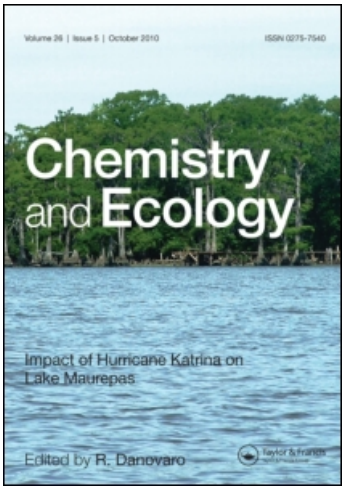

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PERSISTENT OF ORGANOCHLORINE PESTICIDES IN THE GREEN LIP MUSSEL (*PERNA Viridis*) FROM MARINE ESTUARIES IN THAILAND

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Thailand has benefited from the availability of pest control chemicals for improving agricultural products and in public health like any other developing countries. In order to understand the distribution and contamination in the Gulf of Thailand, a Mussel Watch programme has been set up to monitor trace toxic substances of organochlorine pesticides along the Gulf of Thailand since 1989.

This paper focuses on the concentration of organochlorine pesticides in green lip mussels (*Perna viridis*) during 1989 to 1996. The mussel samples were used as a biological indicator and analyzed for 23 organochlorine compounds. The highest frequencies of organochlorine pesticide residues in Thailand were found in aldrin, dieldrin and DDTs in all samples and the highest concentration of DDT-isomer were p,p'-DDE and p,p'-DDT during 1989–1990 and 1991. The p,p'-DDE was only found during 1993–1995.

In 1996 the level of the DDT isomer (o,p-DDE) were found in the range of 11–16 ppb (ng g^{-1} wet weight), HCH isomer (α-HCH) was found ND – 1.6 ppb (ng g^{-1} wet weight) and *cis*-chlordane found at ND – 3.4 ppb (ng g^{-1} wet weight). Other organochlorine pesticide compounds such as dieldrin, heptachlor, aldrin, endrin and endosulfan isomers were not detected. The detection limit of all organochlorine compounds were 1.3–8.2 ppb (ng g^{-1} wet weight). Trends of organochlorine pesticide residue in this area were found differently from the previous data during 1989–1991.

Keywords: Organochlorine; distribution; biological indicator; monitoring

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INTRODUCTION

The usage of pesticides in developing countries are related to early stages with agricultural development of the pest control activities and in public health, chemicals continue to play a significant role; this trends will be sustained for many years to come. Although pesticides are poisons, the majority of people recognize the advantages of their use in view of the benefits they bring to the control of disease vectors and nuisance pests particularly the increase of food and fibre production. Many developing countries are lacking still of appropriate pesticide legislation and adequate mechanism to enforce regulation effectively.

Organochlorine pesticides are used extensively in developing countries because they are inexpensive, effective and persistent. Pesticides, primarily insecticides because of their acute toxicity to mammals and other vertebrates, and their ability to persist (Ramesh, 1991) and accumulate in organisms (Perry *et al.*, 1983; Schmitt *et al.*, 1985) have induced serious effects on wildlife and livestock.

Thailand, like other developing countries, has benefited from the availability of pest control chemicals and has also produced its share of pesticides poisoning. Amount of some pesticides have been decreased as shown in Table I, but the total load of pesticide residues has increased over the past ten years.

Monitoring of trace toxic substances in the aquatic environment using green lip mussels (*Perna viridis*) as biological indicators are used commonly because of their many advantages such as the wide

TABLE I Quality (a.i) of import pesticides (1985–1996)

	Quality tons				
Year	*Insecticide*	*Fungicide*	*Herbicide*	*Others*	*Total*
1985	5,146	2,646	4,830	210	12,832
1986	5,799	2,512	4,262	204	12,777
1987	5,881	4,530	3,967	247	14,625
1988	7,050	4,362	5,596	205	17,213
1989	6,937	4,724	6,747	317	18,725
1990	7,176	2,800	8,272	346	18,594
1991	5,560	2,087	7,071	311	15,029
1992	6,098	3,513	8,450	418	18,479
1993	5,305	3,988	9,056	476	18,825
1994	5,252	4,885	9,554	640	20,331
1995	6,573	4,828	11,934	727	24,062
1996	6,608	4,446	14,041	446	25,541

geographical distribution, and immolibity, and easy to sample. The organs of mussels can accumulate chemicals from the surrounding water, and in water itself, as a state in the mussel watch concept has been favoured (Tanabe *et al.*, 1987; Phillip, 1980; O'Connor *et al.*, 1992).

In order to assess the current status of the contamination of organochlorine pesticides residue were transferred to the environment along the Gulf of Thailand, where the mussel watch programme was established since 1989. This information will be useful in establishing the guideline for coastal resources management plan for the future.

MATERIALS AND METHODS

The green lip mussels (*Perna viridis*) have been collected at 13 stations along the Gulf of Thailand from 1989–1996 as shown in Figures 1 and 2. The samples were cleaned and removed carefully to avoid any contamination. The samples were kept frozen at −20°C until analysis. Mussel tissues were homogenized from all the mussels, about 200 g in each sample. The mussel samples were analyzed for 23 compounds of organochlorine pesticides such as heptachlor, endrin, dieldrin, aldrin, HCH-isomers and DDT-isomers.

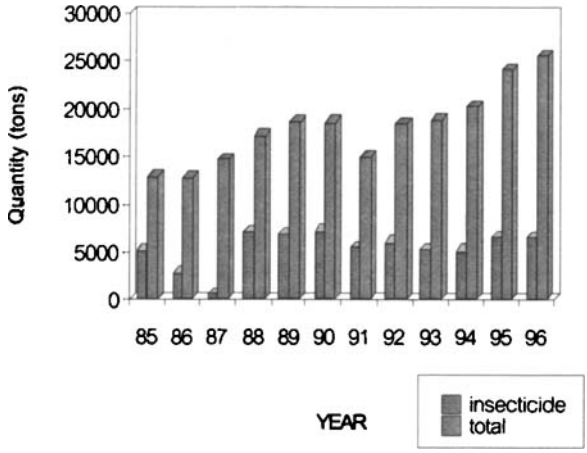

FIGURE 1 Comparison of quality (a.i) of insecticides and total import pesticides during 1985–1996.

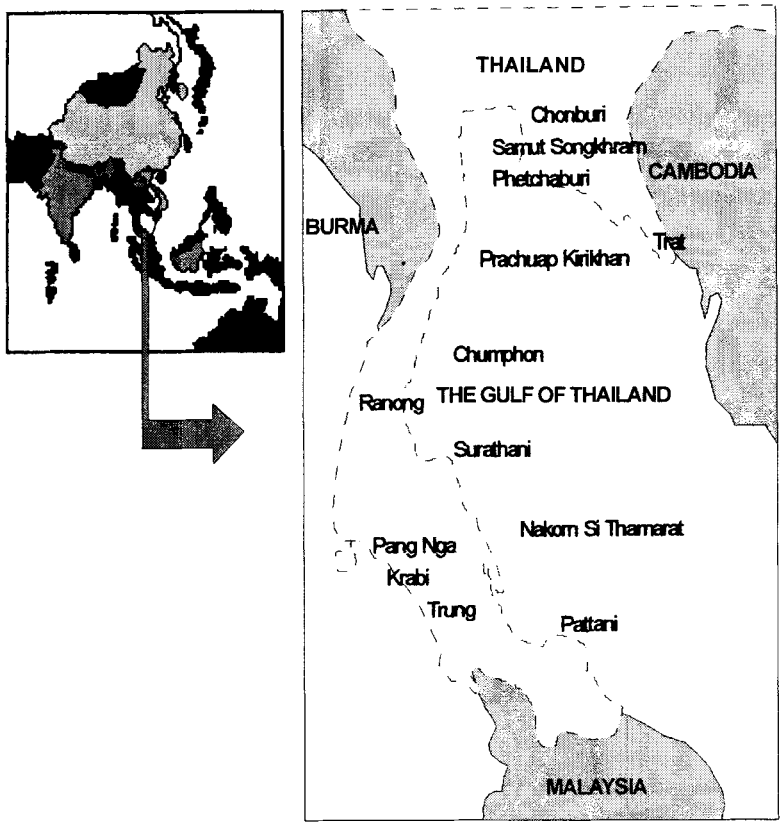

FIGURE 2 Sampling Location of Green Mussels from the Gulf of Thailand. Station Number and Location (Province) 1. Samut Songkhram, 2. Phetchaburi, 3. Prachuap Kirikhan, 4. Chumphon, 5. Nakhon Sri Thammarat, 6. Trad, 7. Thrung, 8. Chonburi, 9. Suratthani, 10. Ranong, 11. Pung-ga, 12. Pattani, 13. Krabi.

Mussel samples of 10 grams were extracted with 35 ml acetone and 10 ml hexane by using homogenization, centrifugation and filtration. The mussel residues were extracted again with hexane : diethyl ether (9 : 1). The filtrate was merged and extracted with 50 ml sodium chloride in 0.1 N ortho-phosphoric acid. The organic layer was concentrated until dryness to determine the fat content. Organochlorine pesticides were cleaned and eliminated from the fat by using Florisil dry column and Florisil column chromatography clean-up and to separate the PCBs and organochlorine pesticides with 100 ml hexane and 200 ml 20% diethyl ether in hexane.

Quantification of organochlorine pesticides residues were performed by gas chromatography (Hewlett Packard 5890) equipped with a Ni 63 Electron Capture Detector. The fused silica capillary column was OV – 1701 (30 m length × 0.25 mm i.d., 0.25 film thickness of stationary phase) The temperature programme is 100°C for 1 min to 180°C at the range of 10°C min^{-1}, held 1 min and increased by 1°C to 200°C, and the last step is increased by 1°C to 220°C and held for 20 mins. The carrier gas was hydrogen (flow rate 2 ml min^{-1}) and make up gas used the nitrogen (flow 40 ml min^{-1}). The detector and injector temperature were 300°C and 220°C.

RESULTS AND DISCUSSION

The yearly concentration of organochlorine pesticides and PCBs in green lip mussel from the Gulf of Thailand during 1989 – 1996 were reported on a wet weight basis. Percentage of fat contents were 1.0 to 2.4% and moisture contents were 80 – 87%. Method detection limits are shown in Table II.

The organochlorine pesticides compounds during 1989 – 1990 such as aldrin, dieldrin, HCH-isomers and DDT-isomers were found at all stations. The range of total of DDT-isomers (o,p- and p,p-DDD, DDE, DDT) concentration in 1989 and 1990 were 0.94 – 5.1 ng g^{-1} and 0.74 – 5.4 ng g^{-1} (Cherdhan Siriwong *et al.*, 1991). The concentration of DDT-isomers ranged from 0.74 – 5.38 ng g^{-1}. Although the use of DDT for agricultural purposes was banned in 1983, it is being used

TABLE II Information of organochlorine pesticides and method detection limit (MDL)

Organochlorine pesticides	*MDL* (ng g^{-1})	*Organochlorine pesticides*	*MDL* (ng g^{-1})	*Organochlorine pesticides*	*MDL* (ng g^{-1})
HCB	1.0	Endosulfan-α	5.5	o,p-DDE	2.7
Heptachlor	0.6	Endosulfan-β	2.0	p,p′-DDE	3.8
Heptachlor-epoxide	2.5	Endosulfan sulphate	3.2	o,p-DDD	9.3
Aldrin	1.4	α-HCH	0.8	p,p′-DDD	3.8
Endrin	4.2	β-HCH	1.5	o,p′-DDT	7.8
Dieldrin	2.8	γ-HCH	2.4	p,p′-DDT	6.0
tran-chlordane	2.4	δ-HCH	1.4	methoxychlor	8.2
cis-chlordane	2.9	oxychlordane	2.8		

still for a malaria vector control by the Ministry of Public Health (Suthep Ruangwises *et al.*, 1994). Trends of concentration of DDT-isomers in 1989 – 1996 are shown in Table III.

Hexachlorobenzene (HCB) is produced as a by-product material of several chlorinated hydrocarbons such as the production of tetrachloro-ethylene, trichloro-ethylene, chlorine, vinyl chloride. In 1989 – 1990 HCB was found at the range of ND – 0.21 ng g^{-1}.

Aldrin and dieldrin which were used as an insecticide against termites and to be converted. Dieldrin has been used extensively in the past as an insecticide for corn and termite control. Although it was no longer registered for general use, the concentration of aldrin were found in 1989 and 1990 at the range of 0.34 – 1.3 ng g^{-1} and 0.20 – 0.62 ng g^{-1} and dieldrin at the range of 0.08 – 0.48 ng g^{-1} and 0.08 – 0.22 ng g^{-1}. The trend of aldrin and dieldrin from 1989 to the present has been decreased due to the product of aldrin and dieldrin were banned in 1983 as shown in Table IV.

In case of HCH-isomers, α-HCH and γ-HCH were found at relatively low concentration in a few mussel samples. The concentration of α-HCH and γ-HCH ranged from < 0.02 – 0.06 ng g^{-1} and < 0.02 – 0.04 ng g^{-1} in 1989. (Siriwong *et al.*, 1991) γ-HCH was found at 3.9 ng g^{-1} at Pattani station in 1994. The β-HCH was found in the range of 22 – 26 ng g^{-1} at Suritthari and Patchap Kirikhan stations in 1994, while α-HCH were 1.6 ng g^{-1} Nakhon Sri Thammarat in 1996.

TABLE III Trend of organochlorine pesticide residues during 1989 – 1996

Year	*Trend of organochlorine pesticide (not DDTs)*	*Trend of organochlorine pesticide (DDTs)*
1989	DDTs > aldrin > dieldrin > HCB > heptachlor > HCHs	p,p′-DDE > p,p′-DDT > p,p′-DDD > o,p′-DDD > o,p′-DDT > o,p′-DDE >
1990	DDTs > dieldrin > aldrin > HCHs	p,p′-DDE > o,p′-DDE p,p′-DDT > p,p′-DDD > o,p′-DDT > o,p′-DDD
1991	DDTs > aldrin > dieldrin > heptachlor	p,p′-DDT > p,p′-DDE > p,p′-DDD > o,p′-DDD > o,p′-DDE > o,p′-DDT
1993 – 1995	β-HCH α-HCH > δ-HCH > endosulfan II	p,p′-DDE > o,p′-DDE > o,p′-DDT
1996	cis-chlordane & α-HCH	o,p-DDE

References of the data in 1989 : Siriwong *et al.*, 1991, 1990 : Tabucanon *et al.*, 1990, 1991 : Ruangwises *et al.*, 1994, 1992 – 1996 : Boonyatumanond *et al.*, 1996.

TABLE IV Organochlorine pesticides banned and/or restricted under the Ministry of Agriculture and Cooperatives, Thailand

Chemical	*Effect year*
HCHs	1980
endrin	1981
DDTs	1983
aldrin	1983
endrin	1983
toxaphene	1983
heptachlor	1988

Endosulfan is used as an insecticide against a variety of insects on a variety of crops. Technical endosulfan is composed of α-endosulfan and β-endosulfan. In 1994 the organochlorine pesticides were imported only 3 compounds (chlordane, lindane, and endosulfan) from other countries such as U.S.A., Germany, Israel and South Korea. The concentration of endosulfan and isomer residues were found at the range of ND$-5.7\,\mathrm{ng\,g^{-1}}$ at Trad in 1994.

In 1995 the concentration of organochlorine pesticides were found in only 2 out of 23 compounds. The γ-HCH and p,p′-DDE were found at the range of $0.21-0.82\,\mathrm{ng\,g^{-1}}$ and $0.09-0.32\,\mathrm{ng\,g^{-1}}$. The trend of organochlorine pesticides residues during 1989–1996 are shown in Table III.

Relation of frequency of organochlorine pesticides residues in green lip mussels during 1993–1996, p,p′-DDE was the highest level found frequently among organochlorine pesticides. The major source of p,p′-DDE were from metabolic transformation of p,p′-DDT to p,p′-DDE isomer under oxidation conditions.

The methoxychlor is a one of pesticides that we used and the quality of import is increased. We have just monitored the methoxychlor since 1994, but we never found that in the mussel samples.

The Royal Thai Government banned many organochlorine pesticide compounds for use in agriculture since 1980 as shown in Table IV, except DDT was used mainly for mosquito control in some areas where malaria disease could occur from 1988–1997 as shown in Table V. At present Department of Communicable Disease Control, Ministry of Health, has used delta-methrin, lambda-cyhalothrin and permetrin instead of DDT since 1995. However, the concentrations of residues were lower than the maximum residue limit (MRL) for

TABLE V The use of DDT 75% Water Disperse Powder (WDP) for malaria vector control in 1988–1997

Location (*provinces*)	*Amount of DDT 75% used (kg)*							
	1988	1989	1990	1993	1994	1995	1996	1997
Phetchaburi	14480	16665	15145	2476	1917	4468	3038	3024
Prachuap Kiri Khan	19565	20630	3390	4432	4083	0	0	1555
Chumphon	17982	19565	20630	22092	8161	4332	0	0
Nakhon Si Thammarat	11828	28834	18655	11816	7812	4101	8938	6226
Pattani	7875	12285	28385	0	0	0	0	0
Trung	**	**	**	7016	5241	432	400	257
Ranong	**	**	**	8967	6668	5505	5025	4960
Chonburi	**	**	**	1880	1494	3193	0	1770
Trad	16715	5075	6090	5523	3957	1893	1552	1768
Suratthani	**	**	**	26272	18905	13739	10334	8952
Krabi	**	**	**	**	**	0	0	1336
Pung-ga	**	**	**	**	**	11504	1815	2062

Ref: from Malaria Division (1988–1997), Department of Communicable Disease Control, Ministry of Public Health, Thailand.
** Not available.

aquatic animals as recommended by Ministry of Health *i.e.*, DDTs in 5 mg kg^{-1} food and aldrin 0.1 mg kg^{-1} food.

The data from some locations indicated the trend of decreased spraying amount of DDT by 75% Water Disperse Powder (WDP) for controlling mosquito in Thailand since 1988–1997 as it is shown in Table V.

CONCLUSIONS

The trend of organochlorine pesticides concentration of the green lip mussel (*Perna viridis*) were indicated that the number of the most persistent of organochlorine compounds were lower than the early data (1989–1991) and it remained still for some of them to metabolize to p,p′-DDE. It seems to be that contamination sites were decreased because the government banned and restricted use under the management plan for control. The mussel watch monitoring programme can provide the information to understand the status of contamination of organochlorine pesticides in this area and to enforce legislation and some management of a toxic substance in the future.

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