoate	II
Fenthion	II

(Source: DOA, 2009c; WHO 2010)

The OP pesticides are recommended for a variety of insect pest combinations in rice, vegetables and fruits (Table 3). Figure 2 shows the statistics of typically imported OP formulations from 2005 to 2009 (DOA 2006, 2007, 2008a, 2009a, 2010). Chlorpyrifos had the highest amounts of importation compared to other OP formulations. The amounts of dimethoate decreased during the period due to its phasing out program decided by the Office of the Registrar of Pesticides. Diazinon and phenthoate were imported as formulations as well as the technical grade (Figure 3) in order to formulate within the country.

**Table 3. Crop pest combinations in Sri Lanka where OP pesticides are recommended**

<i>Crop</i>	<i>Pest</i>
Rice	Thrips, gall midge, leaf folder, stem borer, paddy bug, case worm, mole cricket
Beans and Pulses	Bean fly, legume pod borer
Cabbage, Radish, Knolkhol and Cauliflower	Leaf eating caterpillars
Potato	Green peach aphid
Cucurbits	Melon fly, gall fly
Chilli	Leaf curl complex
Onion	Thrips, caterpillars
Sesame	Leaf webber
Mango	Fruit fly, leaf hopper, seed weevil, leaf cutting weevil
Pineapple	Mealy bug
General pests	Aphids, scales & mealy bugs, cut worm, root rating ants, termites

(Source: DOA, 2009c)

In 2009, about 40% of the total volume of insecticide formulations and 10% of the total pesticide formulations imported to Sri Lanka were OP (Table 4).

Table 4. Annual Imports of Pesticides (Broad Category) in 2009

Category	Technical Material (mt or kL)	Formulations (mt or kL)
Insecticides	107 (23-OP)	1037 (420-OP)
Herbicides	275	2745
Fungicides	0.25	600

(Source: DOA, 2010)

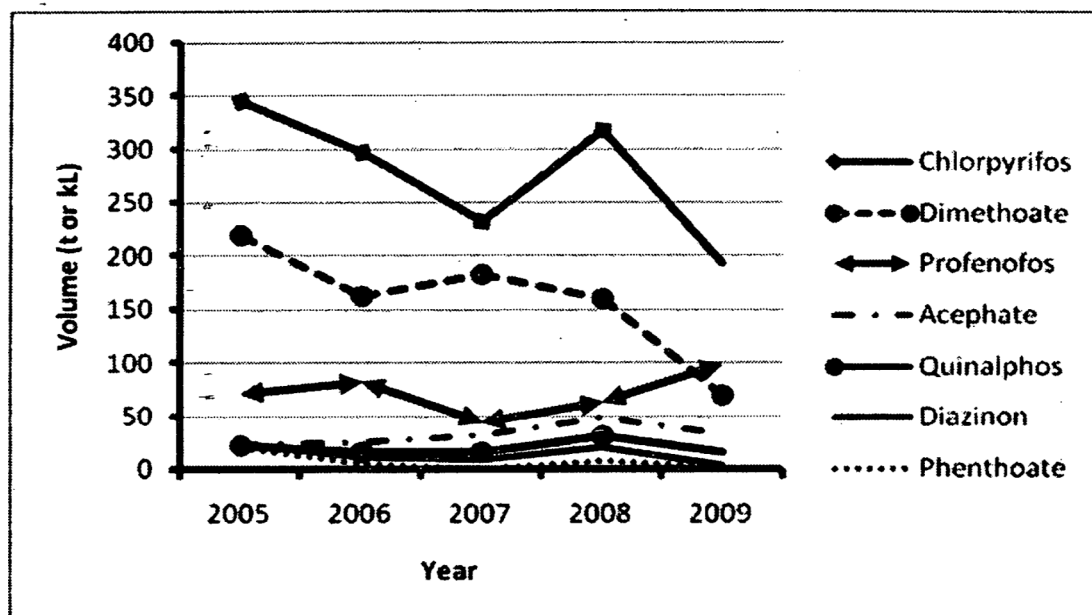


Figure 2. Annual Imports of OP formulations in Sri Lanka

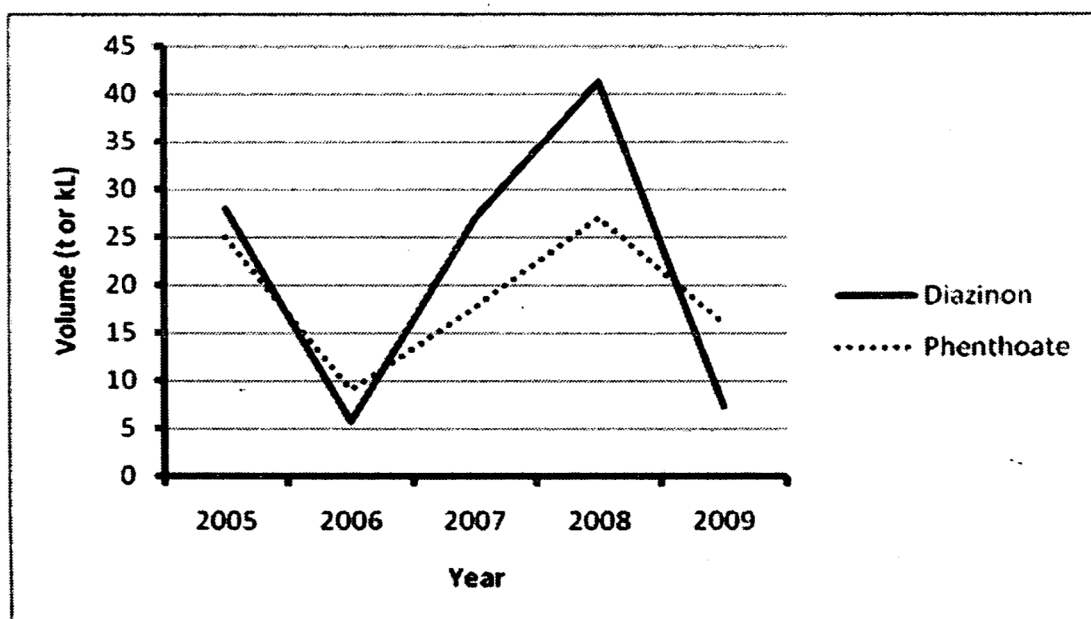


Figure 3. Annual Imports of OP technical grade in Sri Lanka

## Organophosphate residues in food commodities

Chlorpyrifos residues were reported in raw rice, vegetables and water sources in Sri Lanka during the period 2001-2009 (Tables 5 and 6). Samples for the above studies have been collected from many parts of the country and have covered several agricultural instructional divisions or/and 'grama niladhari divisions' (GND). The cabbage (*Brassica oleracea*) and bean (*Phaseolus vulgaris*) samples were obtained from up country vegetable growing areas in Nuwara-eliya, Badulla and Kandy Districts while mukunuwenna (*Alternanthera sessilis*) samples were from four Assistant Government Divisions in the Colombo district representing twenty four GNDs. Rice (*Oryza sativa*) samples were collected from research plots of the Department of Agriculture in Gannoruwa in the Kandy district. Residue data reported for water (Aponso *et al.*, 2003; Aravinna *et al.*, 2005; DOA, 2003; Eramudugolla, 2002; Bandara, 2007) were from various sources in intensive agricultural areas in the dry and wet zones in the country. They represent reservoirs, protected wells and unprotected wells which are used for drinking, bathing and irrigation. The sample analyses have been carried out in different laboratories. The data were presented as individual values (De Alwis *et al.*, 2006; DOA, 2003, 2009b; Eramudugolla, 2002; Bandara, 2007) and in ranges (Aravinna *et al.*, 2008) or graphs (Aponso *et al.*, 2003).

The limits of quantifications of chlorpyrifos in each food commodity are given in Table 5. The percentage of detection of residues in cabbage, beans, mukunuwenna and rice were 20, 13, 15 and 80, respectively (Table 5). Rice samples showed a higher proportion of detections compared to the others. The chlorpyrifos detection levels in water ranged in between  $10^1$  to  $10^6$  ng/L (Table 6). The detection limits for water analyses were not available. Only 22 water samples were reported during the study period, as positive with chlorpyrifos (3.5% of the total water samples).

Four out of twenty two detections in leafy mukunuwenna exceeded the Maximum Residue Limit (MRL) of  $5 \times 10^5$  ng/kg, which is set by the Sri Lanka Standard Institution (De Alwis *et al.*, 2006, Figure 4). Four of the cabbage samples and four of the bean samples also have exceeded the respective MRLs of  $1 \times 10^6$  ng/kg and  $1 \times 10^4$  ng/kg established by the Codex Alimentarius Committee (Aravinna *et al.*, 2008, Figure 4). None of the rice samples exceeded the Codex MRL (Joint FAO/WHO Codex Alimentarius Commission, 1995) of  $5 \times 10^6$  ng/kg (Figure 4). Three of the water samples out of twenty two detections exceeded the WHO Guideline Value (GV) for drinking water (Table 6).

**Table 5. Chlorpyrifos Residues in Rice and Vegetables**

<i>Dietary component (Number detected/ Number analyzed)</i>	<i>Chlorpyrifos-Range of detections (Minimum and Maximum) (10<sup>3</sup> ng/kg)</i>	<i>LOQ (10<sup>3</sup> ng/kg)</i>	<i>Reference</i>
Rice (12/15)	29 -417	20	DOA, 2009b
Mukūnuwenna (22/144)	14-1942	10	De Alwis, <i>et al.</i> , 2006
Cabbage (18/90)	10-5000	10	Aravinna <i>et al.</i> , 2008
Beans (7/54)	3.3-100	N/A	Aravinna <i>et al.</i> , 2008; DOA, 2009b
Lettuce (1/30)	10-50	4	Aravinna <i>et al.</i> , 2008
Leeks (2/129)	100-500	10	Aravinna <i>et al.</i> , 2008
Egg plant (1/2)	102	N/A	DOA, 2009b
Green chilli (1/3)	2.6	N/A	DOA, 2009b
Snake gourd (3/3)	5-101	N/A	DOA, 2009b
Green chilli* (8/42)	0.8-12	N/A	DOA, 2008b
Beans* (12/52)	0.4-14.7	N/A	DOA, 2008b
Cabbage* (3/48)	0.2-1.2	N/A	DOA, 2008b
Snake gourd*(7/28)	0.1-1	N/A	DOA, 2008b
Egg plant*(2/41)	0.9-1.4	N/A	DOA, 2008b

N/A- Not available; LOQ- Limit of Quantification; \* Detected in surface washings of the food commodity

The residues of profenofos, diazinon and dimethoate have been detected in various raw dietary components in Sri Lanka (Table 8). Profenofos has been the second highest OP in frequency of detections. Profenofos also showed a wider detection range from 5000 (excluding surface washings) to  $5 \times 10^6$  ng/kg. Many of the diazinon detections were reported in surface washings of the food commodities. Dimethoate was the least detected OP insecticide. In water sources, profenofos, diazinon and dimethoate were reported in some samples from 3.6 ng/L to 150 ng/L (Table 8).

The past information on residues of OP indicates that there is a relationship between the frequency of detection of chlorpyrifos (Table 5) and its imported amounts (Figure 2).

The residues were reported in different ranges in each food commodity (Table 5, Figure 4).

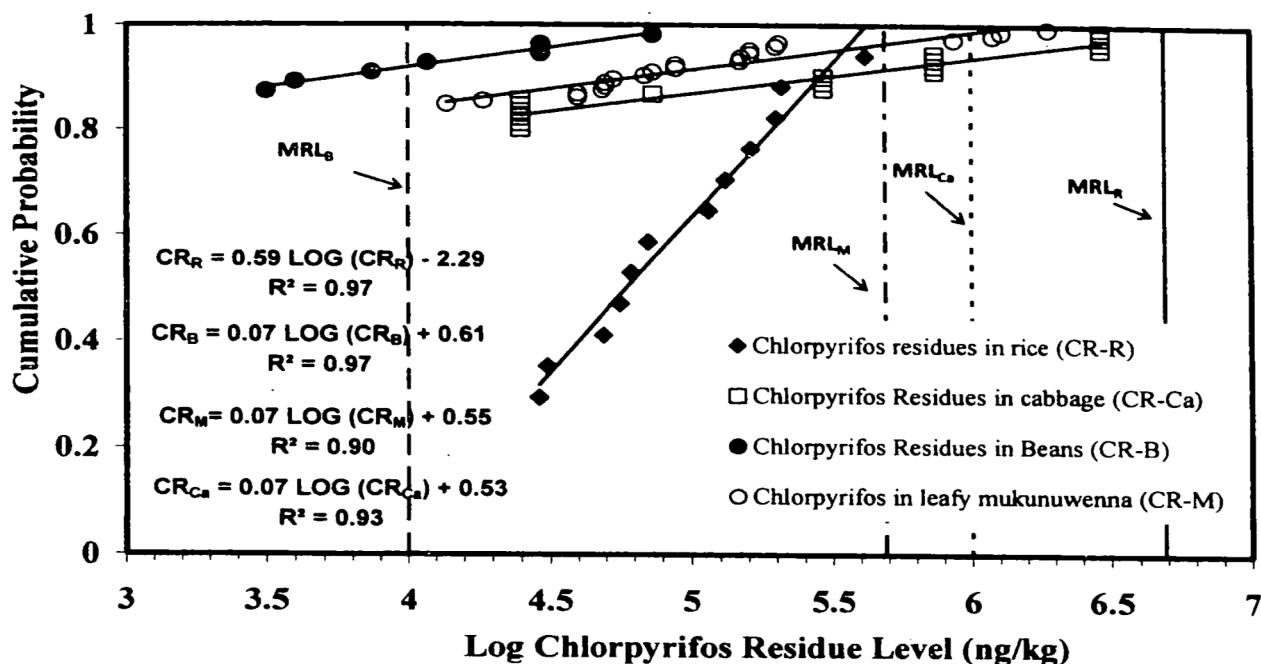


Figure 4. Chlorpyrifos Residues in Dietary Components in Sri Lanka

The wider distribution ranges (e.g. cabbage) are reflections of the variations in detection levels occurred due to many reasons. One reason could be the possible differences in agricultural practices carried out in areas where the samples were collected. For example, the method, frequency and the concentration of pesticide application could be different from one farmer to the other especially with lower levels of adherence to the pesticide recommendations, which is supported by the detection of residues in cabbage and mukunuwenna for which chlorpyrifos is not recommended for pest control under Sri Lankan conditions. The variation of the detection levels could also be due to the time spent from the last application to the time of chlorpyrifos analyses, which is unknown.

Moreover, the aspects of the analyses and different analytical methods practiced by different analysts could also have an impact on the detection levels. For example, chlorpyrifos residues reported in rice were analyzed in a study carried out for special research purposes (DOA, 2009b) and hence there is a possibility to detect the residues in a higher number of samples within a narrow range of concentrations ( $10^4$ - $10^5$  ng/kg, Table 5, Figure 4) with the effect of chlorpyrifos treatment carried out necessarily at the same rate with the whole crop. However, the number of treatments and the interval between pesticide application and sampling were reported to be different for each replicated rice culture plot. It was also noted that the residue analysis in rice

was carried out at the time of harvesting in which the residue levels in rice could still be high. Dimethoate, profenofos and diazinon residues in food commodities (Table 7) were reported less frequently compared to chlorpyrifos. One possible reason assumed for this is the lower amounts of importation and subsequent less usage of these products within the country. Less data were reported on OP residues in water bodies.

**Table 6. Chlorpyrifos Residues in Water**

Source of water	Chlorpyrifos- Range of detections ( $10^3$ ng/L)(Number detected/Number analyzed)	Reference	Guideline Value ( $10^3$ ng/L)
Reservoirs (DZ)	0.03-0.1 (3/544)	Aponso <i>et al.</i> , 2003	30.0
Reservoirs (WZ)	0.02-0.03(3/12)	DOA, 2003	(WHO, 2004)
Wells (DZ)	0.04-6.4 (12/14)	Eramudugolla, 2002	
Wells (WZ)	0.7 (1/60)	Aravinna <i>et al.</i> ,2005	
Reservoirs (DZ)	1450-6770 (3/5)	Bandara, 2007	

DZ - Dry zone, WZ - Wet zone

Apart from the expected analytical differences discussed above, it can be assumed that indirect contamination as another reason for less frequency of detection of residues in water. Among the reported data, a few unusually high levels of chlorpyrifos have been reported from catchment areas of reservoirs in an agricultural area with heavy use of pesticides. Yet the solubility of chlorpyrifos in water at 25°C is 2 mg/ L (WHO, 2004). Therefore the unusually high levels of chlorpyrifos indicate either analytical error or that the water samples have been taken from area where chlorpyrifos was directly contaminated with water.

**Table 7. Profenofos, Diazinon and Dimethoate Residues in Rice and Vegetables**

Pesticide	Dietary component (Number detected/ Number analyzed)	Range ( $10^3$ ng/kg)	LOQ ( $10^3$ ng/kg)	Reference
Profenofos	Mukunuwenna(5/144)	11-50	10	De Alwis <i>et al.</i> , 2006
	Beans (3/51)	5-50	8	Aravinna <i>et al.</i> , 2008
	Lettuce (3/30)	500-5000	16	Aravinna <i>et al.</i> , 2008
	Cabbage(3/90)	10-5000	20	Aravinna <i>et al.</i> , 2008
	Leek (2/129)	50-100	20	Aravinna <i>et al.</i> , 2008
	Green chilli* (2/48)	0.1-12	N/A	DOA, 2008b
	Beans* (2/52)	1.2-5	N/A	DOA, 2008b
Diazinon	Rice (7/16)	12-103	10	DOA, 2009b
	Mukunuwenna(4/144)	12-90	10	De Alwis <i>et al.</i> , 2006
	Green chilli* (1/42)	2	N/A	DOA, 2008b
	Beans* (3/52)	1.2-1.6	N/A	DOA, 2008b
	Snake gourd* (1/28)	0.8	N/A	DOA, 2008b
Dimethoate	Egg plant* (4/41)	0.8-1.7	N/A	DOA, 2008b
	Beans* (1/52)	0.8	N/A	DOA, 2008b
	Snake gourd* (1/28)	0.5	N/A	DOA, 2008b

\* Detected in surface washings of the food commodity; N/A= Not available

**Table 8. Profenofos, Diazinon and Dimethoate Residues in Water**

<i>Pesticide</i>	<i>Source of water</i>	<i>Range in water (ng/L) (Number detected/Number analyzed)</i>	<i>Reference</i>
Profenofos	Reservoirs	3.6 – 73 (7/12)	DOA, 2003
	Wells	10 (1/60)	Aravinna <i>et al.</i> , 2005
Diazinon	Reservoirs	12- 150 (4/544)	Aponso <i>et al.</i> , 2003
Dimethoate	Reservoirs	14 - 52 (1/544)	Aponso <i>et al.</i> , 2003

## CONCLUSIONS

During the period 2001-2009, 31 rice samples, 1043 vegetable samples and 554 surface washings of vegetables were reported to be analysed for OP with, 19, 125 and 15 detections of residues respectively. Chlorpyrifos, profenofos and diazinon were the most frequently detected organophosphates in rice and vegetables with concentrations in the range of  $2.6 \times 10^3$  to  $5.0 \times 10^6$  ng/kg,  $5.0 \times 10^3$  to  $5.0 \times 10^6$  ng/kg and  $1.2 \times 10^4$  to  $1.0 \times 10^5$  ng/kg, respectively. Dimethoate was not reported except for in the surface washings of vegetables. Out of 1795 sample analyses carried out for water, 35 samples reported with OP residues. The detections contained chlorpyrifos in the range of 20 to 6400 ng/L although a few unusually high levels were observed up to  $10^6$  ng/L, profenofos (3.6 to 73 ng/L) and diazinon (12 to 150 ng/L). The detections in vegetables exceed the maximum residue levels in 1% of the samples. This indicates that, either the pesticides were applied at higher rates than recommended or the crops were harvested before completion of the pre-harvest interval. Also, use of pesticides on un-recommended crops is evidenced by presence of OP residues in the leafy vegetable, mukunuwenna. Using past data it can be concluded that food commodities, particularly vegetables in Sri Lanka could contain pesticide residues at low levels but could reach unacceptable levels occasionally. Therefore, it is recommended that regular residue monitoring is necessary to identify the areas and crops contaminated with unacceptable residue levels which ultimately would contribute to alter the use pattern of pesticides to minimize the dietary exposure. For this it is important to select more sensitive analytical techniques and equipment to reach the low limit of quantification (LOQ) than defined MRL while using the accreted laboratories for pesticide residue analysis to ensure the reliability of analytical results.

The reported data for rice is lacking compared to the consumption rate in the country. Therefore more analyses at different sampling intervals are needed to understand the residue status. Although water samples in potential areas contained low levels of residues, the less frequency of detections indicates the contamination is low.

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