

การสะสมของสารตกค้างกลุ่มออร์กาโนคลอรีนในน้ำ ดินตะกอนและสัตว์ไม่มีกระดูกสันหลัง ณ คลอง 7
พื้นที่เกษตรกรรมรังสิต จังหวัดปทุมธานี



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ACCUMULATION OF ORGANOCHLORINE RESIDUES IN WATER, SEDIMENT AND
AQUATIC INVERTEBRATES AT KHLONG 7, RANGSIT AGRICULTURAL AREA,
PATHUM THANI PROVINCE



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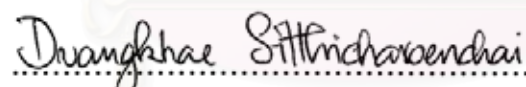
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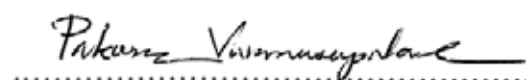
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ศึกษาสารตกค้างยาฆ่าแมลงกลุ่มออร์กาโนคลอรีน ได้แก่ สารกลุ่มบีเอชซี เฮปตาคลอ อัลดริน
ดีดีที เอนโดซัลแฟน เอ็นคริน และเมทอกซีคลอร์ โดยใช้แกสโครมาโตกราฟี ในตัวอย่างน้ำ ดินตะกอน
และสัตว์ไม่มีกระดูกสันหลังในน้ำ ได้แก่ กุ้งฝอย หอยเชอรี่ และหอยขม ณ คลอง 7 พื้นที่เกษตรกรรมรังสิต
จังหวัดปทุมธานี โดยทำการเก็บตัวอย่างตั้งแต่เดือนมิถุนายน 2547 ถึงเดือนพฤษภาคม 2548 ค่าเฉลี่ยความ
เข้มข้นของสารตกค้างยาฆ่าแมลงกลุ่มออร์กาโนคลอรีนในน้ำที่ตรวจพบมีค่าตั้งแต่ 0.004 ถึง 0.08
ไมโครกรัมต่อลิตร ในดินตะกอนมีค่าตั้งแต่ 0.16 ถึง 14.67 ไมโครกรัมต่อกิโลกรัมน้ำหนักแห้ง ในกุ้งฝอยมี
ค่าตั้งแต่ 2.08 ถึง 53.04 ไมโครกรัมต่อกิโลกรัมน้ำหนักเปียก ในหอยเชอรี่มีค่าตั้งแต่ 6.77 ถึง 47.83
ไมโครกรัมต่อกิโลกรัมน้ำหนักเปียก และในหอยขมมีค่าตั้งแต่ 5.39 ถึง 79.61 ไมโครกรัมต่อกิโลกรัม
น้ำหนักเปียก สารเอ็นโดซัลแฟนมีปริมาณความเข้มข้นเฉลี่ยสูงสุดในน้ำ (0.08 ไมโครกรัมต่อลิตร) ส่วนใน
ดินตะกอนพบว่าสารเฮปตาคลอมีความเข้มข้นเฉลี่ยสูงสุด (14.67 ไมโครกรัมต่อลิตร) และพบสารดีดีทีที่มี
ความเข้มข้นเฉลี่ยสูงสุดในตัวอย่างสัตว์ไม่มีกระดูกสันหลังทั้ง 3 ชนิด (47.83 53.04 และ 79.61 ไมโครกรัม
ต่อกิโลกรัมน้ำหนักเปียก ตามลำดับ) จากค่าสัดส่วนระหว่างความเข้มข้นในสัตว์ต่อในน้ำพบว่าสารกลุ่ม
ดีดีทีที่มีปริมาณการตกค้างสะสมคิดเป็น 2,483 เท่าในกุ้งฝอย 2,754 เท่าในหอยเชอรี่ และ 4,133 เท่าในหอย
ขม ส่วนค่าสัดส่วนระหว่างความเข้มข้นของสารกลุ่มดีดีทีในสัตว์ต่อในดินคิดเป็น 4.0 เท่าในหอยเชอรี่ 4.4
เท่าในกุ้งฝอย และ 6.6 เท่าในหอยขม การสะสมตกค้างของสารฆ่าแมลงกลุ่มออร์กาโนคลอรีนมีการตรวจ
พบมากในหอยขมซึ่งน่าจะมาจากพฤติกรรมการกินอาหารของหอยขมซึ่งเป็นผู้กินซาก และรูปแบบการ
ดำรงชีพอยู่หน้าดิน ด้วยเหตุนี้หอยขมจึงมีแนวโน้มที่จะได้รับยาฆ่าแมลงผ่านทาง การซึมซับและการกิน
มากกว่าหอยเชอรี่และกุ้งฝอย ซึ่งสองชนิดหลังมักอาศัยอยู่บริเวณผิวน้ำเป็นส่วนใหญ่ อย่างไรก็ตามระดับ
สารตกค้างยาฆ่าแมลงกลุ่มออร์กาโนคลอรีนในสัตว์ไม่มีกระดูกสันหลังที่ตรวจพบยังจัดอยู่ในระดับที่
ยอมรับได้ โดยเทียบกับระดับสารตกค้างที่ยอมให้พบได้มากที่สุดซึ่งกำหนดโดยโคเดกซ์และกระทรวง
สาธารณสุขประเทศไทย

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PREMKAMOL THONGKONGOWM : ACCUMULATION OF ORGANOCHLORINE RESIDUES IN WATER, SEDIMENT AND AQUATIC INVERTEBRATES AT KHLONG 7, RANGSIT AGRICULTURAL AREA, PATHUM THANI PROVINCE. THESIS ADVISOR : DUANGKHAE SITTHICHAROENCHAI, Ph.D. THESIS COADVISOR : ASST. PROF. KUMTHORN THIRAKHUPT, Ph.D., 90 pp. ISBN 974-14-2099-4.

Organochlorine pesticides residues (OCPs) such as Σ BHCs, Σ Heptachlor, Σ Aldrin, Σ DDTs, Σ Endosulfans, Σ Endrin, and methoxychlor collected from water, sediment, and aquatic invertebrates such as Lanchester's freshwater prawn *Machrobrachium lanchesteri*, apple snail *Pomacea* sp., and freshwater snail *Filopaludina martensi* from Khlong 7, Rangsit agricultural area, Pathum Thani province from June 2004 to May 2005, were measured using gas chromatography-micro electron capture detector (GC- μ ECD). The means of minimum and maximum concentrations of OCPs in water, sediment, *M. lanchesteri*, *Pomacea* sp., and *F. martensi* were 0.004 - 0.08 μ g/L, 0.16 - 14.67 μ g/kg dry weight, 2.08 - 53.04 μ g/kg dry wet weight, 6.77 - 47.83 μ g/kg dry weight, and 5.39 - 79.61 μ g/kg dry weight, respectively. The highest amount of OCPs were Σ Endosulfans (0.08 μ g/L) in water samples, Σ Heptachlors (14.67 μ g/kg dry weight) in sediment samples, and Σ DDTs in invertebrate samples (47.83, 53.04, and 79.61 μ g/kg wet weight in *Pomacea* sp., *M. lanchesteri*, and *F. martensi*, respectively). For the OCPs accumulations, the invertebrate-water ratios of Σ DDTs were 2,483, 2,754, and 4,133 folds in *Pomacea* sp., *M. lanchesteri*, and *F. martensi*, respectively, whereas the invertebrate-sediment ratios were 4.0, 4.4, and 6.6 folds in *Pomacea* sp., *M. lanchesteri*, and *F. martensi*, respectively. Accumulations of OCPs were mostly found in *F. martensi* more than in *Pomacea* sp. and *M. lanchesteri*. The higher OCPs accumulation in *F. martensi* might be explained by its feeding behavior as a scavenger and its benthic life form. Therefore, *F. martensi* tends to expose to pesticides, by both absorption and ingestion, higher than *Pomacea* sp. and *M. lanchesteri* which prefer to live mainly at the littoral zone. However, the levels of OCPs in all aquatic invertebrates were in acceptable limit recommended by the Codex Alimentarius Commission and the Ministry of Public Health in Thailand.

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NOMENCLATURES

AOAC	Association of Analytical Communities
AR	Analytical grade
ASE	Accelerated solvent extractor
ATSDR	Agency for Toxic Substances and Disease Registry
CNS	Central nervous system
ECD	Electron capture detector
FAO	Food and Agriculture Organization
GC	Gas chromatography
IARC	International Agency Research on Cancer
LD	Lethal dose
LOD	Limit of detection
LOQ	Limit of quantitation
MRLs	Maximum residue limits
ND	Not detectable
OC	Organochlorine pesticide
OCPRs	Organochlorine pesticide residues
PE	Polyethylene
ppm	Part per million
PR	Pesticide grade
SOP	Standard Operating Procedure
SPE	Solid phase extraction
USEPA	United States Environmental Protection Agency

CHAPTER I

INTRODUCTION

1.1 Statement of problems

Thailand is one of the agricultural countries in Asia. Greater than 53 percents of the total land area, approximately 513,000 square kilometers of land, is agriculture. The most important four food crops in terms of the planted area and the value are rice, maize, sugarcane, and cassava. The productions are both for domestic consumption and exportation (Poblap and Silkavute, 2001). In view of the fact that the central plain is a flush and fertile valley suitable for agriculture, it is the most extensive rice-producing area in the country. Rice which is the most important agricultural product is grown over 50 percents of the farmland around the country (Office of Agricultural Economic, 1992). The immense serious problems in producing are rice diseases and pests that destroy plants. Extremely, farmers want their yields as much as possible. As a result, more pesticides are widely applied to control pests for increasing crop yields.

During 1994-1998, 38.6 percents of imported chemicals in Thailand were for agricultural sectors. In 1998, the total amount of 2.9 million tons of chemicals was imported for agricultural uses, 2.8 million tons (98.9%) of fertilizers and 0.032 million tons (1.1%) of pesticides. In 1999, the total amount of imported pesticides increased to 0.041 million tons (Poblap and Silkavute, 2001). In 2003, the total amount of imported pesticides was 0.08 million tons (Imported Toxic Substances Report, 2003) which increased two-fold.

Even though the use of pesticides has increased crop production and other benefits, it has raised concerns about potential adverse effects on the environment and human health. In many regards, the greatest potential for involuntary adverse effects of pesticides is through contamination of the aquatic system such as ponds, canals, rivers, and streams which supports aquatic life and related food chains, recreation, drinking water, irrigation, and many other purposes.

Currently, contamination in aquatic system has been a significant problem in the Asian countries. The primary concern pesticides up until the present in sediment, water, and aquatic biota have been the organochlorine pesticides which were heavily used as an agricultural pesticide and as malaria repellent.

Pesticides are toxic to living organisms in many ways and one of the concerning substances is organochlorine pesticides. The reason behind this is that they are water insoluble and present in the environment for a long time. Moreover, organochlorine pesticides, being with slow chemical and biological degradation, can be concentrated in organisms and biomagnified along the food chain (Ahlborg *et al.*, 1992). Although some organochlorine pesticides such as DDT, dieldrin, and lindane, have been banned in Thailand for more than ten years, most recent studies have reported that these pesticide residues can still be detected in crops, water, sediment and biota of many aquatic ecosystems (Pipithsangchan *et al.*, 1997; Thapinta and Hudak, 2000).

For more than 50 years, pesticides of several kinds have been applied to the Rangsit Great Plain or Rangsit agricultural area, especially the organochlorine pesticides. This toxic substance had been heavily used in this area during 1950-1990 and some still being used till the present day. The rice growing system in paddy fields of the Rangsit Great Plain is a type of agricultural practice which requires pump-in water from the canal at the beginning of cultivation and the water will be released to the canal before harvesting. Therefore, the organochlorine pesticide could contaminate the canal and its components via the released water. Moreover, each paddy field can be cultivated three times a year with continuous use of pesticides. As a result, it is expected that the contamination of OCPR could be very high in water and sediment as well as in aquatic invertebrates such as mollusks, shrimps, crabs, and aquatic insects.

At present, the Rangsit Great Plain has increasingly become human settlement and the water from the irrigation canal has been used for many purposes. Local people health risks associated with using contaminated water and consuming animals and plants in the canal, have never been evaluated. This study was aimed to provide

information for ecological risk assessment and proper management in the future. Khlong 7 canal, the one of 14 Rangsit sub-canals, which is situated at the middle of Rangsit irrigation system in Nong Sua district, Pathum Thani province, was selected as a representative area in this study.

1.2 Objectives

1.2.1 To examine OCPR concentrations in water, sediment, and some aquatic invertebrates at Khlong 7, Rangsit agricultural area, from June 2004 to May 2005

1.2.2 To compare the differences in OCPR concentrations among water, sediment, and each aquatic invertebrate species collected from different study sites and in different seasons.

1.2.3 To assess the risk of consuming some edible invertebrates by compare with the maximum residue limits (MRLs).

1.3 Hypotheses

1.3.1 Although some types of organochlorine pesticides have been banned for several years, some residues still persist in water, sediment, and some aquatic invertebrates.

1.3.2 The concentrations of OCPR in parameters measured among study sites and seasons are different.

1.3.3 All concentrations of OCPR found in edible invertebrates are in the acceptable limit.

1.4 Scope of the study

1.4.1 Examination was on the OCPR concentration cited in standard pesticide catalog No. 47913 which includes 17 kinds of organochlorine pesticides such as α -BHC, β -BHC, γ -BHC, δ -BHC, heptachlor, aldrin, heptachlor epoxide, endosulfan I, endosulfan II, endrin, dieldrin, 4,4'-DDE, 4,4'-DDD, 4,4'-DDT, endrin aldehyde, endosulfan sulfate and methoxychlor.

1.4.2 The study area was situated along 20 kms of the Khlong 7 canal, Rangsit agricultural area, Pathum Thani province.

1.4.3 Aquatic invertebrate samples were identified at least to the genus level.

1.4.4 The samples were collected once a month for one-year period.

1.5 Anticipated benefits

The results of this study provide information on the level of organochlorine pesticide contamination in aquatic ecosystem of Khlong 7, Rangsit agricultural area, Pathum Thani province. It can also provide information for the ecological risk assessment. Furthermore, the result can enhance proper public awareness and management in this study area.



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CHAPTER II

BACKGROUND AND LITERATURE REVIEWS

2.1 Study area

Topographically, the Rangsit agricultural area is a wide expanse of lowland. It is bounded by the Chao Phraya River on the western side and the Nakhon Nayok River on the northeastern side, with a network of many irrigation canals inside the Great Plain that have been connected with the purposes to irrigate the whole agricultural areas and to protect Bangkok Metropolis from annual flooding by acting as a main water-holding area. Its main portion comprises more or less waterlogged fields which are suitable habitats for many forms of aquatic life. Rangsit agricultural area is regarded as one of productive spots of fishery resources in the central plain of Thailand (National Science Museum, Ministry of Science and Technology, 2001).

The Rangsit agricultural area comprises 14 sub-canals (Khlung) which receive water from Raphi Phat canal, north of Pathum Thani province. The water from these sub-canals flows to Rangsitprayulasakdi canal which transfers water to the portion of the lower Chao Phraya River, north of Bangkok.

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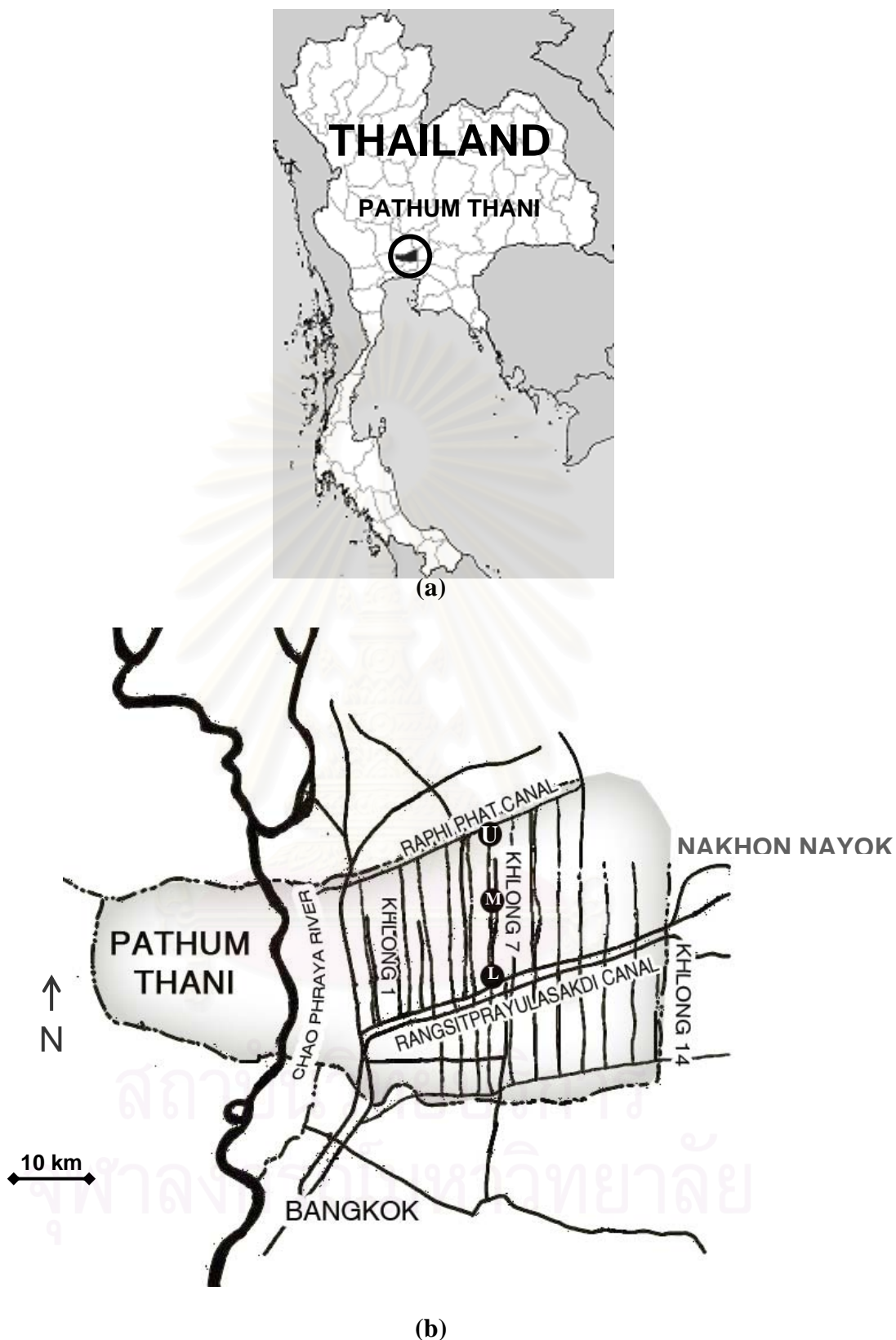


Figure 2.1 Study area and sampling sites investigated from June 2004 to May 2005: (a) location of Pathum Thani province in central Thailand (b) three sampling sites at Khlong 7, Pathum Thani province (U: upper stream, M: middle stream, and L: lower stream).

(Adapt from: th.wikipedia.org/wiki/จังหวัดปทุมธานี)

2.2 Organochlorine pesticides

2.2.1 Organochlorine pesticide groups

Organochlorine pesticides are insecticides composed primarily of carbon, hydrogen, and chlorine. They break down slowly and can remain in the environment long after application and in organisms long after exposure. Organochlorine pesticides are diverse group of agents belonging to three distinct chemical classes including the dichlorodiphenylethane-, the chlorinated cyclodiene-, and also the chlorinated benzene- and cyclohexane-related structure (Table 2.1). From the mid-1940s through the mid-1960s, these compounds were used extensively in all aspects of agriculture and forestry, in building and structural protection, and in human situations to control a wide variety of insect pests. The low volatility, chemical stability, lipid solubility, slow rate of biotransformation, and degradation properties that made these chemicals such effective insecticides also brought about their demise because of their persistence in the environment, bioconcentration, and biomagnification within various food chains. The acquisition of biologically active body burdens in many wildlife species that, if not lethal, undoubtedly interfered with the reproductive success of the species (Klaassen, 1996).

Table 2.1 Structural classification of organochlorine pesticides

Organochlorine pesticide group	Chemical structure	Examples
Dichlorodiphenylethanes		DDT,DDD, DDE, Dicofol, Perthane, Methoxychlor, Methlochlor
Cyclodienes		Aldrin, Dieldrin, Heptachlor, Chlordane, Endosulfan
Chlorinated Benzenes Cyclohexane		BHC, HCH, Lindane (γ -BHC)

Source: Klaassen, 1996

2.2.1.1. Dichlorodiphenylethanes

DDT (Dichlorodiphenyltrichloroethane or 1,1,1-trichloro-2,2-bis(*p*-chlorophenyl) ethane) and its related compounds such as methoxychlor, dicofol, perthane, and methlocholor are the examples of this group. DDT is one of the best known insecticides and it was once widely used to control insects on agricultural crops and insects that carry diseases like malaria and typhus, but it now used in only a few countries to control malaria (ATSDR, 2002). DDT is semi-volatile and can be expected to partition into the atmosphere as a result. It is lipophilic and partitions readily into the fat of all living organisms and has been verified to bioconcentrate and biomagnify. The breakdown products of DDT, 1,1-dichloro-2,2-bis(4-chlorophenyl)ethane (DDD or TDE) and 1,1-dichloro-2,2bis (4-chlorophenyl)ethylene (DDE), are also present almost everywhere in the environment and are more persistent than the parent compound. DDT and its metabolites may enter the air when they evaporate from contaminated water and soil. DDT, DDE, and DDD in the air will then be deposited on land or surface water. This cycle of evaporation and deposition may be repeated many times. As a result, DDT, DDE, and DDD can be carried long distances in the atmosphere.

Some DDT can enter the soil from waste sites. DDT, DDE, and DDD may occur in the atmosphere as a vapor or attach to solid particles in the air. Vapor phases of DDT, DDE, and DDD may be broken down in the atmosphere due to reactions caused by sunlight. The half-life of these chemicals in the atmosphere as vapors (the time it takes for one-half of the chemical to turn into something else) has been calculated to be approximately 1.5-3 days. However, in reality, this half-life estimate is too short to account for the ability of DDT, DDE, and DDD to be carried for the long distances reported. DDT, DDE, and DDD have lasted in the soil for a very long time. Potentially, they may persist for hundred-year long. DDT is broken down slowly to DDE and DDD by the microbial degradation (ATSDR, 2002). These chemicals may also evaporate into the air and then be deposited in other places. They stick strongly to soil, and therefore, they generally remain in the surface layers of soil. Some soil particles with attached DDT, DDE, or DDD may get into rivers and lakes through runoff. Only a very small amount, if any, will seep into the ground and migrate into groundwater. The length of time that DDT will last in soil depends on

many factors including temperature, type of soil, and whether the soil is wet. DDT lasts for a much shorter time in the tropics where the chemical evaporates faster and where microorganisms degrade it faster. DDT disappears faster when the soil is flooded or wet than when it is dry. DDT also disappears faster when it initially enters the soil. Later on, the evaporation slows down and some DDT moves into small spaces in the soil particles where are very difficult for microorganisms to reach and break it down efficiently. In tropical areas, DDT, DDE, and DDD may disappear in much less than a year. In temperate areas, half of a deposit initially present usually disappears in about 5 years. However, in some cases, the half will remain for 20, 30, or more years (ATSDR, 2002). In surface water, DDT binds to particles in the water, then settles and deposits in the sediment. DDT is taken up by small organisms and fish in the water. It accumulates at high levels in fish and marine mammals such as seals and whales, reaching levels many thousand times higher than existing in the water. In these animals, the highest levels of DDT are found in their adipose tissues. DDT in the bottom sediment can also be absorbed by some water plants and by the aquatic animals which consume those plants. DDT metabolites can be transported through food webs to top consumers such as humans (Thirakhupt *et al.*, 2005).

The use of DDT has been banned in at least 34 countries and severely restricted in at least 34 other countries. The countries that have banned DDT include Argentina, Australia, Bulgaria, Colombia, Cyprus, Ethiopia, Finland, Hong Kong, Japan, Lebanon, Mozambique, Norway, Switzerland, and the USA. Countries that have severely restricted its use include Belize, Ecuador, the European Union, India, Israel, Kenya, Mexico, Panama, and Thailand. Other countries, not noted above, may also prohibit or severely restrict the use of DDT (Ritter *et al.*, 1995). DDT has an acute oral toxicity to mammals of 113-450 mg/kg for rats, 100-800 mg/kg for mouse, 250-560 mg/kg for guinea pig and 300-1,770 mg/kg for rabbit. Similarly, DDT has an acute dermal toxicity of 250-3,000 mg/kg for rat, 250-500 mg/kg for mouse, 100 mg/kg for guinea pig and 300-2,820 mg/kg for rabbit (WHO, 1989).

Methoxychlor is efficient against flies, mosquitoes, cockroaches, and a wide multiplicity of other insects. It is used on agricultural crops and livestock, including animal feed, barns, and grain storage bins. It does not readily evaporate into air or dissolve in water. Methoxychlor can accumulate in some living organisms including

algae, bacteria, snails, clams, and some fish. However, most fish and animals convert methoxychlor into other substances that are rapidly released from their bodies, thus methoxychlor does not usually build up in the food chain (Thirakhupt *et al.*, 2005).

2.2.1.2 Chlorinated cyclodienes

Cyclodiene insecticides are cyclic hydrocarbons having a chlorine substituted methanobridge structure. Cyclodiene insecticides were introduced into the western countries during the 1950s and were used in diverse formulations for many different purposes (Thirakhupt *et al.*, 2005). Aldrin, dieldrin, heptachlor, chlordane, and endosulfan are examples of this group. Aldrin, dieldrin, and endrin in various commercial formulations have many end-uses varying from public health schemes to crop protection and industry. Major uses are aldrin which is a broad spectrum insecticide primarily used for the control of a wide range of soil pests, grasshoppers, and certain cotton insects. Another are endrin which control a wide range of foliage pests of cotton, rice, tobacco, maize, sugarcane, and fruit trees such as cutworms, armyworms, aphids, corn borers, cabbage looper, grasshoppers, plant bugs, webworms, and many other pests. Moreover, endrin is particularly effective against caterpillars. Aldrin, dieldrin, and endrin are Shell-products. Chlordane is a similar chemical, but is of lower vertebrate toxicity. Endrin and endosulfan are very high toxicity in vertebrate, but limited biological persistence. Aldrin is readily metabolized to dieldrin by both plants and animals, so aldrin residues are rarely found in foods and animals and reside in small amounts. In general, the cyclodienes resemble DDT in being stable lipophilic solids of very low water solubility, but differ from it in their mode of action. Endosulfan is an exception to this rule, having appreciable water solubility. Due to their water insolubility, emulsifiable concentrates and wettable powders were the formulations normally used for spraying. Sprays were used for the control of certain crop pests and for vectors of diseases. They were also used in dips and sprays to control ectoparasites of livestock and were widely used as seed dressings for cereals and other crops. The use of aldrin, dieldrin, and heptachlor for the latter purpose has caused very serious ecological consequences through food chains and food webs including contamination in soil, water, and groundwater (Thirakhupt *et al.*, 2005).

Endosulfan is a man-made insecticide used for control of a number of insects on food crops such as grains, tea, fruits, and vegetables and on nonfood crops such as tobacco and cotton. It is also used as a wood preservative. Endosulfan is sold as a mixture of two different forms of the same chemical (referred to as alpha- and beta-endosulfan). Endosulfan enters air, water, and soil when it is manufactured or used as a pesticide. Some endosulfan in the air may travel long distances before it lands on crops, soil, or water. Endosulfan on crops usually breaks down within a few weeks. Endosulfan released to soil attaches to soil particles. Endosulfan found near hazardous waste sites is usually found in soil. Some endosulfan in soil evaporates into air and some endosulfan in soil breaks down. However, it may stay in soil for several years before it all breaks down. Rain water can wash endosulfan attached to soil particles into surface water. Endosulfan does not dissolve easily in water. Most endosulfan in surface water is attached to soil particles floating in the water or attached to soil at the bottom. The small amounts of endosulfan that dissolve in water break down over time. Depending on the conditions in the water, endosulfan may break down within 1 day or it may take several months. Some endosulfan in surface water evaporates into air and breaks down. Because it does not dissolve easily in water, only very small amounts of endosulfan are found in groundwater (water below the soil surface; for example, well water). Animals that live in endosulfan-contaminated waters can build up endosulfan in their bodies. The amount of endosulfan in their bodies may be several times greater than in the surrounding water (ATSDR, 2000).

2.2.1.3 Hexachlorocyclohexanes (HCH)

Hexachlorocyclohexane (HCH), formally known as benzene hexachloride (BHC), is a synthetic chemical that exists in eight chemical forms called isomers. The different isomers are named according to the position of the hydrogen atoms in the structure of the chemical (ATSDR, 2005). BHC has similar properties to other organochlorine insecticides, but it is 100 times more polar and water soluble than DDT. BHC is classified into alpha (α), beta (β), gamma (γ), and delta (δ) isomers. Technical grade HCH used in insecticide preparations contains a mixture of isomers: the γ - and α -isomers are convulsive poisons; the β - and δ -isomers are central nervous system (CNS) depressants (Klaassen, 1996). Emulsifiable concentrates of HCH have been used for controlling agricultural pests and parasites on farm animals. It has also

been used as an insecticidal seed dressing. HCH is moderately toxic to rats (LD_{50} 60-250 mg/kg). One of these forms, gamma-HCH (or γ -HCH, commonly called lindane), is produced and used as an insecticide on fruit, vegetables, and forest crops, and also animals and animal premises. This isomer of HCH (lindane) produces signs of poisoning that resemble those caused by DDT. Only the γ -isomer has seen in medical use today, as a component of a pediculicide shampoo for head lice (Klaassen, 1996).

Although technical-grade HCH is no longer used as an insecticide in the United States, α -, β -, γ -, and δ -HCH have been found in the soil and surface water at hazardous waste sites because they persist in the environment. In the air, the different forms of HCH can be present as a vapor or attached to small particles such as soil and dust. The particles may be removed from the air by rain or degraded by other compounds found in the atmosphere. HCH can remain in the air for long periods and travel great distances depending on the environmental conditions. In soil, sediments, and water, HCH is broken down to less toxic substances by algae, fungi, and bacteria, but this process can take a long time (ATSDR, 2005).



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2.2.2 Carcinogenic classification

Organochlorine pesticides are characterized by their low water solubility and high lipid solubility. They are noted for their environmental persistence, long half-lives and their potential to bioaccumulate and biomagnify in organisms once dispersed into the environment (Ritter *et al.*, 1995). As a result, these persistent compounds have the potential to cause significant adverse effects to human health by affect neural transmission.

International Agency Research on Cancer (IARC) classified chemicals into 5 groups as followed.

- Group 1 : Known human carcinogen
- Group 2A : Probable human carcinogen
- Group 2B : Possible human carcinogen
- Group 3 : Not classifiable for human carcinogenicity
- Group 4 : Probably not carcinogenic to humans

IARC has reviewed the study of carcinogenicity of organochlorine pesticides and has concluded that there is evidence for carcinogenicity of organochlorine pesticides in experimental animals, rats, as showed in the Table 2.2.

Table 2.2 Carcinogen classifications of organochlorine pesticides and their target organs.

Organochlorine Pesticides	IARC Carcinogen Classifications	Target organ
Aldrin	3	Liver
Chlordane, heptachlor	3	Liver
DDT	2B	Lung, liver
Dieldrin	3	Liver
Hexachlorocyclohexane (BHC)	2B	Liver, leukemia
Methoxychlor	3	Liver

Source: Pimsaman, 1997

2.3. Biology of aquatic samples collected in this study

The aquatic invertebrates collected in this study were Lanchester's freshwater prawn *Macrobrachium lanchesteri*, apple snail *Pomacea* sp. and freshwater snail *Filopaludina martensi* (Brandt, 1974). These species were selected because they frequently found all along the Khlong 7 throughout the year. Moreover, they are edible animals especially Lanchester's freshwater prawn and freshwater snail.

2.3.1 Lanchester's freshwater prawn

Taxonomy: Phylum Arthropoda

Subphylum Crustacea

Class Malacostraca

Subclass Leptostraca

Superorder Eucarida

Order Decapoda

Suborder Natantia

Section Caridea

Family Palaemonidae

Scientific name : *Macrobrachium lanchesteri*, De Man, 1991

General description : Lanchester's freshwater prawn, body length about 2-7 cm, is common seen in Thailand and Malaysia peninsula. They live in rice fields, swamps and canal. They are able to tolerate low oxygen contents, and salinity up to 20 ppm, and prefer the temperature from 26 to 36°C. They reproduce under pond conditions. They consume algae, microorganisms, zooplankton, and larvae of some aquatic organisms, however, they are human's food (Department of Fisheries, 2005).

2.3.2 Freshwater snail

Taxonomy: Phylum Mollusca,

Class Gastropoda,

Subclass Prosobranchia

Order Mesogastropoda

Family Viviparidae

Scientific name : *Filopaludina martensi*, Gray, 1847

Common name : Freshwater snail, river snail or pond snail

General description : This species inhabit on clay at the base of the pond and on drain with running water (FAO, 2000). This species is a common food of human.

2.3.3 Apple snail

Taxonomy: Phylum Mollusca

Class Gastropoda

Subclass Prosobranchia

Order Mesogastropoda

Family Ampullariidae

Genus *Pomacea*

Scientific name : *Pomacea* sp.

General description : During the 1980's, this introduced snails rapidly spread to Indonesia, Thailand, Cambodia, Hong Kong, southern China, Japan and the Philippines. However, apple snails are considered a delicacy in several regions and they are often sold in Oriental markets for consumption. Apple snails inhabit a wide range of ecosystems from swamps, ditches, and ponds to lakes and rivers. Most of them prefer lentic waters above turbulent water (rivers). Although they occasionally leave the water, they remain mainly submerged. Apple snails have separated sexes (gonochoristic). They are not selective and eat almost everything available in their environment. In general, they prefer soft and digestible vegetation. Tougher plants and algae

are consumed as long as they are able to grasp pieces of with their radula (rasp tongue). When there is not enough food available in the water, apple snails can profit from their amphibian life style to leave the water in search for food. The life cycle of apple snails is determined by the availability of food and the temperature of the water. At high temperatures and abundance of food, some apple snail species exhibit a very short life cycle of less than three months and are reproductive throughout the whole year. As apple snails are a popular food source for various animals like birds, turtles, fishes, insects, and crocodiles, it is not surprising that they have developed several techniques to avoid predation (Ghesquiere, 2003).



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2.4 Maximum Residue Limits (MRLs)

MRL is the maximum concentration of a pesticide residue (expressed as mg/kg or part per million, ppm), recommended by the Codex Alimentarius Commission to be legally permitted in food commodities and animal feeds. In Thailand, pesticides residues limits in food in Thailand are recommended by the Ministry of Public Health. The MRLs of OCPRs in aquatic animals are presented in Table 2.3.

Table 2.3 The MRLs (mg/kg) of organochlorine pesticide residues in aquatic animal recommended by the Codex Alimentarius Commission and the Ministry of Public Health in Thailand.

Organochlorine Pesticides	Maximum Residue Limits (mg/kg aquatic animal)	
	Codex ^a	Ministry of Public Health, Thailand ^b
1. Aldrin	0.3	0.1
2. BHC	-	0.5
3. DDT	5.0	5.0
4. Dieldrin	0.3	0.3
5. Endrin	-	0.3
6. Heptachlor and Heptachlor epoxide	0.3	0.3

Source: ^a Notification of Codex Alimentarius Commission

^b Ministry of Public Health No.71 (B.E.2525) issued under Food Act B.E.2522 (1979), published in the Royal Government Gazette (Special issued) Vol. 169, Part 168, dated November 1982.

2.5 Literature reviews

As a consequence of heavy application of organochlorine pesticides, many scientific studies have looked for residues in sediment, aquatic resources and aquatic biota. The occurrence of organochlorine pesticides varies in different sites depending on the sources of pesticides in the drainage and the characteristics of the aquatic systems such as water flow and the physical and chemical properties of each pesticide. In Turkey, for example, Barlas (1999) reported that the occurrence and concentrations of organochlorines varied among stations and seasons. However, Miglioranza *et al.* (1999) in Argentina considered that differences in height and temperature did not affect organochlorine pesticide concentration but the lack of clay material results in organochlorine pesticides concentration decreasing through the soil profile. In recent years, there has been an increased attentiveness of the potential hazards of pesticides in urban environments and demand for assessing risk for public health.

At present, contamination of aquatic environment by organochlorine pesticides is a critical problem throughout the world. The number of food chains in water and water environment obtain the major amounts of the chemicals and other substances (Caliskan, 2000). Organochlorine pesticides are responsible for negative ecological consequences to wildlife because of their biomagnification in the food web, reaching higher concentrations in the top predators (Barron *et al.*, 1995). Study on the limnology, plankton and biomagnification of organochlorine pesticides at Ignacio Ramirez Reservoir in Mexico by Favari *et al.* (2002) indicated that these pesticides were bioconcentrated 2- to 10-fold from water to algae, 10- to 25-fold in zooplankton, and 8- to 140-fold in fish. This result showed that the bioaccumulation of these contaminants in fish and biomagnification potential in humans are perceived as threats. Currently, there was the status of organochlorine contamination in various environmental media such as sediments, soils, and wildlife in China. DDT and its metabolites were the predominants in most media even though the use of DDTs has been officially banned in China since 1983. However, the concentration of *p,p'*-DDT

and ratio of *p,p'*-DDT to Σ DDTs were significantly higher in marine fishes than in freshwater fishes (Nakata, 2005).

In view of the fact that, benthic invertebrates serve as agents of contaminant transfer between sediment and higher trophic levels in aquatic systems (Menzie, 1980). Numerous researchers reported high concentration of organochlorine pesticides residues in mussels (Kauss and Hamdy, 1985; Kurt and Ozkoc, 2004), crabs (Caliskan, 2000; Mortimer, 2000) and aquatic insect larvae (Thornley and Hamdy, 1984). Several studies indicated that aquatic insects were inherently more susceptible than terrestrial insects (Tang, 1996). Nevertheless, some reports showed the low concentration in the soft tissue of the mussel collected from Lake Faro in Italy that this area is not at contamination risk from organochlorine compounds and besides is free from health problems for the consumer of mussel products (Licata *et al.*, 2004).

Thailand is one of agricultural countries. There have been a number of studies for organochlorine pesticide residues in many organisms and their environments. However, the adverse effect to organisms in the food chain and to humans in Thailand is not well evaluated. In 1989, Pimpan *et al.* analyzed bioaccumulation of organochlorine pesticide residues in water through food chains in 3 freshwater areas of Bueng Boraphed in Nakhon Sawan province, Nonghan in Sakon Nakhon province, and Kwanphayao in Phayao province. The results from water, sediment, aquatic plants, and aquatic animal samples indicated that 5 kinds of insecticides found in most samples were lindane, heptachlor, aldrin, dieldrin, and DDT including their derivatives. Dieldrin residue was found in all samples at high concentration ranged from less than 0.01 ppb to 0.138 ppm. In addition, they reported that the levels of dieldrin residue in sediment, aquatic plant, and aquatic animal samples were 517, 442, and 425 folds higher than those found in water samples, respectively. Organochlorine residues in fish samples were investigated by Chinda (1998) in the Chao Phraya River, the main water resource of Thailand. The results showed that the highest quantities of pesticides was detected in the lower part of the Chao Phraya River and contaminated to the middle and upper parts, respectively. The highest quantities of heptachlor and DDT and derivatives were observed in rainy season whereas dieldrin

was found highest in dry season. However, there was no correlation between the amounts of pesticides residue concentration in fish investigated in different seasons.

Jumruskul *et al.* (1999) studied the distribution of organochlorines in water and sediments in Mae Khlong River and Tributaries. The results showed that organochlorine pesticides in water samples were BHC, heptachlor, heptachlor epoxide, aldrin, dieldrin, dicofol, endrin, endosulfan, and its derivatives including DDT and its derivatives. Their concentrations were less than 0.01-1.65 $\mu\text{g/L}$. Consequently, the amounts of contamination in sediment samples were similar to water samples except endosulfan and its derivative. However, DDT and its derivatives were found in most samples, Noicharoen (2000) studied the accumulation of organochlorine pesticides in sediment and green mussel *Perna viridis* from Tachin estuary, Samut Sakhon province in Thailand. The three dominant organochlorine pesticides were detected such as total-DDT, total-BHC, and chlordane. Moreover, the result showed that the high level of contamination was in sediment affected to high level of pesticides residue in green mussel.

Aquatic organisms are important in the food web of terrestrial organisms such as fish, mollusk, shrimp, or crab as being consumed by people and wildlife. Besides the investigating of bioconcentration of organochlorine pesticide, risk assessments in the contaminated area have been studying. Actually, humans and animals have risked from these aquatic biota. The recent work of Satapornvanit *et al.* (unpublished manuscript) has used new method to assess risk of pesticide applications in central region of Thailand, in Pathum Thani province and Nakhonpathom province. The primary results showed that there was a very high risk that pesticide use could negatively affect the environment and ecology around the farms.

Thoophom *et al.* (1987) showed the concentrations in bivalves from the Upper Gulf of Thailand. The results indicated that DDT, dieldrin, BHC, endrin, and lindane were detected at the level not exceed 10 $\mu\text{g/kg}$. Siriwong *et al.* (1991) studied in Green mussel *Perna viridis* collected from the Gulf of Thailand in 1989, and found that DDTs, aldrin, and dieldrin were the predominant compounds at the levels did not exceed the maximum residue limit for aquatic animals as recommended by the Ministry of Public Health in Thailand. Kumblad *et al.* (2000) reported that the mean

Σ DDT concentrations in fish collected at different location from the Songkhla Lake, Thailand were below the recommended maximum residue levels in aquatic animals used for human consumption (5,000 ng/g fresh weight) in Thailand.

Additionally, risk assessment in human health have been undertaken worldwide to examine the potential health risk due to exposure to toxic contaminants in a variety of environmental media and food materials (NRC, 1993). For example, Stefanell (2004) estimated and monitored the organochlorine residue through the ingestion of edible fishes in Italy. Only *p,p'*-DDE and *p,p'*-DDD were found at the levels up to 25.00 ng/g wet weight. Estimated daily intake of organochlorine pesticides through edible fish was significantly lower than the relevant ADI. Moreover, the study of human health risk assessment of organochlorine pesticides by fish consuming in China showed that the concentrations of organochlorine pesticides ranged from 0.67 to 13 ng/g wet weight (ppb). The *p,p'*-DDE concentration (3.9 ng/g wet weight) found in fish meat was the significantly higher than the other organochlorine pesticides (Jiang *et al.*, 2005).

CHAPTER III

METHODOLOGY

3.1 Chemical reagents

3.1.1 Solvents and chemicals

- 3.1.1.1 Acetone (CH_3COCH_3), (pesticide grade, PR and analytical grade, AR)
- 3.1.1.2 Dichloromethane (DCM) (CH_2Cl_2), (PR)
- 3.1.1.3 Diethyl ether ($(\text{C}_2\text{H}_5)_2\text{O}$), (PR)
- 3.1.1.4 Hexane ($\text{CH}_3(\text{CH}_2)_4\text{CH}_3$), (PR)
- 3.1.1.5 Petroleum ether, (PR)
- 3.1.1.6 Ottawa sand from Applied Separatio
- 3.1.1.7 Granular anhydrous sodium sulfate (Na_2SO_4), (AR)
- 3.1.1.8 Copper powder, (AR)
- 3.1.1.9 Hydrochloric acid (HCl), (AR)
- 3.1.1.10 Distilled water

3.1.2 Standard chemicals

EPA 8080 Pesticides Mix Catalog No. 47913, consisting of aldrin (purity, 99.3%), alpha-benzene hexachloride (α -BHC) (purity, 99.8%), beta-benzene hexachloride (β -BHC) (purity, 98.9%), delta-benzene hexachloride (δ -BHC) (purity, 99.6%), dieldrin (purity, 99.2%), endosulfan I (alpha) (purity, 99.9%), endosulfan II (beta) (purity, 99.9%), endosulfan sulfate (purity, 99.9%), endrin (purity, 98.0%) , endrin aldehyde (purity, 98.6 %) , gamma-benzene hexachloride (γ -BHC) (purity, 99.9%), heptachlor, heptachlor epoxide isomer B (purity, 99.9%), methoxychlor (purity, 99.4%), 4,4'-DDD (purity, 98.5%), 4,4'-DDE (purity, 99.6%) and 4,4'-DDT (purity, 98.9%) from SUPLECO, were used.

3.2 Instruments and equipments

3.2.1 Sample containers for,

3.2.1.1 Water – Bottle-Polyethylene (PE), 1-L volume

3.2.1.2 Sediment – PE bags

3.2.1.3 Aquatic invertebrates – wrapped sample with aluminum foil and kept in PE containers filled with ice

3.2.2 Accelerated Solvent Extractor (ASE 100[®]) from Dionex

3.2.3 Gas Chromatography (GC) model 6890N with micro-Electron Capture Detectors (μ -ECDs) equipped with Agilent 6783 A 8-sample tray autosampler from Agilent Technologies. The data system is an Agilent Chemstation G2070AA operated with Windows Me and an Agilent Kayak XA, 350 MHz Pentium II computer workstation.

3.2.3.1 Primary column: DB-35MS (35% phenyl methyl siloxane, 30 m x 320 mm, 0.25 μ m film thickness)

3.2.3.2 Confirmation column: DB-1701 (30 m long \times 0.25 mm I.D. bonded fused silica column, 0.25 μ m film thickness)

3.2.3.3 Helium carrier gas flow was established at 44 cm/sec linear velocity and nitrogen is set at 60 mL/min as make-up gas.

3.2.3.4 The oven temperature was programmed from 100 °C to 280 °C at 12 °C/min and held for 10 min. Total run time was calculated to be 27.00 min. The injection port temperature was 260 °C and detector temperature was set at 300 °C. The injection volume was 1 μ L splitless mode with a 0.75-min vent delay.

3.2.4 Nitrogen evaporator (Turbo Vap II Concentration workstation, Zymark Corporation)

3.2.5 Solid-Phase Extraction (SPE) with 500 mg Florisil-PR Extract-Clean Columns (4.0 mL column size) from Alltech Associates, Inc.

3.2.6 Blender

3.2.7 2-mL GC amber vial

3.2.8 2-, 4-, and 5-mL vial-glass

3.2.9 Volumetric pipette

3.2.10 2-L separatory funnels

3.2.11 1-L graduated cylinders

3.2.12 Beaker

3.2.13 Whatman paper filter No.1, 70 mm diameter

3.2.14 Suction flask

3.2.15 Suction pump

3.2.16 SPE chamber

3.2.17 Buchner funnel

3.2.18 Glass funnel

3.2.19 Chromatographic column, 2 cm diameter and 30 cm length

3.2.20 Sieve (2 mm)

3.2.21 Mortar

All glassware used was washed with liquid soap and rinsed properly with distilled water, and then with pure acetone (AR). They were then baked in an oven at 200 °C for 24 hours.



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3.3 Sample collection

Water, sediment, and aquatic invertebrate samples were collected once a month in the irrigated Khlong 7 at the same period from June 2004 to May 2005. Three sampling locations along the Khlong 7 were divided as follow the direction flow of the canal into upper stream, middle stream, and lower stream sites, respectively.

3.3.1 Water sample

Three replicates of surface water samples, a measured volume of sample approximately 1 L each, were collected at each site: upper, middle, and lower stream sites by using water sampler. Water samples were suddenly refrigerated at -34°C in laboratory from the time of collection until extraction (SOP for USEPA Method 8081A and 3510).

3.3.2 Sediment sample

Three sediment samples each with 100 grams of surface sediment were collected with grab sampler from each site: upper, middle, and lower stream sites. Then the samples were air-dried at room temperature for 1-2 weeks, homogenized, and sieved with mesh no. 200 (SOP for USEPA Method 8081A and 3541).

3.3.3 Aquatic invertebrate sample

Aquatic invertebrates such as Lanchester's freshwater prawn *Macrobrachium lanchesteri*, apple snail *Pomacea* sp., and freshwater snail *Filopaludina martensi* were obtained using a dip net or a fishnet. On site, the samples were immediately wrapped with aluminum foil and placed on ice and then stored in the refrigerator at temperature lower than 4°C until extraction (SOP for AOAC Method 983.21).

3.4 Sample preparation

3.4.1 Extraction

3.4.1.1 Water sample

Initially, water sample was well-shaken and accurately adjusted the volume to 800 mL with graduated cylinder and filtrated through a Whatman paper (diameter, 70 mm) with a Buchner funnel.

The 800-ml-filtered water sample was poured into a 2-L separatory funnel. Triplicated extraction with 1:1 v/v n-Hexane:Dichloromethane (DCM) 100, 50, and 50 mL, respectively, were performed. The mixtures were shaken vigorously for 2 min before letting phases separate for at least 10 min as following APHA (1992). Then, the water phase was drained from separatory funnel into sample bottle and carefully poured organic phase pass through a glass funnel containing a 20 g anhydrous sodium sulfate through a 200-mL concentrator tube. Then, sample was poured back into separatory funnel. The extract was concentrated to a volume of 2 mL by Turbo Vap.

3.4.1.2 Sediment and invertebrate samples

The air-dried sediment sample was sieved with mesh no. 200 to remove course sands and organic debris. Five grams of sediment sample was mixed with 5 g anhydrous sodium sulfate (1:1 w/w) and filled in a 34-mL vessel of accelerated solvent extractor (ASE, Dionex ASE-100). The vessel was layered with activated copper powder on the ASE filter paper and fulfilled with Ottawa sand.

In the case of invertebrate samples, before extraction, the whole body of each group was homogenized using blender. Five grams of composite samples was mixed with 15 grams anhydrous sodium sulfate (1:3 w/w). The mixture was then placed into the vessel of ASE which was layered with filter paper and fulfilled with the Ottawa sand.

The method from Pan *et al.* (2004) was applied and validated from both sediment and aquatic organism extractions. The pressured liquid extraction was implemented using ASE for these samples. The working conditions were as follows:

preheating for 5 min, extraction temperature at 100°C, pressure at 1500 psi, static cycle of 10 min in twice, and purging with Nitrogen for 60 second by using 1:1 v/v n-Hexane: DCM as extracting solvent. Afterward, the extract was concentrated to 2 mL by using Turbo Vap.

3.4.2 Clean up

Because of the presence of pigment and sulfur in the extract of sediment sample, it was necessary to remove the contaminants before injecting to GC. The extracted sediment sample was de-sulfur and cleaned up using a multi-layer chromatographic column provided from Pan *et al.* (personal communication, 2004). The column was orderly packed with glass wool, activated copper powder, 6 g activated florisil, and 10 g anhydrous sodium sulfate. Then the extract was poured into the multi-layer chromatographic column and eluted with 50 mL of 6%, 15%, and 50% diethyl ether in petroleum ether, respectively. After that, the eluate was concentrated to 2 mL by Turbo Vap for the GC analysis.

For aquatic invertebrate sample, the extract was cleaned through 500 mg extract-clean-florisil SPE cartridge to clean up process. The extract was eluted with 10 mL of 6%, 15%, and 50% diethyl ether in petroleum ether, respectively. Before GC analysis, the eluate was concentrated in Turbo Vap to a volume of 2 mL.

3.5 Sample analysis

The extracted samples were analyzed by GC (Agilent Technologies 6890N) with micro-Electron Capture Detectors (μ -ECDs). The data system was an Agilent Chemstation G2070AA operated with Windows Me and an Agilent Kayak XA, 350 MHz Pentium II computer workstation.

One microlitre of the extracted sample was injected into the GC-ECD with splitless mode for 0.75 min vent delay. The injector and detector temperature was maintained at 260°C and 300°C, respectively. The oven temperature was initially maintained at 100°C, and then programmed to increase at 12 °C /min to 280 °C and held for 10 min. Total run time was calculated to be 27.00 min. Ultra-pure helium and

nitrogen gasses were used as carrier and make-up gasses, respectively. For optimum performance, the carrier gas flow rate was established at 2 mL/min linear velocity and nitrogen is set at 60 mL/min as make-up gas.

3.6 Method validation

Following the SOP for AOAC method 983.21, performing blank analysis and assessing recovery from pre-extracted and fortified matrices were done in each sampling batch. Triplicate analyses of extracted samples from each sampling time were done to ensure that the measurement remained stable. The spiked recovery (%) and relative standard deviation (%) were calculated.

3.6.1 Limit of detection (LOD) and limit of quantitation (LOQ)

The limit of detection (LOD) and limit of quantitation (LOQ) were defined as the peak height of analyte in standard solution that signaled significantly different from the peak height of noise. They were 3 and 10 times of signal per noise for LOD and LOQ, respectively. LOD and LOQ were done. In case of the OCPRs concentrations below the LOD, the results were described as ND or not detectable.

3.6.2 Spike recovery

Fortified samples were done in every sampling batch to ensure that the extraction efficiency would be under control (Kebbekus and Mitra, 1998). In addition, the acceptable recovery of the OCPRs should be ranged from 70 to 130 %, following SOP for USEPA METHOD 8081 + 3510 (waters). The recovery percentage can be calculated by the equation below,

$$\% \text{ Recovery} = \frac{\text{amount of OCPR determined}}{\text{amount of OC standard}} \times 100$$

3.6.3 Blanks

To avoid the effect of interferences, the set of blanks were done. The blanks included solvent blank, system blank, and fortified sample blank. These blanks were done every sample batch. The blanks must be free from contaminants, or the concentration of contaminated analytes must be at least level.

3.6.4 Replications

The replications of samples were done to evaluate repeatability. The samples were extracted and analyzed in triplicate to be sure that the measurement remained stable. The % RSD was calculated from the equation as below;

$$\% \text{ RSD} = \frac{\text{standard deviation}}{\text{mean}} \times 100$$

3.6.5 Method detection limit (MDL)

The detection limit of the selected method was calculated based on the replicated determinations as following.

$$\text{MDL} = t_{0.95[n-1]} \times \text{SD}$$

Where t is the threshold value of student t -distribution at the degree of $(n-1)$, n represents the number of replications, and SD represents the standard deviation. The confidence interval is 95% ($\alpha=0.05$).

3.7 Statistical analyses

The SPSS for windows version 10.00 and the SigmaStat version 2.0 were used in statistical analyses at 0.05 levels of significance ($p \leq 0.05$). For the samples with OCPRs concentrations below LOD, zero was used for calculations.

3.7.1 To compare OCPR concentrations in each matrix such as water, sediment, and aquatic invertebrates throughout the year (from June 2004 to May 2005)

After GC analysis, overall mean concentrations of OCPRs in water, sediment, and three species of invertebrates were compared using descriptive statistics (mean \pm standard error, SE) in order to indicate the level of accumulations and fluctuation trends.

3.7.2 To compare the differences in OCPR concentrations in water, sediment, and each aquatic invertebrate species collected in different seasons and from different study sites

Comparisons of OCPRs concentrations among different sites: upper, middle, and lower streams, and among different samples: water, sediment, Lanchester's freshwater prawn, apple snail, and freshwater snail samples, were analyzed. All data were checked for normal distribution. If any data was normal distribution, One Way ANOVA and T-Test were used. But, if any data was not normal distribution, Kruskal-Wallis One Way ANOVA and Mann-Whitney U Test were used.

3.8 Risk evaluation

The Maximum Residue Limits (MRLs), the maximum concentration of a pesticide residue (mg/kg) recommended by the Codex Alimentarius Commission and the Ministry of Public Health in Thailand, were used to evaluate the potential health risk compared with OCPRs in all aquatic organism samples.

CHAPTER IV

RESULTS AND DISCUSSION

4.1 Result of organochlorine analysis

4.1.1 Result of retention time of 17 mixed organochlorine pesticides standard

By using DB-35MS, the 17 mixed organochlorine pesticides standard consisting of α -BHC, γ -BHC, β -BHC, heptachlor, δ -BHC, aldrin, heptachlor epoxide, endosulfan I, 4,4'-DDE, dieldrin, endrin, 4,4'-DDD, endosulfan II, 4,4'-DDT, endrin aldehyde, endosulfan sulfate, and methoxychlor, respectively, were consequently separated. Figure 4.1 showed the chromatogram of the retention times of 17 organochlorine pesticides standard (100 ng/L in hexane) on DB-35MS.

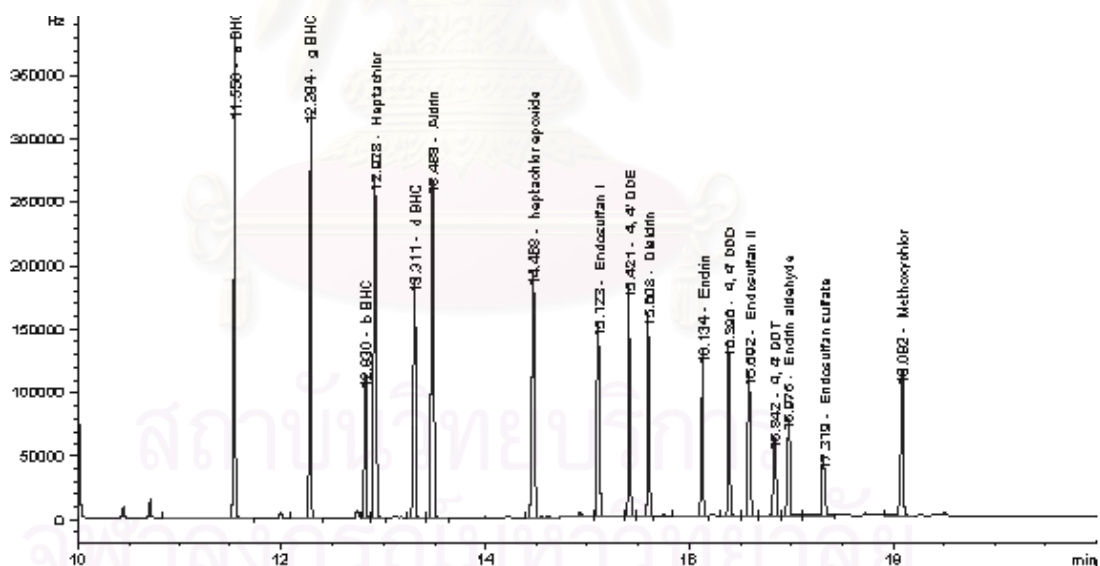


Figure 4.1 Gas chromatogram of 17 mixed organochlorine pesticides standard 100 ng/L in hexane included α -BHC, γ -BHC, β -BHC, heptachlor, δ -BHC, aldrin, heptachlor epoxide, endosulfan I, 4,4'-DDE, dieldrin, endrin, 4,4'-DDD, endosulfan II, 4,4'-DDT, endrin aldehyde, endosulfan sulfate, and methoxychlor, respectively.

4.1.2 Result of limit of detection (LOD) and limit of quantitation (LOQ)

The LOD and LOQ are defined as the peak height of analyses in standard solution that signaled significantly different from the peak height of noise about 3 times for LOD and 10 times for LOQ of each compound. Figure 4.2 showed that the LOD in this study was ranged from 0.0007 to 0.0508 and the LOQ was ranged from 0.0024 to 0.1695. The OCPRs concentrations which below the LOD were described as ND or not detectable.

4.1.3 Result of method detection limit (MDL)

The results in Table 4.1 showed that the MDL of 17 mixed organochlorine pesticides ranged from 0.12 to 3.74 $\mu\text{g/L}$ in water samples, from 2.26 to 14.87 $\mu\text{g/kg}$ dry weight in sediment samples, from 1.37 to 14.27 $\mu\text{g/kg}$ dry weight in Lanchester's freshwater prawn samples, and from 0.39 to 12.93 $\mu\text{g/kg}$ wet weight in apple snail samples.

4.1.4 Result of recovery

The results in Table 4.1 showed that the recoveries of 17 mixed organochlorine pesticides ranged from 70.62 % to 121.87 % in water samples, from 74.87 % to 95.16 % in sediment samples, from 70.92 % to 115.14 % in Lanchester's freshwater prawn samples, and from 70.24 % to 116.60 % in apple snail samples. All spiked matrices were in the acceptable range, from 70 % to 130 % (SOP USEPA METHOD 8081 + 3510). The repeatability was also described by % relative standard deviation (% RSD). The % RSD in water samples, ranging from 0.74 to 18.32, were in the acceptable range recommended by AOAC that referred to not exceed 21 at 10 ppb. Likewise, % RSD were ranged from 0.72 in apple snail to 14.62 in Lanchester's freshwater prawn which were not exceed 15 at 50 ppb. Accordingly, the recovery percentages and repeatability of all analytes were accepted in this study.

Table 4.1 Limit of detection (LOD), limit of quantitation (LOQ), method detection limit (MDL) (ppb), spiked recovery (%), and relative standard deviation (% RSD) of organochlorine pesticides standard solution in water, sediment, Lanchester's freshwater prawn, and apple snail samples.

Organochlorine Pesticides (OCPRs)	LOD (ppb)	LOQ (ppb)	MDL (ppb)				Matrices spiked recovery (%)				Relative standard deviation (% RSD)			
			Water	Sediment	Lanchester's freshwater prawn	Apple snail	Water	Sediment	Lanchester's freshwater prawn	Apple snail	Water ^a	Sediment ^b	Lanchester's freshwater prawn ^b	Apple snail ^b
α -BHC	0.03	0.09	2.06	3.38	1.37	0.56	95.21	74.87	73.87	72.62	6.02	4.93	1.74	0.72
γ -BHC	0.05	0.2	2.03	2.26	3.66	5.93	98.92	77.62	67.84	68.83	5.86	3.17	5.06	8.08
β -BHC	0.01	0.05	1.59	2.72	5.60	4.07	100.16	76.85	104.06	84.17	6.34	3.32	7.66	4.33
δ -BHC	0.0007	0.002	2.34	1.64	4.43	1.68	107.18	77.34	73.44	85.26	2.12	2.31	5.66	1.85
Heptachlor	0.04	0.1	0.35	2.11	5.60	4.51	119.94	84.08	83.22	70.82	13.28	2.73	7.15	5.97
Heptachlor epoxide	0.02	0.05	1.04	5.38	14.27	0.39	86.75	83.02	91.59	70.76	6.57	7.07	14.62	0.53
Aldrin	0.02	0.07	0.70	6.94	3.59	4.10	70.62	79.99	70.92	86.84	7.52	9.47	4.74	4.20
Dieldrin	0.05	0.2	3.46	7.94	5.90	1.15	88.82	89.30	99.78	104.85	18.32	9.70	4.05	1.03
4,4'-DDE	0.04	0.1	1.17	9.10	3.66	9.72	76.50	85.81	88.33	104.84	5.76	11.58	3.89	7.84
4,4'-DDD	0.03	0.1	0.08	3.98	10.14	1.61	121.87	86.15	101.48	106.05	0.74	5.04	5.42	1.42
4,4'-DDT	0.02	0.07	0.93	4.46	8.29	6.93	85.85	92.51	103.43	77.58	6.76	5.26	7.52	8.38
Endosulfan I	0.003	0.01	1.49	10.38	3.93	9.72	95.40	90.03	76.85	86.40	4.30	12.58	4.80	10.55
Endosulfan II	0.003	0.009	1.31	10.19	5.91	2.65	94.98	88.87	109.12	116.60	5.08	12.51	5.08	2.13
Endosulfan sulfate	0.002	0.008	3.74	14.87	9.56	12.93	109.86	108.30	90.81	94.85	2.61	12.88	9.87	12.78
Endrin	0.002	0.007	2.05	11.44	6.82	3.81	89.61	85.48	92.11	111.55	11.32	14.60	6.94	3.20
Endrin aldehyde	0.002	0.007	1.69	9.92	6.87	2.17	115.72	90.61	95.82	70.24	5.05	11.95	6.72	2.89
Methoxychlor	0.02	0.06	0.12	10.61	14.01	7.94	71.30	93.55	115.14	71.29	0.86	12.20	14.25	10.45

^a % RSD at 10 ppb of analyses

^b % RSD at 50 ppb of analyses

4.2 Levels of OCPRs in water, sediment, and aquatic invertebrate samples throughout the year

4.2.1 OCPRs in water samples

A total number of 108 water samples was collected and analyzed. The results were summarized in Table 4.2. The overall means \pm SE of the OCPRs concentrations in water samples at Khlong 7, Rangsit agricultural area, Pathum Thani province collected once a month from June 2004 to May 2005 were shown in Table 4.2. Σ Endosulfans (including endosulfan I, endosulfan II, and endosulfan sulfate) were the highest, following by Σ DDTs (including 4,4'-DDT, 4,4'-DDD, and 4,4'-DDE), Σ BHCs (including α -BHC, γ -BHC, β -BHC, and δ -BHC), Σ Aldrin (including aldrin and dieldrin), Σ Heptachlor (including heptachlor and heptachlor epoxide), Σ Endrin (including endrin and endrin aldehyde), and methoxychlor, respectively.

Generally, endosulfan is a mixture of two stereoisomers: 70% is the exo configuration (endosulfan I or α -endosulfan) and 30% is the endo configuration (endosulfan II or β -endosulfan). High amount of endosulfan and its derivative (endosulfan sulfate) were found in water samples in this study because it was recently banned in 2004 in Thailand from now. This result may probably reflect the usage of prohibited organochlorine pesticides. The reason may be because of continuous using of the endosulfan by farmers to control the apple snail which has been a major pest problem in paddy fields since 1975 (Thirakhupt *et al.*, 2006). Furthermore, this study area is crowded with rice cultivation area, therefore, it is not surprise to detect the high amount of endosulfan and its derivative. Endosulfan was widely used in Thailand because of lower production cost than the other groups of pesticides, for instances, pyrethroid, organophosphate pesticides, or biopesticide (neem) and because of the ease of purchasing in pesticide grocery (Haruthaithanasan, 1999).

To determine the components of Σ Endosulfans, the main endosulfan metabolite (endosulfan sulfate) was found more than the parent forms (endosulfan I and endosulfan II). To consider the water solubility, the partition coefficient (K_{ow}) was used to describe this property. The log K_{ow} for endosulfan sulfate is 3.77 which lower than log K_{ow} of the parent forms, endosulfan I and endosulfan II which are 4.65

and 4.34, respectively. The degradation product (endosulfan sulfate) was, therefore, less hydrophobic and less bioaccumulative when compared to endosulfan (German Federal Environment Agency, 2004). As a result, this reason may support the elevated presence of endosulfan sulfate.

According to this study, the presence of OCPRs concentrations at Khlong 7, Rangsit agricultural area was similar to the concentrations of OCPRs reported in water samples from agricultural areas in the eastern part of Thailand collected in 1996 and the northern part of Thailand collected in 1997 (PCD, 1997, cited in Thapinta and Hudak, 1998). The OCPRs in water samples were also the highest found in endosulfan group, ranging from 0.003 - 1.350 $\mu\text{g/L}$ in the northern part of Thailand and from 0.001 - 0.460 $\mu\text{g/L}$ in the eastern part of Thailand. Similarly, the study in surface water samples from the Ganges river at Farrukhabad area and at Varanasi area in India found vast amount of Σ Endosulfans (0.232 $\mu\text{g/L}$ and 0.083 - 66.516 $\mu\text{g/L}$, respectively (Agnihotri *et al.*, 1994; Nayak *et al.*, 1995) which much higher than the result obtained in this study (0.08 $\mu\text{g/L}$). Likewise, Miles and Pfeuffer (1997) found the Σ Endosulfans concentrations up to 0.748 $\mu\text{g/L}$ occasionally above the Florida water quality criterion (0.056 $\mu\text{g/L}$) for the surface water of South Florida in which endosulfan was used for control whiteflies on vegetable crops such as tomato crops.

Table 4.2 Overall means (\pm SE) of the OCPRs concentrations ($\mu\text{g/L}$) in water samples at Khlong 7, Rangsit agricultural area, Pathum Thani province collected once a month from June 2004 to May 2005.

Organochlorine pesticide residues (OCPRs)	Concentration of OCPRs (mean \pm SE in $\mu\text{g/L}$) (n = 108)
α -BHC	0.0017 \pm 0.0002
γ -BHC	0.0051 \pm 0.0003
β -BHC	0.007 \pm 0.002
δ -BHC	0.0005 \pm 0.0002
Σ BHCs	0.014 \pm 0.002
Heptachlor	0.0063 \pm 0.0009
Heptachlor epoxide	0.0005 \pm 0.0001
Σ Heptachlor	0.0068 \pm 0.0009
Aldrin	0.0028 \pm 0.0003
Dieldrin	0.0045 \pm 0.0008
Σ Aldrin	0.0072 \pm 0.0010
4,4'-DDE	0.0004 \pm 0.0001
4,4'-DDD	0.0008 \pm 0.0002
4,4'-DDT	0.018 \pm 0.001
Σ DDTs	0.019 \pm 0.001
Endosulfan I	0.0052 \pm 0.0008
Endosulfan II	0.007 \pm 0.002
Endosulfan sulfate	0.07 \pm 0.01
Σ Endosulfans	0.08 \pm 0.01
Endrin	0.0038 \pm 0.0005
Endrin aldehyde	0.0010 \pm 0.0002
Σ Endrin	0.0048 \pm 0.0006
Methoxychlor	0.004 \pm 0.001

4.2.2 OCPRs in sediment samples

Table 4.3 showed the results of the overall means of the OCPRs concentrations in sediment samples throughout the year. The results showed that the means of Σ Heptachlor, Σ DDTs, Σ BHCs, and Σ Endosulfans were detected at high concentrations in sediment samples whereas Σ Aldrin, Σ Endrins, and methoxychlor were detected at low concentrations.

Heptachlor is converted to heptachlor epoxide and other degradation products in the environment. Among the Σ Heptachlor, the average concentrations of heptachlor ($13.4 \pm 0.4 \mu\text{g/kg}$ dry weight) were much higher than heptachlor epoxide ($1.29 \pm 0.09 \mu\text{g/kg}$ dry weight). People always use heptachlor for killing insects especially termites and fire ants, in homes, buildings, and on food crops (ATSDR, 1993). Because heptachlor sticks to sediment very strongly and does not dissolve easily in water, the existence of heptachlor in this study was detected. Generally, both heptachlor and heptachlor epoxide adsorb strongly to sediments (ATSDR, 2005). Heptachlor epoxide degrades more slowly and, as a result, it is more persistent than heptachlor. Therefore, heptachlor epoxide should be found more than heptachlor. However, heptachlor was still detected in this study more than heptachlor epoxide. The reasons may be because of the heavily usage in the past and the illegal usage of heptachlor nowadays although it has been banned since 1998. The level of Σ Heptachlor was similar to the concentrations reported in earlier studies which also found the highest amount of heptachlor in sediment from Göksu delta in Turkey (Ayas, 1997) and from the Eastern Cape in South Africa (Awofolu, 2003). In the earlier study, heptachlor was found in the sediment of Casco Bay, Washington in concentrations ranging from 0.04 to 0.13 $\mu\text{g/kg}$ dry weight (Kennicutt *et al.* 1994) which much lower than the results in this study ($13.4 \pm 0.4 \mu\text{g/kg}$ dry weight).

Among the popular pesticide of Σ DDTs, the existence of 4,4'-DDT was the uppermost ($7.4 \pm 0.2 \mu\text{g/kg}$ wet weight) following by its metabolites, 4,4'-DDE ($3.03 \pm 0.08 \mu\text{g/kg}$ wet weight) and 4,4'-DDD ($1.66 \pm 0.05 \mu\text{g/kg}$ wet weight), respectively. The metabolic forms that were 4,4'-DDE and 4,4'-DDD should be found more than the parent form (4,4'-DDT). However, the study showed that the average concentrations of 4,4'-DDT were highest even though DDT has been banned since

1983 in Thailand. Most DDT breaks down slowly into DDE and DDD, generally by the action of microorganisms. They stick strongly to soil, and therefore, generally remain in the surface layers of soil. Some soil particles with attached DDT, DDE, or DDD may get into aquatic environment in runoff (ATSDR, 2002). Σ DDTs were entered to surface water and then bound to particles in the water. Due to the hydrophobic property, the Σ DDTs were then settled and deposited in the sediment. The high means concentrations of 4,4'-DDT may be because of its persistence in which a half life can remain for 20, 30, or more years (ATSDR, 2002). The other evidences were resulted by the illegal sale from retail shops around the Rangsit area, and some farmers had illegally used for rice cultivation (interview, March 2003). The Σ DDTs concentrations in this study ($12.1 \pm 0.3 \mu\text{g/kg}$ dry weight) were compared to the amount of sediments from Korea by Lee *et al.* (2001). Σ DDTs concentrations obtained in this study were close to the amount of Σ DDTs concentrations from Kyeonggi Bay, Korea ($0.048 - 32 \mu\text{g/kg}$ dry weight) but higher than in Namyang Bay and Lake Shihwa, Korea ($0.088 - 0.38$ and $0.62 - 2.3 \mu\text{g/kg}$ dry weight).

Among the Σ BHCs, γ -BHC (lindane) had the highest average concentrations of $5.13 \pm 0.08 \mu\text{g/kg}$ dry weight in sediment samples, following by $3.3 \pm 0.2 \mu\text{g/kg}$ dry weight for β -BHC, $0.65 \pm 0.09 \mu\text{g/kg}$ dry weight for δ -BHC, and $0.26 \pm 0.07 \mu\text{g/kg}$ dry weight for α -BHC. The high amount of lindane revealed the current usage of technical BHC in this region even though technical BHC has been banned since 2001 in Thailand. This study was related to the study of Kan-atireklap *et al.* (1997) who reported that γ -BHC was the most prevalent isomers in Thailand due to the continuous use of this pesticide. Also, the γ -BHC was the dominant isomer of Σ BHCs from the Merhei lake from the Danube Delta, Romania ($2.8 \mu\text{g/kg}$ dry weight) (Covaci, 2006) which slightly lower than the results in this study.

Table 4.3 Overall means (\pm SE) of the OCPRs concentrations ($\mu\text{g}/\text{kg}$ dry weight) in sediment samples at Khlong 7, Rangsit agricultural area, Pathum Thani province collected once a month from June 2004 to May 2005.

OCPRs	Concentration of OCPRs (mean \pm SE in $\mu\text{g}/\text{kg}$ dry weight) (n=108)
α -BHC	0.26 \pm 0.07
γ -BHC	5.13 \pm 0.08
β -BHC	3.3 \pm 0.2
δ -BHC	0.65 \pm 0.09
Σ BHCs	9.4 \pm 0.3
Heptachlor	13.4 \pm 0.4
Heptachlor epoxide	1.29 \pm 0.09
Σ Heptachlor	14.7 \pm 0.5
Aldrin	0.81 \pm 0.06
Dieldrin	2.2 \pm 0.2
Σ Aldrin	3.0 \pm 0.2
4,4'-DDE	3.03 \pm 0.08
4,4'-DDD	1.66 \pm 0.05
4,4'-DDT	7.4 \pm 0.2
Σ DDTs	12.1 \pm 0.3
Endosulfan I	0.87 \pm 0.03
Endosulfan II	2.3 \pm 0.1
Endosulfan sulfate	3.2 \pm 0.2
Σ Endosulfans	6.4 \pm 0.3
Endrin	0.6 \pm 0.1
Endrin aldehyde	0.23 \pm 0.07
Σ Endrin	0.8 \pm 0.1
Methoxychlor	0.16 \pm 0.06

4.2.3 OCPRs in aquatic invertebrate samples

The results of average OCPRs concentrations collected from June 2004 to May 2005 from aquatic invertebrate samples such as Lanchester's freshwater prawn, apple snail, and freshwater snail were shown in Table 4.4. The OCPRs concentrations revealed that the four highest concentrations were \sum DDTs (79.6 ± 8.8 , 53.0 ± 7.9 , and 47.8 ± 5.1 $\mu\text{g/kg}$ wet weight for freshwater snail, Lanchester's freshwater prawn, and apple snail samples, respectively), \sum Endosulfans (36.7 ± 5.7 , 36.5 ± 4.0 , and 27.9 ± 3.4 $\mu\text{g/kg}$ wet weight for Lanchester's freshwater prawn, apple snail, and freshwater snail samples, respectively), \sum BHCs (42.3 ± 4.6 , 34.4 ± 2.6 , and 27.1 ± 2.1 $\mu\text{g/kg}$ wet weight for freshwater snail, apple snail, and Lanchester's freshwater prawn samples, respectively), and \sum Heptachlor (19.0 ± 1.5 , 18.9 ± 2.3 , and 14.5 ± 0.9 $\mu\text{g/kg}$ wet weight for apple snail, freshwater snail, and Lanchester's freshwater prawn, respectively) (Figure 4.2). For other OCPRs, the means concentrations of \sum Aldrin, \sum Endrin, and methoxychlor were relatively low in these biological samples.

Among \sum DDTs, the average concentrations of 4,4'-DDT were detected at the highest levels in all species (45.8 ± 7.7 , 30.6 ± 2.6 , and 55.2 ± 6.6 $\mu\text{g/kg}$ wet weight for Lanchester's freshwater prawn, apple snail, and freshwater snail, respectively), following by 4,4'-DDE and 4,4'-DDD in Lanchester's freshwater prawn and freshwater snail samples, but following by 4,4'-DDD and 4,4'-DDE in apple snail samples. Higher compositions of 4,4'-DDT in all invertebrate samples may reflect that a contaminant source of \sum DDTs may be around the study area. In addition, the contamination of OCPRs in biological samples may probably from the water pump-in from paddy fields surrounding the Khlong 7 canal which farmers could apply organochlorine pesticides into their farms. Besides, the high amount of \sum DDTs in biological samples and sediment samples may be due to their higher particle affinity, lipophilicity, and biochemical stability among the organochlorine pesticides (Tanabe *et al.*, 1989). Thus their persistence with the long half-life may be incorporated in the biota samples in this study. Likewise, DDT and its metabolites were highest in green mussel *Perna viridis*, L. from the coastal waters of Thailand, ranging from 1.2 - 38 $\mu\text{g/kg}$ wet weight (Kan-atireklap *et al.*, 1997) which slightly lower than the results obtained in this study. To compare with other areas, the \sum DDTs concentrations in this study were higher than the invertebrates from Danube Delta in Romania such as

chironomids *Chironomus plumosus* (26 – 38 $\mu\text{g}/\text{kg}$ dry weight), but close to the ΣDDTs concentrations in zooplankton from the Razim lake from the Danube Delta (99 $\mu\text{g}/\text{kg}$ dry weight) (Covaci *et al.*, 2006). The ΣDDTs concentrations in gastropods such as freshwater and apple snail in this study were higher than in *Pluoploca trapezium* gastropod samples (7.5 $\mu\text{g}/\text{kg}$ wet weight) from the Ferry in Tanzania (Mwevura *et al.*, 2002). The other important OCPs were $\Sigma\text{Endosulfans}$ and ΣBHCs . The average concentrations of ΣBHCs in freshwater snail samples were significantly higher than in Lanchester's freshwater prawn samples (Tamhane' T2, $P < 0.05$), but not different in apple snail samples. Among ΣBHCs , the ratios of $\beta\text{-BHC}$ to the ΣBHCs were highest in all of aquatic invertebrates. For $\Sigma\text{Endosulfans}$, degradation product of endosulfan (endosulfan sulfate) was the dominance.



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Table 4.4 Overall means (\pm SE) of the OCPRs concentrations ^a ($\mu\text{g}/\text{kg}$ wet weight) in Lanchester's freshwater prawn, apple snail, and freshwater snail samples at Khlong 7, Rangsit agricultural area in Pathum Thani province collected once a month from June 2004 to May 2005.

OCPRs	Concentration of OCPRs (mean \pm SE in $\mu\text{g}/\text{kg}$ wet weight aquatic invertebrates)		
	Lanchester's freshwater prawn (n=93)	Apple snail (n=72)	Freshwater snail (n=57)
α -BHC	2.8 \pm 0.5 ^a	10.0 \pm 1.1 ^b	9.2 \pm 0.7 ^b
γ -BHC	2.4 \pm 0.3 ^a	0.7 \pm 0.3 ^b	1.5 \pm 0.6 ^{ab}
β -BHC	12.3 \pm 1.1 ^a	18.3 \pm 1.4 ^{ab}	25.6 \pm 2.9 ^b
δ -BHC	9.6 \pm 1.9 ^a	5.3 \pm 1.4 ^a	5.9 \pm 1.2 ^a
Σ BHCs	27.1 \pm 2.1 ^a	34.4 \pm 2.6 ^{ab}	42.3 \pm 4.6 ^b
Heptachlor	12.4 \pm 0.8 ^a	15.4 \pm 1.3 ^a	14.1 \pm 1.2 ^a
Heptachlor epoxide	2.1 \pm 0.4 ^a	3.6 \pm 0.5 ^{ab}	4.8 \pm 1.8 ^b
Σ Heptachlor	14.5 \pm 0.9 ^a	19.0 \pm 1.5 ^a	18.9 \pm 2.3 ^a
Aldrin	3.5 \pm 0.3 ^a	6.7 \pm 0.6 ^b	6.9 \pm 0.5 ^b
Dieldrin	2.4 \pm 0.4 ^a	9.0 \pm 1.4 ^b	11.4 \pm 1.2 ^b
Σ Aldrin	5.9 \pm 0.5 ^a	15.7 \pm 1.7 ^b	18.3 \pm 1.6 ^b
4,4'-DDE	5.0 \pm 0.4 ^a	7.8 \pm 1.6 ^a	15.9 \pm 1.1 ^b
4,4'-DDD	2.2 \pm 0.6 ^a	9.4 \pm 2.0 ^b	8.4 \pm 2.3 ^{ab}
4,4'-DDT	45.8 \pm 7.7 ^{ab}	30.6 \pm 2.6 ^a	55.2 \pm 6.6 ^b
Σ DDTs	53.0 \pm 7.9 ^a	47.8 \pm 5.1 ^a	79.6 \pm 8.8 ^a
Endosulfan I	3.3 \pm 0.3 ^a	8.7 \pm 1.8 ^b	8.1 \pm 1.0 ^b
Endosulfan II	5.4 \pm 0.7 ^{ab}	10.1 \pm 1.7 ^a	2.6 \pm 1.2 ^b
Endosulfan sulfate	27.9 \pm 5.6 ^a	17.7 \pm 1.7 ^a	17.2 \pm 2.8 ^a
Σ Endosulfans	36.7 \pm 5.7 ^a	36.5 \pm 4.0 ^a	27.9 \pm 3.4 ^a
Endrin	3.2 \pm 0.7 ^a	9.5 \pm 2.5 ^{ab}	8.7 \pm 1.6 ^b
Endrin aldehyde	2.7 \pm 0.7 ^a	7.3 \pm 1.2 ^b	0.7 \pm 0.7 ^a
Σ Endrin	5.8 \pm 1.2 ^a	16.8 \pm 3.2 ^b	9.3 \pm 1.8 ^{ab}
Methoxychlor	2.1 \pm 0.6 ^a	6.8 \pm 1.1 ^b	5.4 \pm 1.1 ^{ab}

^a means of the OCPRs concentrations with the same letter in the same row are not significantly different at $p \leq 0.05$

n: number of sampling

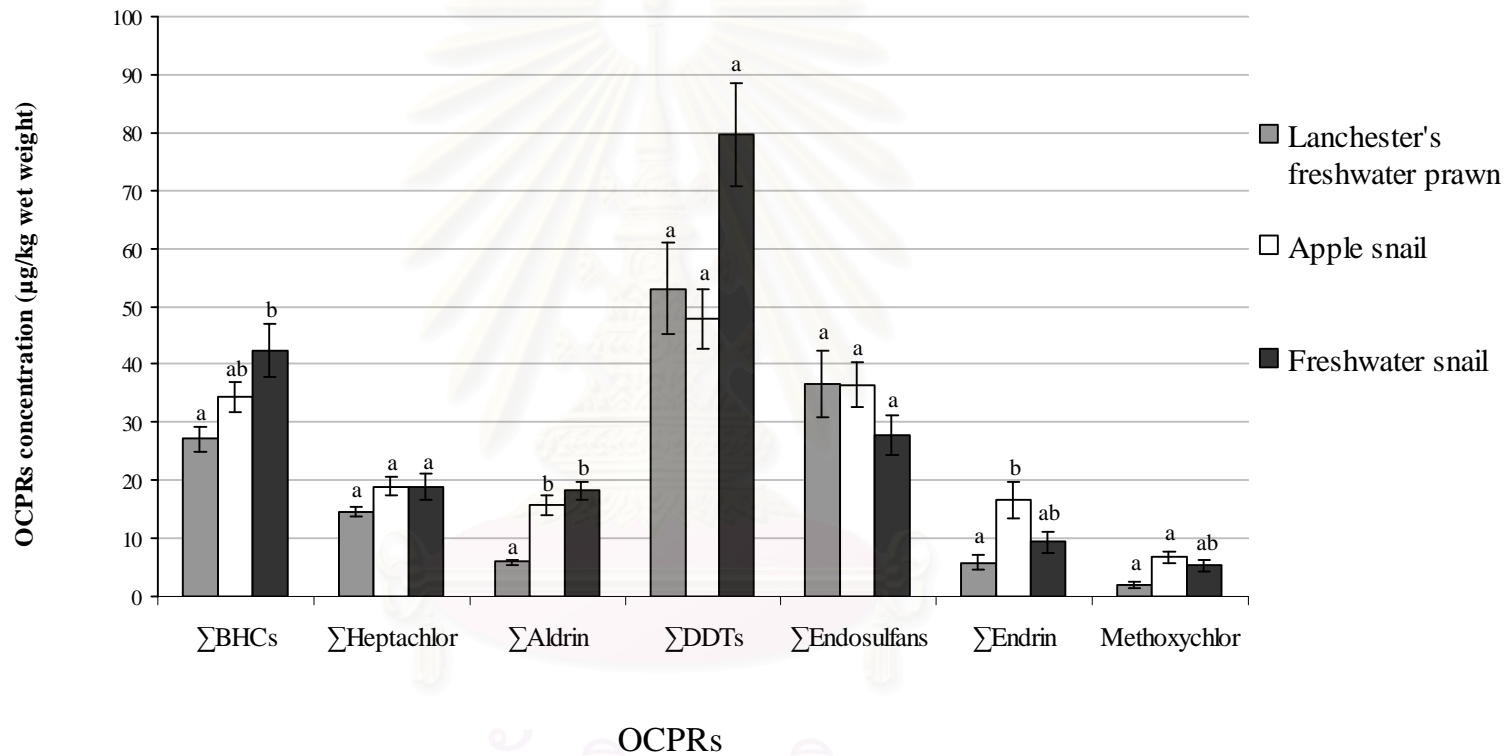


Figure 4.2 Overall means of OCPs concentrations (\pm SE) of Lanchester's freshwater prawn, apple snail, and freshwater snail samples collected once a month from June 2004 to May 2005 at Khlong 7, Rangsit agricultural area in Pathum Thani province.

4.3 Comparisons of OCPRs concentrations in water, sediment, and aquatic invertebrate samples from different study sites and different seasons

4.3.1 Water samples

The results in Table 4.5 presented the average concentrations of OCPRs in water samples in the different study sites such as upper stream, middle stream, and lower stream in wet season (June to November 2004) and dry season (December 2004 to May 2005) at Khlong 7, Rangsit agricultural area in Pathum Thani province. The highest amount of OCPRs concentrations in all locations were in wet season especially the predominant compounds of Σ Endosulfans as shown in Figure 4.3.

Statistical comparisons of average OCPRs concentrations between wet and dry seasons found that Σ Endosulfans in the upper stream were significantly higher in wet season than in dry season (Mann-Whitney U Test, Σ Endosulfans, $Z = -4.271$, P-value = 0.000, $p \leq 0.05$) whereas Σ Aldrin and methoxychlor were significantly higher in dry season than in wet season (Mann-Whitney U Test, Σ Aldrin, $Z = -2.642$, P-value = 0.008, $p \leq 0.05$ and methoxychlor, $Z = -2.198$, P-value = 0.028, $p \leq 0.05$). In the case of other OCPRs, the means of concentrations of Σ BHCs, Σ Heptachlor, Σ DDTs, and Σ Endrin were no statistical differences between wet and dry seasons. Similarly, the average concentrations of Σ Endosulfans in the middle stream were lower in the dry season than in wet season whereas methoxychlor were statistically higher in wet season than in dry season (Mann-Whitney U Test, Σ Endosulfans, $Z = -3.892$, P-value = 0.000, $p \leq 0.05$ and methoxychlor, $Z = -2.403$, P-value = 0.016, $p \leq 0.05$). The average concentrations of Σ Heptachlor were significantly higher in wet season than in dry season (Mann-Whitney U Test, $Z = -2.135$, P-value = 0.033, $p \leq 0.05$). For the rests of the average OCPRs concentrations, the Σ BHCs, Σ Aldrin, Σ DDTs, and Σ Endrin were not significantly different between wet and dry seasons. In the lower stream, the average concentrations of Σ Heptachlor, Σ Aldrin, and methoxychlor were significantly different between wet and dry seasons (Mann-Whitney U Test, Σ Heptachlor, $Z = -2.750$, P-value = 0.006, $p \leq 0.05$, Σ Aldrin, $Z = -2.784$, P-value = 0.005, $p \leq 0.05$, and methoxychlor, $Z = -3.840$, P-value = 0.000, $p \leq 0.05$). Similarly

to the middle stream, the average concentrations of Σ Heptachlor were higher in wet season than in dry season, and the average concentrations of methoxychlor were also higher in dry season than in wet season. For the Σ Aldrin, the means of concentrations in the dry season were higher than wet season. There were not significantly different for the average concentrations of the Σ BHCs, Σ DDTs, Σ Endosulfans, and Σ Endrin between wet and dry season.

According to the average concentrations overall the year of OCPRs of each site (June 2004 to May 2005) as shown in Table 4.5, the average concentrations of Σ Endosulfans were no differences among the upper stream, middle stream, and lower stream (Kruskal-Wallis One Way ANOVA, $df = 2$, $P\text{-value} = 0.147$, $p \leq 0.05$). Among the residues of Σ Endosulfans, endosulfan sulfate metabolite was the major composition of Σ Endosulfans and was highest detected in every location overall the year (Figure 4.3). Additionally, the average concentrations of Σ BHCs, Σ Heptachlor, Σ Aldrin, Σ DDTs, and methoxychlor were not significantly different among the three sites. In contrast, the results indicated that the overall means of Σ Endrins concentrations in the lower stream were statistically higher than the middle stream (Kruskal-Wallis One Way ANOVA, $df = 2$, $P\text{-value} = 0.009$, $p \leq 0.05$).

Along the Khlong 7, there are a number of paddy fields. Usually, farmers in this area have three crops per year or sometimes five crops per 2 years. The highest concentrations of Σ Endosulfans in wet season probably resulted from the usage of endosulfan as molluscicide in paddy fields to control apple snails in this area. Moreover, during the wet season, the concentrations of Σ Endosulfans were high due to the rainfall that leached the endosulfan contaminating in paddy fields into the aquatic system as well as the rice growing practice using pump-in water and pump-out water from the canal to the paddy fields that could transferred OCPRs into the Khlong 7.

Table 4.5 Average concentrations ^a of OCPs in water samples in the upper stream, middle stream, and lower stream in wet season (June - November 2004) and dry season (December 2004 - May 2005); and the average concentrations of OCPs of the each site overall the year (June 2004 - May 2005) at Khlong 7, Rangsit agricultural area, Pathum Thani province.

OCPs	Average concentrations of OCPs in water samples (mean ± SE in µg/L)								
	Upper stream		Middle stream		Lower stream		Overall the year		
	Wet season (n = 18)	Dry season (n = 18)	Wet season (n = 18)	Dry season (n = 18)	Wet season (n = 18)	Dry season (n = 18)	Upper stream (n = 36)	Middle stream (n = 36)	Lower stream (n = 36)
α-BHC	0.0012±0.0004 ^a	0.0012±0.0004 ^a	0.0016±0.0004 ^a	0.0015±0.0004 ^a	0.0033±0.0009 ^a	0.0018±0.0004 ^a	0.0012±0.0003 ^a	0.0015±0.0003 ^{ab}	0.0025±0.0005 ^b
γ-BHC	0.0054±0.0007 ^a	0.0051±0.0004 ^a	0.0043±0.0004 ^a	0.0048±0.0005 ^a	0.005±0.001 ^a	0.0059±0.0008 ^a	0.0052±0.0004 ^a	0.0045±0.0003 ^a	0.0055±0.0007 ^a
β-BHC	0.0009±0.0004 ^a	0.010±0.005 ^a	0.0011±0.0004 ^a	0.015±0.007 ^a	0.0016±0.0008 ^a	0.014±0.005 ^a	0.005±0.003 ^a	0.008±0.004 ^a	0.008±0.003 ^a
δ-BHC	0.0004±0.0002 ^a	ND ^a	0.0002±0.0002 ^a	0.0001±0.0050 ^a	0.003±0.001 ^a	ND ^b	0.0002±0.0001 ^a	0.0001±0.0001 ^a	0.0013±0.0006 ^a
ΣBHCs	0.008±0.002 ^a	0.016±0.006 ^a	0.0071±0.0006 ^a	0.021±0.007 ^a	0.013±0.002 ^a	0.021±0.005 ^a	0.012±0.003 ^a	0.014±0.004 ^a	0.017±0.003 ^a
Heptachlor	0.008±0.002 ^a	0.010±0.004 ^a	0.007±0.001 ^a	0.0003±0.0002 ^b	0.0072±0.0002 ^a	0.002±0.001 ^b	0.009±0.002 ^a	0.005±0.001 ^a	0.0049±0.0009 ^a
Heptachlor epoxide	0.0004±0.0002 ^a	0.0011±0.0004 ^a	0.0002±0.0001 ^a	0.0031±0.0007 ^a	0.0022±0.0003 ^a	0.0007±0.0003 ^a	0.0007±0.0003 ^a	0.0003±0.0001 ^a	0.0006±0.0002 ^a
ΣHeptachlor	0.008±0.002 ^a	0.011±0.004 ^a	0.007±0.001 ^a	0.003±0.001 ^b	0.008±0.001 ^a	0.003±0.001 ^b	0.010±0.002 ^a	0.005±0.001 ^a	0.0054±0.0008 ^a
Aldrin	0.0017±0.0004 ^a	0.0024±0.0004 ^a	0.0021±0.0003 ^a	0.0031±0.0007 ^a	0.0022±0.0003 ^a	0.005±0.001 ^b	0.0020±0.0003 ^a	0.0026±0.0004 ^a	0.0037±0.0006 ^a
Dieldrin	0.0022±0.0004 ^a	0.007±0.002 ^a	0.0019±0.0002 ^a	0.008±0.004 ^a	0.0016±0.0004 ^a	0.006±0.002 ^a	0.005±0.001 ^a	0.005±0.002 ^a	0.004±0.001 ^a
ΣAldrin	0.0038±0.0005 ^a	0.009±0.002 ^b	0.0039±0.0005 ^a	0.011±0.004 ^a	0.0038±0.0006 ^a	0.011±0.003 ^b	0.007±0.001 ^a	0.008±0.002 ^a	0.007±0.002 ^a
4,4'-DDE	0.0007±0.0004 ^a	0.001±0.002 ^a	0.0002±0.0002 ^a	0.0005±0.0003 ^a	0.0002±0.0002 ^a	ND ^a	0.0008±0.0002 ^a	0.0004±0.0002 ^{ab}	0.0004±0.0002 ^b
4,4'-DDD	ND ^a	0.0013±0.0005 ^a	0.0002±0.0001 ^a	0.0008±0.0003 ^a	0.0011±0.0006 ^a	0.0013±0.0005 ^a	0.0007±0.0003 ^a	0.0005±0.0002 ^a	0.0012±0.0004 ^a
4,4'-DDT	0.017±0.003 ^a	0.016±0.002 ^a	0.017±0.002 ^a	0.016±0.003 ^a	0.018±0.002 ^a	0.025±0.004 ^a	0.016±0.002 ^a	0.016±0.002 ^a	0.021±0.002 ^b
ΣDDTs	0.018±0.003 ^a	0.018±0.002 ^a	0.017±0.002 ^a	0.017±0.003 ^a	0.019±0.002 ^a	0.026±0.004 ^a	0.018±0.002 ^a	0.017±0.002 ^a	0.023±0.002 ^a
Endosulfan I	0.0028±0.0004 ^a	0.0036±0.0009 ^a	0.006±0.003 ^a	0.005±0.002 ^a	0.007±0.003 ^a	0.007±0.002 ^a	0.0032±0.0005 ^a	0.006±0.002 ^a	0.007±0.002 ^a
Endosulfan II	0.003±0.001 ^a	0.007±0.004 ^a	0.008±0.004 ^a	0.007±0.004 ^a	0.006±0.002 ^a	0.017±0.007 ^a	0.0035±0.0008 ^a	0.007±0.003 ^a	0.011±0.004 ^a
Endosulfan sulfate	0.13±0.04 ^a	0.016±0.003 ^a	0.11±0.03 ^a	0.016±0.003 ^b	0.12±0.04 ^a	0.034±0.006 ^a	0.07±0.02 ^a	0.06±0.02 ^a	0.07±0.02 ^a
ΣEndosulfans	0.14±0.04 ^a	0.018±0.003 ^b	0.12±0.04 ^a	0.028±0.008 ^b	0.13±0.01 ^a	0.06±0.01 ^a	0.08±0.02 ^a	0.08±0.02 ^a	0.09±0.02 ^a
Endrin	0.0017±0.0004 ^a	0.003±0.001 ^a	0.0023±0.0004 ^a	0.003±0.001 ^a	0.0034±0.0002 ^a	0.008±0.002 ^a	0.0030±0.0007 ^a	0.0027±0.0006 ^a	0.006±0.001 ^a
Endrin aldehyde	0.0008±0.0004 ^a	0.0010±0.0004 ^a	0.0003±0.0002 ^a	0.007±0.002 ^a	0.0002±0.0001 ^a	0.0016±0.0007 ^a	0.0014±0.0004 ^a	0.0006±0.0002 ^a	0.0009±0.0004 ^a
ΣEndrin	0.0025±0.0004 ^a	0.006±0.001 ^a	0.0026±0.0004 ^a	0.004±0.001 ^a	0.0036±0.0002 ^a	0.009±0.002 ^a	0.0044±0.0008 ^b	0.0033±0.0006 ^a	0.007±0.001 ^b
Methoxychlor	0.00007±0.00007 ^a	0.007±0.002 ^b	0.0011±0.0006 ^a	0.007±0.002 ^b	ND ^a	0.015±0.007 ^b	0.0010±0.0004 ^a	0.004±0.001 ^a	0.007±0.004 ^a

^a the average concentrations, in each data block: upper stream, middle stream, lower stream and overall the year data blocks, with the same letter in the same row are not significantly different at $p \leq 0.05$.

ND: samples with organochlorine pesticide concentrations below limit of detection (LOD), n: number of sampling

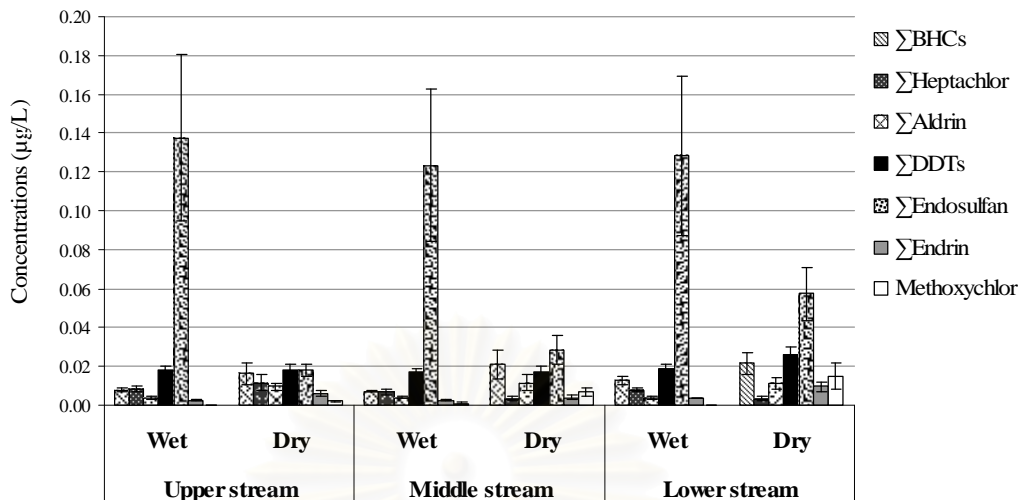


Figure 4.3 Comparisons of average OCPs concentrations (\pm SE) in water samples between wet season (June - November 2004) and dry season (December 2004 - May 2005) among the upper stream, middle stream, and lower stream at Klong 7, Rangsit agricultural area in Pathum Thani province.

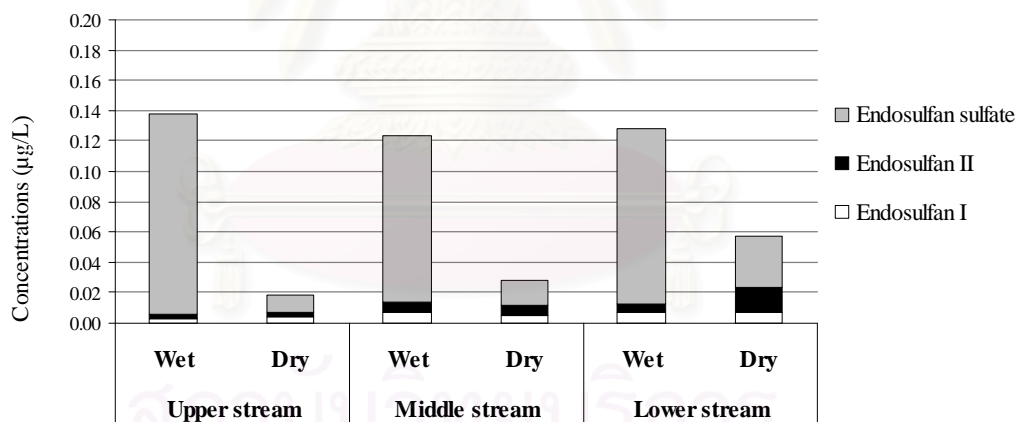


Figure 4.4 Compositions of Σ Endosulfans concentrations including endosulfan I, endosulfan II, and endosulfan sulfate in water samples between wet season (June - November 2004) and dry season (December 2004 - May 2005) among upper stream, middle stream, and lower stream at Klong 7, Rangsit agricultural area in Pathum Thani province.

4.3.2 Sediment Samples

Table 4.6 presented the overall average concentrations of OCPRs in sediment samples among the upper stream, middle stream, and lower stream in wet and dry seasons. The results showed that almost the three study sites had the highest concentration of Σ Heptachlor in both wet and dry seasons (Figure 4.5).

In the upper stream, statistical comparisons of OCPRs concentrations between wet and dry seasons showed that Σ Heptachlor, Σ Endosulfans, and Σ Aldrin were significantly higher in wet season than in dry season (Mann-Whitney U Test, Σ Heptachlor, $Z = -2.310$, $P\text{-value} = 0.021$, $p \leq 0.05$, Σ Endosulfans, $Z = -3.354$, $P\text{-value} = 0.001$, $p \leq 0.05$, and Σ Aldrin, $Z = -3.133$, $P\text{-value} = 0.002$, $p \leq 0.05$). In contrast, the concentrations of Σ BHCs, Σ DDTs, Σ Endrin, and methoxychlor were no significant differences between wet and dry seasons. In the middle stream, the concentrations of Σ DDTs and Σ Endrin were significantly higher in dry season than in wet season (Mann-Whitney U Test, Σ DDTs, $Z = -2.847$, $P\text{-value} = 0.004$, $p \leq 0.05$, Σ Endrin, $Z = -4.126$, $P\text{-value} = 0.000$, $p \leq 0.05$) whereas there were no significant differences for the rests of OCPRs. In the case of the lower stream, the means concentrations of Σ Heptachlor, Σ BHCs, Σ Endosulfans, and Σ Aldrin were significantly higher in wet season than in dry season (Mann-Whitney U Test; Σ Heptachlor, $Z = -3.069$, $P\text{-value} = 0.002$, $p \leq 0.05$, Σ BHCs, $Z = -2.310$, $P\text{-value} = 0.021$, $p \leq 0.05$, Σ Endosulfans, $Z = -3.101$, $P\text{-value} = 0.002$, $p \leq 0.05$, and Σ Aldrin, $Z = -2.246$, $P\text{-value} = 0.025$, $p \leq 0.05$.) which were similarly to many OCPRs found in wet season in the upper stream.

Considering the overall average concentrations of OCPRs, every compound was varied among the different sites. Among the average concentrations of Σ Heptachlor in Figure 4.6, heptachlor were the main compositions in sediment samples both in wet and dry seasons in every location sites. Among the average concentrations of Σ DDTs, 4,4'-DDT was the major contaminant.

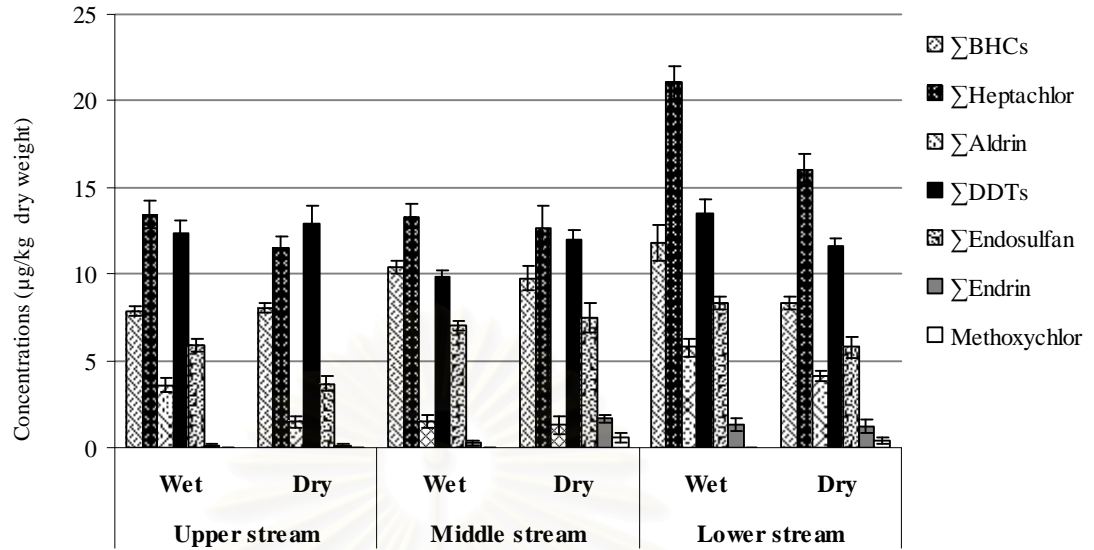


Figure 4.5 Comparisons of average OCPs concentrations (\pm SE) in sediment samples between wet season (June - November 2004) and dry season (December 2004 – May 2005) among upper stream, middle stream, and lower stream in Khlong 7, Rangsit agricultural area in Pathum Thani province.

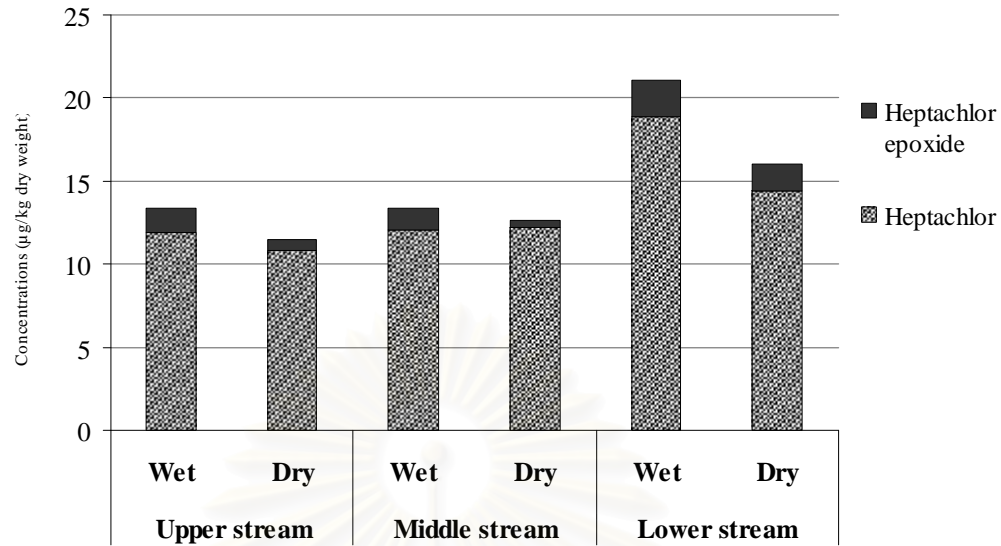


Figure 4.6 Compositions of Σ Heptachlors concentrations including heptachlor and heptachlor epoxide in sediment samples between wet season (June - November 2004) and dry season (December 2004 – May 2005) among upper stream, middle stream, and lower stream at Khlong 7, Rangsit agricultural area in Pathum Thani province.

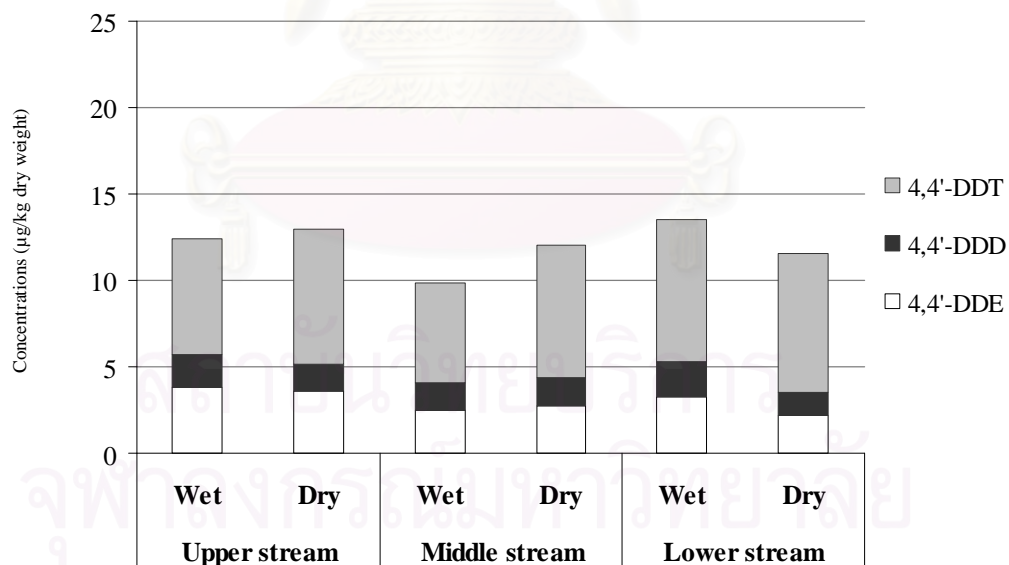


Figure 4.7 Compositions of Σ DDTs concentrations including 4,4'-DDT, 4,4'-DDD, and 4,4'-DDE in sediment samples between wet season (June - November 2004) and dry season (December 2004 – May 2005) among upper stream, middle stream, and lower stream at Khlong 7, Rangsit agricultural area in Pathum Thani province.

Table 4.6 Average concentrations ^a of OCPRs in sediment samples in the upper stream, middle stream, and lower stream in wet season (June - November 2004) and dry season (December 2004 - May 2005); and the average concentrations of OCPRs of the each site overall the year (June 2004 - May 2005) at Khlong 7, Rangsit agricultural area in Pathum Thani province.

OCPRs	Average concentrations of OCPRs in sediment samples (mean ± SE in µg/kg dry weight)								
	Upper stream		Middle stream		Lower stream		Overall the year		
	Wet season (n = 18)	Dry season (n = 18)	Wet season (n = 18)	Dry season (n = 18)	Wet season (n = 18)	Dry season (n = 18)	Upper stream (n = 36)	Middle stream (n = 36)	Lower stream (n = 36)
α-BHC	ND ^a	ND ^a	0.29 ± 0.16 ^a	0.35 ± 0.19 ^a	0.16 ± 0.16 ^a	0.74 ± 0.25 ^a	ND ^a	0.32 ± 0.12 ^b	0.45 ± 0.16 ^b
γ-BHC	4.61 ± 0.14 ^a	4.71 ± 0.14 ^a	5.53 ± 0.20 ^a	5.03 ± 0.15 ^a	5.90 ± 0.22 ^a	4.99 ± 0.22 ^b	4.66 ± 0.10 ^a	5.28 ± 0.13 ^b	5.45 ± 0.17 ^b
β-BHC	2.98 ± 0.17 ^a	2.44 ± 0.14 ^b	2.86 ± 0.08 ^a	3.44 ± 0.39 ^a	5.70 ± 0.82 ^a	2.52 ± 0.10 ^b	2.71 ± 0.12 ^a	3.15 ± 0.20 ^{ab}	4.11 ± 0.49 ^b
δ-BHC	0.28 ± 0.15 ^a	0.87 ± 0.19 ^a	1.69 ± 0.19 ^a	0.96 ± 0.23 ^b	ND ^a	0.08 ± 0.08 ^a	0.57 ± 0.13 ^a	1.32 ± 0.16 ^b	0.04 ± 0.04 ^c
∑BHCs	7.87 ± 0.25 ^a	8.03 ± 0.30 ^a	10.37 ± 0.37 ^a	9.78 ± 0.73 ^a	11.75 ± 1.03 ^a	8.33 ± 0.36 ^b	7.95 ± 0.19 ^a	10.08 ± 0.41 ^b	10.04 ± 0.61 ^b
Heptachlor	11.92 ± 0.82 ^a	10.79 ± 0.56 ^a	12.06 ± 0.50 ^a	12.20 ± 1.10 ^b	18.87 ± 0.88 ^a	14.42 ± 0.86 ^b	11.36 ± 0.50 ^a	12.13 ± 0.60 ^a	16.65 ± 0.71 ^b
Heptachlor epoxide	1.47 ± 0.08 ^a	0.73 ± 0.18 ^b	1.27 ± 0.23 ^a	0.45 ± 0.20 ^a	2.20 ± 0.16 ^a	1.63 ± 0.16 ^b	1.10 ± 0.12 ^a	0.86 ± 0.17 ^a	1.92 ± 0.12 ^b
∑Heptachlor	13.39 ± 0.80 ^a	11.52 ± 0.69 ^b	13.33 ± 0.68 ^a	12.64 ± 1.28 ^a	21.08 ± 0.95 ^a	16.05 ± 0.93 ^b	12.46 ± 0.54 ^a	12.98 ± 0.72 ^a	18.56 ± 0.78 ^b
Aldrin	1.10 ± 0.14 ^a	0.37 ± 0.13 ^b	0.66 ± 0.14 ^a	0.30 ± 0.12 ^a	1.50 ± 0.07 ^a	0.96 ± 0.11 ^b	0.73 ± 0.11 ^a	0.48 ± 0.10 ^a	1.23 ± 0.08 ^b
Dieldrin	2.50 ± 0.30 ^a	1.12 ± 0.26 ^b	0.85 ± 0.35 ^a	0.97 ± 0.39 ^a	4.29 ± 0.43 ^a	3.18 ± 0.21 ^a	1.81 ± 0.23 ^a	0.91 ± 0.26 ^b	3.73 ± 0.25 ^c
∑Aldrin	3.60 ± 0.43 ^a	1.48 ± 0.32 ^b	1.51 ± 0.40 ^a	1.27 ± 0.48 ^a	5.78 ± 0.50 ^a	4.14 ± 0.30 ^b	2.54 ± 0.32 ^a	1.39 ± 0.31 ^b	4.96 ± 0.32 ^c
4,4'-DDE	3.86 ± 0.15 ^a	3.63 ± 0.11 ^a	2.49 ± 0.14 ^a	2.73 ± 0.06 ^b	3.26 ± 0.21 ^a	2.18 ± 0.17 ^b	3.75 ± 0.10 ^a	2.61 ± 0.08 ^b	2.72 ± 0.16 ^b
4,4'-DDD	1.87 ± 0.08 ^a	1.67 ± 0.12 ^b	1.3 ± 0.05 ^a	1.67 ± 0.12 ^a	1.53 ± 0.05 ^a	1.33 ± 0.17 ^b	1.69 ± 0.06 ^a	1.60 ± 0.07 ^a	1.68 ± 0.12 ^a
4,4'-DDT	6.65 ± 0.51 ^a	7.80 ± 0.92 ^a	5.79 ± 0.33 ^a	7.63 ± 0.45 ^b	8.20 ± 0.60 ^a	8.09 ± 0.43 ^a	7.23 ± 0.53 ^{ab}	6.71 ± 0.31 ^a	8.14 ± 0.36 ^b
∑DDTs	12.39 ± 0.68 ^a	12.94 ± 1.02 ^a	9.82 ± 0.42 ^a	12.03 ± 0.49 ^b	13.50 ± 0.86 ^a	11.59 ± 0.48 ^a	12.67 ± 0.61 ^a	10.92 ± 0.37 ^b	12.55 ± 0.51 ^a
Endosulfan I	0.65 ± 0.08 ^a	0.86 ± 0.06 ^b	0.84 ± 0.03 ^a	0.84 ± 0.02 ^a	1.15 ± 0.09 ^a	0.87 ± 0.06 ^b	0.76 ± 0.05 ^a	0.84 ± 0.02 ^a	1.01 ± 0.06 ^b
Endosulfan II	2.62 ± 0.37 ^a	0.92 ± 0.27 ^b	3.15 ± 0.25 ^a	1.96 ± 0.32 ^b	3.01 ± 0.19 ^a	2.13 ± 0.28 ^a	1.77 ± 0.27 ^a	2.56 ± 0.22 ^{ab}	2.57 ± 0.18 ^b
Endosulfan sulfate	2.63 ± 0.17 ^a	1.89 ± 0.21 ^b	3.06 ± 0.06 ^a	4.68 ± 0.83 ^a	4.15 ± 0.22 ^a	2.77 ± 0.32 ^b	2.26 ± 0.15 ^a	3.87 ± 0.43 ^b	3.46 ± 0.22 ^b
∑Endosulfans	5.90 ± 0.37 ^a	3.67 ± 0.41 ^b	7.05 ± 0.26 ^a	7.47 ± 0.86 ^a	8.31 ± 0.35 ^a	5.77 ± 0.63 ^b	4.78 ± 0.33 ^a	7.26 ± 0.44 ^b	7.04 ± 0.41 ^b
Endrin	0.13 ± 0.07 ^a	ND ^a	ND ^a	1.56 ± 0.26 ^b	0.77 ± 0.17 ^a	0.83 ± 0.39 ^a	0.06 ± 0.04 ^a	0.78 ± 0.18 ^b	0.80 ± 0.21 ^b
Endrin aldehyde	ND ^a	0.09 ± 0.09 ^a	0.24 ± 0.16 ^a	0.11 ± 0.11 ^a	0.57 ± 0.27 ^a	0.38 ± 0.21 ^a	0.04 ± 0.04 ^a	0.17 ± 0.10 ^a	0.47 ± 0.17 ^a
∑Endrin	0.13 ± 0.07 ^a	0.09 ± 0.09 ^a	0.24 ± 0.16 ^a	1.67 ± 0.24 ^b	1.34 ± 0.37 ^a	1.21 ± 0.40 ^a	0.11 ± 0.05 ^a	0.95 ± 0.19 ^b	1.27 ± 0.27 ^b
Methoxychlor	ND ^a	ND ^a	ND ^a	0.55 ± 0.30 ^a	ND ^a	0.39 ± 0.21 ^a	ND ^a	0.27 ± 0.15 ^a	0.19 ± 0.11 ^a

^a the average concentrations, in each data block: upper stream, middle stream, lower stream and overall the year data blocks, with the same letter in the same row are not significantly different at $p \leq 0.05$.

ND: samples with organochlorine pesticide concentrations below limit of detection (LOD), n: number of sampling

4.3.3 Aquatic invertebrates

The overall average concentrations of OCPRs in aquatic invertebrate samples such as Lanchester's freshwater prawn, apple snail, and freshwater snail samples in different study sites such as upper stream, middle stream, and lower stream from wet season and dry season were summarized in Table 4.7, 4.8, and 4.9. Almost three location sites showed the concentrations of Σ DDTs were highest in both wet and dry seasons.

In Figure 4.8, Σ DDTs concentrations in freshwater snail and apple snail samples in the middle stream presented the same tendency that the highest peak of Σ DDTs (274.84 ± 12.02 and 143.25 ± 9.59 $\mu\text{g}/\text{kg}$ wet weight for freshwater snail and apple snail, respectively) were significantly higher in dry season than in wet season (Mann-Whitney U Test, in apple snail, $Z = -3.504$, $P = 0.000$, $p \leq 0.05$ and in freshwater snail, $Z = -2.496$, $P = 0.013$, $p \leq 0.05$) whereas Σ DDTs concentrations in Lanchester's freshwater prawn were no difference between wet and dry seasons in the same location. To determine the other sites such as upper stream and lower stream, the average Σ DDTs concentrations were not significantly different between wet and dry seasons from the both sites in all aquatic invertebrates except in apple snail samples from upper stream (Mann-Whitney U Test, $Z = -3.667$, $P = 0.000$, $p \leq 0.05$). Among Σ DDTs in the aquatic invertebrates, the average concentrations of 4,4'-DDT were higher than its degradation products, 4,4'-DDD and 4,4'-DDE (Table 4.7 and Figure 4.8). To determine the comparisons of Σ DDTs concentrations among the upper, middle, and lower stream overall the year, the results presented that there were not different Σ DDTs concentrations among the three sites in both apple snail and freshwater snail whereas the Σ DDTs concentrations in the upper stream were significantly higher than in the middle and lower streams in Lanchester's freshwater prawn (Tamhane's T2, $P < 0.05$).

Similarly to Σ DDTs, in the middle stream, the average concentrations of Σ BHCs in both apple snail and freshwater snail samples in dry season (77.62 ± 9.01 and 70.92 ± 3.53 $\mu\text{g}/\text{kg}$ wet weight for apple snail and freshwater snail, respectively) were significant higher in dry season than in wet season (Mann-Whitney U Test, in apple snail, $Z = -3.503$, $P = 0.000$, $p \leq 0.05$ and in freshwater snail, $Z = -2.496$, $P =$

0.013, $p \leq 0.05$), but there were not different between wet and dry seasons in Lanchester's freshwater prawn in the same location site. In the lower stream, the average concentrations of Σ BHCs were not significantly different between wet and dry seasons in all aquatic invertebrate samples. In the upper stream, average Σ BHCs concentrations were varied in each invertebrate samples. In freshwater snail samples, Σ BHCs concentrations were not different between wet and dry seasons. In contrast, in Lanchester's freshwater prawn samples, the concentrations in dry season were statistically higher than in wet season (Mann-Whitney U Test, $Z = -3.600$, $P = 0.000$, $p \leq 0.05$); but in apple snail samples, the concentrations in dry season were significantly lower than in wet season (Mann-Whitney U Test, $Z = -4.025$, $P = 0.000$, $p \leq 0.05$). To determine the comparisons of average Σ BHCs concentrations among the upper, middle, and lower stream overall the year, the results showed that there were not significantly different these among three sites in every aquatic invertebrate sample such as Lanchester's freshwater prawn, apple snail, and freshwater snail samples. To determine the compositions of Σ BHCs concentrations, β -isomer was dominant in almost aquatic invertebrate samples, but γ -isomer was relatively low in all samples.

Among Σ Endosulfans concentrations, there were not significantly different between wet season and dry season from the upper, middle, and lower stream in freshwater snail samples. For Σ Endosulfans concentrations in Lanchester's freshwater prawn, the concentrations were no differences between wet and dry seasons from the upper stream and middle stream, but significant differences in the lower stream (Mann-Whitney U Test, $Z = -2.278$, $P = 0.023$, $p \leq 0.05$). For Σ Endosulfans concentrations in apple snail, there were not significantly different between wet and dry seasons in lower stream. However, in the middle stream, Σ Endosulfans concentrations in wet season were significantly lower than in dry season (Mann-Whitney U Test, $Z = -2.335$, $P = 0.020$, $p \leq 0.05$) whereas in the upper stream, the concentrations in wet season were statistically higher than in dry season (Mann-Whitney U Test, $Z = -3.221$, $P = 0.001$, $p \leq 0.05$). The comparisons of average Σ Endosulfans concentrations among the upper, middle, and lower stream overall the year showed that there were not significantly different among the three location sites in both apple snail and freshwater snail samples while the average Σ Endosulfans concentrations in lower stream were significantly lower than in the middle stream

(Tamhane's T2, $P < 0.05$). Among Σ Endosulfans concentrations, Figure 4.9 showed that the major composition was endosulfan sulfate.

Figure 4.8, 4.9, and 4.10 presented the average concentrations of three dominant OCPRs such as Σ DDTs, Σ BHCs, and Σ Endosulfans. All of them showed the same trend that the peak showed that the highest peaks were shown in the middle stream. Almost average concentrations of Σ DDTs, Σ BHCs, and Σ Endosulfans were higher in dry season than in wet season in every organism in the middle stream.



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Table 4.7 Average concentrations ^a of OCPs in Lanchester's freshwater prawn samples in the upper stream, middle stream, and lower stream in wet season (June - November 2004) and dry season (December 2004 - May 2005); and the average concentrations of OCPs of the each site overall the year (June 2004 - May 2005) at Khlong 7, Rangsit agricultural area in Pathum Thani province.

OCPs	Average concentrations of OCPs in Lanchester's freshwater prawn samples (mean ± SE in µg/kg wet weight)								
	Upper stream		Middle stream		Lower stream		Overall the year		
	Wet season (n = 12)	Dry season (n = 18)	Wet season (n = 12)	Dry season (n = 18)	Wet season (n = 15)	Dry season (n = 18)	Upper stream (n = 30)	Middle stream (n = 30)	Lower stream (n = 33)
α-BHC	3.49 ± 1.83 ^a	4.39 ± 1.49 ^a	1.94 ± 1.03 ^a	1.27 ± 0.66 ^a	4.05 ± 1.75 ^a	1.68 ± 0.64 ^a	4.03 ± 1.14 ^a	1.54 ± 0.56 ^a	2.76 ± 0.88 ^a
γ-BHC	1.83 ± 0.71 ^a	2.62 ± 0.58 ^a	0.81 ± 0.34 ^a	3.22 ± 0.90 ^a	2.85 ± 0.77 ^a	2.41 ± 1.08 ^a	2.30 ± 0.45 ^a	2.25 ± 0.59 ^a	2.61 ± 0.68 ^a
β-BHC	7.01 ± 3.29 ^a	23.40 ± 3.10 ^b	5.54 ± 1.93 ^a	11.53 ± 1.86 ^b	6.94 ± 1.74 ^a	14.53 ± 1.62 ^b	16.84 ± 2.69 ^a	9.13 ± 1.44 ^b	11.08 ± 1.34 ^{ab}
δ-BHC	1.72 ± 0.65 ^a	6.51 ± 2.16 ^a	8.84 ± 2.86 ^a	11.12 ± 4.65 ^a	13.98 ± 6.67 ^a	13.27 ± 5.58 ^a	4.59 ± 1.38 ^a	10.21 ± 2.98 ^a	13.59 ± 4.23 ^a
∑BHCs	14.05 ± 3.93 ^a	36.92 ± 2.87 ^b	17.13 ± 3.18 ^a	27.14 ± 4.13 ^a	27.83 ± 8.34 ^a	31.88 ± 5.00 ^a	27.77 ± 3.09 ^a	23.13 ± 2.89 ^a	30.04 ± 4.61 ^a
Heptachlor	15.73 ± 2.21 ^a	13.66 ± 1.51 ^a	14.42 ± 2.20 ^a	8.71 ± 1.30 ^b	11.22 ± 1.75 ^a	12.44 ± 2.08 ^a	14.49 ± 1.25 ^a	10.99 ± 1.26 ^a	11.88 ± 1.37 ^a
Heptachlor epoxide	0.44 ± 0.19 ^a	1.99 ± 1.09 ^a	2.46 ± 0.57 ^a	2.41 ± 0.66 ^a	1.39 ± 0.75 ^a	3.29 ± 1.16 ^a	1.37 ± 0.67 ^a	2.43 ± 0.45 ^a	2.43 ± 0.73 ^a
∑Heptachlor	16.17 ± 2.08 ^a	15.65 ± 1.95 ^a	16.88 ± 1.74 ^a	11.12 ± 1.29 ^b	12.61 ± 1.55 ^a	15.73 ± 2.92 ^a	15.86 ± 1.41 ^a	13.42 ± 1.15 ^a	14.31 ± 1.74 ^a
Aldrin	3.27 ± 0.55 ^a	5.65 ± 1.10 ^a	3.01 ± 0.63 ^a	2.64 ± 0.41 ^a	3.14 ± 0.51 ^a	3.12 ± 0.25 ^a	4.70 ± 0.72 ^a	2.79 ± 0.34 ^a	3.13 ± 0.26 ^a
Dieldrin	2.52 ± 1.20 ^a	0.81 ± 0.44 ^a	3.25 ± 1.41 ^a	3.52 ± 1.10 ^a	1.58 ± 0.70 ^a	2.70 ± 0.71 ^a	1.50 ± 0.56 ^a	3.41 ± 0.85 ^a	2.19 ± 0.50 ^a
∑Aldrin	5.79 ± 1.38 ^a	6.46 ± 1.07 ^a	6.26 ± 1.61 ^a	6.16 ± 1.37 ^a	4.72 ± 1.00 ^a	5.82 ± 0.55 ^a	6.19 ± 0.83 ^a	6.20 ± 1.02 ^a	5.32 ± 0.54 ^a
4,4'-DDE	4.69 ± 0.36 ^a	7.34 ± 1.05 ^b	4.51 ± 1.01 ^a	4.81 ± 1.04 ^a	2.97 ± 0.26 ^a	5.26 ± 1.16 ^a	6.28 ± 0.68 ^a	4.69 ± 0.73 ^a	4.22 ± 0.67 ^a
4,4'-DDD	1.25 ± 0.61 ^a	4.32 ± 2.28 ^a	2.50 ± 1.28 ^a	0.35 ± 0.24 ^a	2.49 ± 1.33 ^a	2.04 ± 0.86 ^a	3.09 ± 1.40 ^a	1.21 ± 0.56 ^a	2.24 ± 0.75 ^a
4,4'-DDT	28.46 ± 2.87 ^a	118.40 ± 32.18 ^a	15.59 ± 3.28 ^a	39.85 ± 10.21 ^a	20.95 ± 4.16 ^a	31.64 ± 10.37 ^a	82.42 ± 20.79 ^a	30.15 ± 6.57 ^{ab}	26.78 ± 5.96 ^b
∑DDTs	34.41 ± 3.25 ^a	130.06 ± 31.87 ^a	22.60 ± 4.90 ^a	45.00 ± 10.88 ^a	26.41 ± 5.32 ^a	38.94 ± 10.26 ^a	91.80 ± 20.85 ^a	36.04 ± 7.03 ^b	33.24 ± 6.11 ^b
Endosulfan I	3.55 ± 0.84 ^a	4.48 ± 0.74 ^a	2.68 ± 1.09 ^a	3.75 ± 0.83 ^a	2.07 ± 0.88 ^a	3.14 ± 0.42 ^a	4.11 ± 0.56 ^a	3.32 ± 0.66 ^a	2.65 ± 0.46 ^a
Endosulfan II	8.64 ± 1.75 ^a	7.59 ± 2.68 ^a	6.43 ± 1.67 ^a	2.66 ± 1.23 ^b	6.38 ± 1.12 ^a	2.35 ± 1.04 ^b	8.01 ± 1.73 ^a	4.17 ± 1.04 ^a	4.18 ± 0.83 ^a
Endosulfan sulfate	20.10 ± 8.03 ^a	32.33 ± 15.89 ^a	33.20 ± 13.68 ^a	58.71 ± 19.66 ^a	5.10 ± 1.29 ^a	13.52 ± 6.31 ^a	27.44 ± 9.99 ^{ab}	48.51 ± 13.03 ^a	9.69 ± 3.52 ^b
∑Endosulfans	32.29 ± 9.68 ^a	44.40 ± 14.85 ^a	42.31 ± 15.24 ^a	65.12 ± 20.14 ^a	13.55 ± 1.45 ^a	19.01 ± 6.73 ^b	39.55 ± 9.64 ^{ab}	56.00 ± 13.50 ^a	16.53 ± 3.71 ^b
Endrin	1.73 ± 0.55 ^a	8.90 ± 3.33 ^a	1.34 ± 0.44 ^a	1.11 ± 0.47 ^a	2.32 ± 0.51 ^a	2.37 ± 0.72 ^a	6.03 ± 2.09 ^a	1.21 ± 0.33 ^a	2.35 ± 0.45 ^a
Endrin aldehyde	ND ^a	6.40 ± 2.68 ^b	ND ^a	5.11 ± 2.20 ^b	0.73 ± 0.39 ^a	1.58 ± 0.86 ^a	3.84 ± 1.69 ^a	3.07 ± 1.38 ^a	1.19 ± 0.50 ^a
∑Endrin	1.73 ± 0.55 ^a	15.30 ± 5.41 ^b	1.34 ± 0.44 ^a	6.22 ± 2.09 ^b	3.05 ± 0.72 ^a	3.95 ± 0.91 ^a	9.87 ± 3.45 ^a	4.27 ± 1.33 ^a	3.54 ± 0.59 ^a
Methoxychlor	2.74 ± 0.91 ^a	4.36 ± 2.60 ^a	3.15 ± 0.93 ^a	0.12 ± 0.09 ^b	2.81 ± 1.36 ^a	ND ^b	3.71 ± 1.59 ^a	1.33 ± 0.46 ^a	1.28 ± 0.65 ^a

^a the average concentrations, in each data block: upper stream, middle stream, lower stream and overall the year data blocks, with the same letter in the same row are not significantly different, $p \leq 0.05$.

ND: samples with organochlorine pesticide concentrations below limit of detection (LOD), n: number of sampling

Table 4.8 Average concentrations ^a of OCPs in apple snail samples in the upper stream, middle stream, and lower stream in wet season (June - November 2004) and dry season (December 2004 - May 2005); and the average concentrations of OCPs of the each site overall the year (June 2004 - May 2005) at Khlong 7, Rangsit agricultural area in Pathum Thani province.

OCPs	Average concentrations of OCPs in apple snail samples (mean ± SE in µg/kg wet weight)								
	Upper stream		Middle stream		Lower stream		Overall the year		
	Wet season (n = 9)	Dry season (n = 15)	Wet season (n = 15)	Dry season (n = 6)	Wet season (n = 12)	Dry season (n = 15)	Upper stream (n = 24)	Middle stream (n = 21)	Lower stream (n = 27)
α-BHC	15.65 ± 0.89 ^a	5.06 ± 0.67 ^b	6.59 ± 1.56 ^a	27.75 ± 6.53 ^b	9.31 ± 2.24 ^a	8.52 ± 1.90 ^a	9.03 ± 1.19 ^a	12.64 ± 2.97 ^a	8.87 ± 1.43 ^a
γ-BHC	ND ^a	0.73 ± 0.39 ^a	ND ^a	ND ^a	0.53 ± 0.28 ^a	2.37 ± 1.27 ^a	0.46 ± 0.25 ^a	ND ^a	1.55 ± 0.73 ^a
β-BHC	28.98 ± 3.12 ^a	13.34 ± 1.78 ^b	11.02 ± 1.49 ^a	22.16 ± 9.91 ^a	20.46 ± 2.64 ^a	20.78 ± 2.77 ^a	19.21 ± 2.23 ^a	14.21 ± 3.06 ^a	20.64 ± 1.90 ^a
δ-BHC	9.31 ± 3.13 ^a	2.52 ± 0.57 ^a	1.17 ± 0.62 ^a	27.71 ± 12.39 ^a	2.49 ± 0.80 ^a	3.11 ± 0.58 ^a	5.07 ± 1.37 ^a	8.75 ± 4.28 ^a	2.84 ± 0.47 ^a
∑BHCs	53.94 ± 3.57 ^a	21.65 ± 2.32 ^b	18.78 ± 3.29 ^a	77.62 ± 9.01 ^b	32.79 ± 3.71 ^a	34.78 ± 5.44 ^a	33.76 ± 3.79 ^a	35.59 ± 6.82 ^a	33.90 ± 3.39 ^a
Heptachlor	23.35 ± 1.81 ^a	10.15 ± 0.77 ^b	10.61 ± 1.66 ^a	34.94 ± 7.67 ^b	15.48 ± 3.11 ^a	12.83 ± 2.60 ^a	15.10 ± 1.56 ^a	17.56 ± 3.41 ^a	14.01 ± 1.98 ^a
Heptachlor epoxide	ND ^a	1.96 ± 0.65 ^b	5.11 ± 1.32 ^a	5.19 ± 2.32 ^a	6.03 ± 1.65 ^a	3.34 ± 1.05 ^a	1.23 ± 0.44 ^a	5.13 ± 1.12 ^b	4.53 ± 0.95 ^b
∑Heptachlor	23.35 ± 1.81 ^a	12.11 ± 0.92 ^b	15.72 ± 2.77 ^a	40.13 ± 5.35 ^b	21.51 ± 4.69 ^a	16.18 ± 3.45 ^a	16.33 ± 1.43 ^a	22.69 ± 3.46 ^a	18.54 ± 2.82 ^a
Aldrin	14.06 ± 2.21 ^a	4.00 ± 0.53 ^b	6.54 ± 1.53 ^a	3.69 ± 1.65 ^a	7.62 ± 0.86 ^a	5.74 ± 1.20 ^a	7.77 ± 1.33 ^a	5.73 ± 1.20 ^a	4.53 ± 0.95 ^a
Dieldrin	6.07 ± 3.03 ^a	8.27 ± 1.16 ^a	3.27 ± 1.55 ^a	26.02 ± 11.64 ^a	13.52 ± 3.01 ^a	6.66 ± 1.98 ^b	7.44 ± 1.33 ^a	9.77 ± 4.02 ^a	9.71 ± 1.82 ^a
∑Aldrin	20.12 ± 5.17 ^a	12.27 ± 1.64 ^a	9.81 ± 2.26 ^a	29.71 ± 13.29 ^a	21.14 ± 3.85 ^a	12.40 ± 2.98 ^b	15.21 ± 2.27 ^a	15.50 ± 4.38 ^a	16.28 ± 2.48 ^a
4,4'-DDE	8.06 ± 4.03 ^a	5.20 ± 0.84 ^a	1.20 ± 0.64 ^a	27.05 ± 12.10 ^a	12.39 ± 4.24 ^a	5.65 ± 1.91 ^a	6.27 ± 1.57 ^a	8.58 ± 4.18 ^a	8.65 ± 2.21 ^a
4,4'-DDD	18.63 ± 5.17 ^a	1.07 ± 0.57 ^b	ND ^a	45.91 ± 8.03 ^b	11.26 ± 5.63 ^a	5.50 ± 2.94 ^a	7.65 ± 2.60 ^a	13.12 ± 5.11 ^a	8.06 ± 2.98 ^a
4,4'-DDT	56.83 ± 2.46 ^a	21.81 ± 4.25 ^b	21.28 ± 3.13 ^a	70.29 ± 10.55 ^b	20.50 ± 3.27 ^a	25.13 ± 4.49 ^a	34.94 ± 4.49 ^a	35.28 ± 6.11 ^a	23.07 ± 2.87 ^a
∑DDTs	83.51 ± 9.03 ^a	28.08 ± 4.88 ^b	22.47 ± 3.19 ^a	143.25 ± 9.59 ^b	44.15 ± 12.77 ^a	36.28 ± 6.95 ^a	48.87 ± 7.14 ^a	56.98 ± 12.67 ^a	39.78 ± 6.76 ^a
Endosulfan I	21.10 ± 10.55 ^a	4.50 ± 0.34 ^a	7.54 ± 2.68 ^a	22.70 ± 10.15 ^a	3.34 ± 0.81 ^a	5.48 ± 1.24 ^a	10.72 ± 4.17 ^a	11.87 ± 3.65 ^a	4.53 ± 0.79 ^a
Endosulfan II	ND ^a	3.27 ± 1.13 ^b	12.00 ± 3.43 ^a	11.51 ± 5.15 ^a	16.69 ± 5.40 ^a	15.15 ± 5.01 ^a	2.04 ± 0.77 ^a	11.86 ± 2.79 ^b	15.84 ± 3.61 ^b
Endosulfan sulfate	27.47 ± 3.20 ^a	5.62 ± 0.80 ^b	18.81 ± 4.15 ^a	32.53 ± 3.68 ^b	23.25 ± 4.79 ^a	12.41 ± 3.23 ^b	13.81 ± 2.54 ^a	22.73 ± 3.39 ^a	17.22 ± 2.92 ^a
∑Endosulfans	48.57 ± 12.25 ^a	13.38 ± 1.38 ^b	38.36 ± 9.95 ^a	66.74 ± 8.68 ^b	43.28 ± 9.73 ^a	33.04 ± 9.32 ^a	26.58 ± 5.74 ^a	46.46 ± 7.94 ^a	37.59 ± 6.69 ^a
Endrin	34.97 ± 8.92 ^a	0.31 ± 0.18 ^b	0.51 ± 0.28 ^a	40.52 ± 18.12 ^a	ND ^a	7.59 ± 2.87 ^b	13.31 ± 4.76 ^a	11.95 ± 6.31 ^a	4.22 ± 1.74 ^a
Endrin aldehyde	26.62 ± 2.69 ^a	5.02 ± 1.46 ^b	7.28 ± 2.41 ^a	ND ^a	ND ^a	6.57 ± 2.17 ^b	13.12 ± 2.55 ^a	5.20 ± 1.86 ^b	3.65 ± 1.35 ^b
∑Endrin	61.59 ± 9.94 ^a	5.33 ± 1.40 ^b	7.79 ± 2.31 ^a	40.52 ± 18.12 ^a	ND ^a	14.16 ± 4.50 ^b	26.43 ± 6.77 ^a	17.14 ± 6.09 ^{ab}	7.87 ± 2.82 ^b
Methoxychlor	11.56 ± 5.80 ^a	9.40 ± 0.77 ^a	7.65 ± 2.32 ^a	ND ^b	6.62 ± 2.53 ^a	3.22 ± 1.73 ^a	10.21 ± 2.16 ^a	5.46 ± 8.30 ^a	4.73 ± 1.49 ^a

^a the average concentrations, in each data block: upper stream, middle stream, lower stream and overall the year data blocks, with the same letter in the same row are not significantly different at $p \leq 0.05$.

ND: samples with organochlorine pesticide concentrations below limit of detection (LOD), n: number of sampling

Table 4.9 Average concentrations ^a of OCPRs in freshwater snail samples in the upper stream, middle stream, and lower stream in wet season (June - November 2004) and dry season (December 2004 - May 2005); and the average concentrations of OCPRs of the each site overall the year (June 2004 - May 2005) at Khlong 7, Rangsit agricultural area in Pathum Thani province.

OCPRs	Average concentrations of OCPRs in freshwater snail samples (mean ± SE in µg/kg wet weight)								
	Upper stream		Middle stream		Lower stream		Overall the year		
	Wet season (n = 12)	Dry season (n = 18)	Wet season (n = 9)	Dry season (n = 3)	Wet season (n = 6)	Dry season (n = 9)	Upper stream (n = 30)	Middle stream (n = 12)	Lower stream (n = 15)
α-BHC	10.23 ± 1.86 ^a	9.11 ± 0.86 ^a	7.51 ± 1.81 ^a	19.38 ± 2.37 ^b	5.51 ± 2.47 ^a	9.05 ± 0.93 ^a	9.55 ± 0.89 ^a	10.47 ± 2.11 ^a	7.64 ± 1.17 ^a
γ-BHC	3.12 ± 1.66 ^a	ND ^b	ND ^a	2.10 ± 2.10 ^a	7.12 ± 3.19 ^a	ND ^b	1.25 ± 0.71 ^a	0.53 ± 0.53 ^a	2.85 ± 1.52 ^a
β-BHC	21.77 ± 3.65 ^a	22.91 ± 2.58 ^a	16.89 ± 2.25 ^a	41.13 ± 2.43 ^b	54.65 ± 22.60 ^a	20.08 ± 1.53 ^a	22.45 ± 2.09 ^a	22.95 ± 3.61 ^a	33.91 ± 9.71 ^a
δ-BHC	9.13 ± 4.18 ^a	1.63 ± 0.59 ^a	9.30 ± 3.15 ^a	8.31 ± 0.09 ^a	11.45 ± 5.12 ^a	2.49 ± 1.31 ^a	4.63 ± 1.80 ^a	9.05 ± 2.33 ^a	6.07 ± 2.39 ^a
∑BHCs	44.24 ± 10.17 ^a	33.65 ± 2.90 ^a	33.70 ± 6.90 ^a	70.92 ± 3.53 ^b	78.73 ± 33.36 ^a	31.62 ± 2.98 ^a	37.88 ± 4.42 ^a	43.01 ± 7.08 ^a	50.46 ± 14.15 ^a
Heptachlor	17.31 ± 2.80 ^a	12.46 ± 1.41 ^a	14.42 ± 3.78 ^a	27.81 ± 1.09 ^b	11.74 ± 5.25 ^a	9.89 ± 2.50 ^a	14.40 ± 1.44 ^a	17.77 ± 3.30 ^a	10.63 ± 2.48 ^a
Heptachlor epoxide	1.75 ± 0.92 ^a	ND ^b	3.00 ± 1.44 ^a	7.15 ± 0.11 ^a	32.94 ± 11.90 ^a	3.11 ± 1.59 ^b	0.70 ± 0.39 ^a	2.25 ± 1.14 ^a	15.04 ± 6.03 ^b
∑Heptachlor	19.06 ± 2.64 ^a	12.46 ± 1.41 ^b	17.42 ± 3.88 ^a	27.81 ± 1.09 ^a	44.68 ± 17.15 ^a	13.00 ± 1.04 ^a	15.10 ± 1.45 ^a	20.02 ± 3.18 ^a	25.67 ± 7.72 ^a
Aldrin	7.02 ± 1.11 ^a	9.46 ± 0.87 ^a	4.49 ± 0.91 ^a	7.15 ± 0.11 ^b	1.01 ± 0.45 ^a	8.07 ± 0.65 ^b	8.48 ± 0.71 ^a	5.16 ± 0.76 ^b	5.24 ± 1.01 ^b
Dieldrin	9.63 ± 3.71 ^a	15.79 ± 1.60 ^a	12.34 ± 2.33 ^a	ND ^b	ND ^a	15.47 ± 1.61 ^b	13.33 ± 1.82 ^a	9.25 ± 2.35 ^a	9.28 ± 2.23 ^a
∑Aldrin	16.65 ± 4.00 ^a	25.25 ± 2.26 ^a	16.83 ± 2.86 ^a	7.15 ± 0.11 ^a	1.01 ± 0.45 ^a	23.53 ± 2.22 ^b	21.81 ± 2.20 ^a	14.41 ± 2.46 ^a	14.52 ± 3.23 ^a
4,4'-DDE	18.22 ± 3.53 ^s	17.59 ± 1.74 ^a	12.23 ± 3.28 ^a	18.90 ± 0.02 ^a	8.29 ± 1.68 ^a	17.39 ± 1.32 ^b	17.84 ± 1.72 ^a	13.90 ± 2.58 ^a	13.75 ± 1.56 ^a
4,4'-DDD	10.19 ± 5.37 ^s	5.49 ± 3.03 ^a	ND ^a	33.82 ± 0.93 ^b	26.46 ± 11.83 ^a	ND ^b	7.37 ± 2.79 ^a	8.46 ± 4.42 ^a	10.58 ± 5.66 ^a
4,4'-DDT	68.49 ± 18.14 ^a	44.53 ± 6.09 ^a	31.19 ± 7.39 ^a	195.12 ± 11.36 ^b	58.01 ± 13.28 ^a	34.55 ± 3.31 ^a	54.12 ± 8.23 ^a	72.17 ± 22.22 ^a	43.93 ± 6.20 ^a
∑DDTs	96.90 ± 23.54 ^a	67.61 ± 10.34 ^a	43.42 ± 10.63 ^a	247.84 ± 12.02 ^b	92.76 ± 26.74 ^a	51.94 ± 3.63 ^a	79.33 ± 11.35 ^a	94.53 ± 27.94 ^a	68.26 ± 11.63 ^a
Endosulfan I	1.67 ± 0.55 ^a	11.98 ± 1.82 ^b	10.26 ± 3.09 ^a	8.97 ± 0.09 ^a	1.06 ± 0.56 ^a	11.17 ± 1.27 ^b	7.85 ± 1.44 ^a	9.93 ± 2.29 ^a	7.13 ± 1.53 ^a
Endosulfan II	ND ^a	ND ^a	ND ^a	ND ^a	20.43 ± 9.28 ^a	2.99 ± 1.52 ^a	ND ^a	ND ^a	9.97 ± 4.28 ^b
Endosulfan sulfate	25.64 ± 9.12 ^a	8.90 ± 3.08 ^a	24.21 ± 10.83 ^a	ND ^a	24.21 ± 10.83 ^a	17.66 ± 4.84 ^a	15.60 ± 4.28 ^a	17.11 ± 5.20 ^a	20.28 ± 5.05 ^a
∑Endosulfans	27.30 ± 8.80 ^a	20.88 ± 4.41 ^a	33.07 ± 7.94 ^a	8.97 ± 0.09 ^a	45.70 ± 19.58 ^a	31.82 ± 4.20 ^a	23.45 ± 4.35 ^a	27.05 ± 6.65 ^a	37.37 ± 8.01 ^a
Endrin	12.18 ± 1.98 ^a	4.86 ± 2.46 ^b	7.87 ± 3.96 ^a	10.90 ± 0.06 ^a	20.64 ± 9.25 ^a	3.59 ± 1.84 ^a	7.79 ± 1.78 ^a	8.63 ± 2.95 ^a	10.41 ± 4.29 ^a
Endrin aldehyde	ND ^a	ND ^a	4.31 ± 4.31 ^a	ND ^a	ND ^a	ND ^a	ND ^a	3.23 ± 3.23 ^a	ND ^a
∑Endrin	12.18 ± 1.98 ^a	4.86 ± 2.46 ^b	12.19 ± 6.95 ^a	10.90 ± 0.06 ^a	20.64 ± 9.25 ^a	3.59 ± 1.84 ^a	7.79 ± 1.78 ^a	11.86 ± 5.13 ^a	10.41 ± 4.29 ^a
Methoxychlor	1.83 ± 0.96 ^a	7.02 ± 1.85 ^a	9.20 ± 2.56 ^a	ND ^a	ND ^a	8.44 ± 4.22 ^a	4.94 ± 1.25 ^a	6.90 ± 2.24 ^a	5.06 ± 2.71 ^a

^a the average concentrations, in each data block: upper stream, middle stream, lower stream and overall the year data blocks, with the same letter in the same row are not significantly different at $p \leq 0.05$.

ND: samples with organochlorine pesticide concentrations below limit of detection (LOD), n: number of sampling

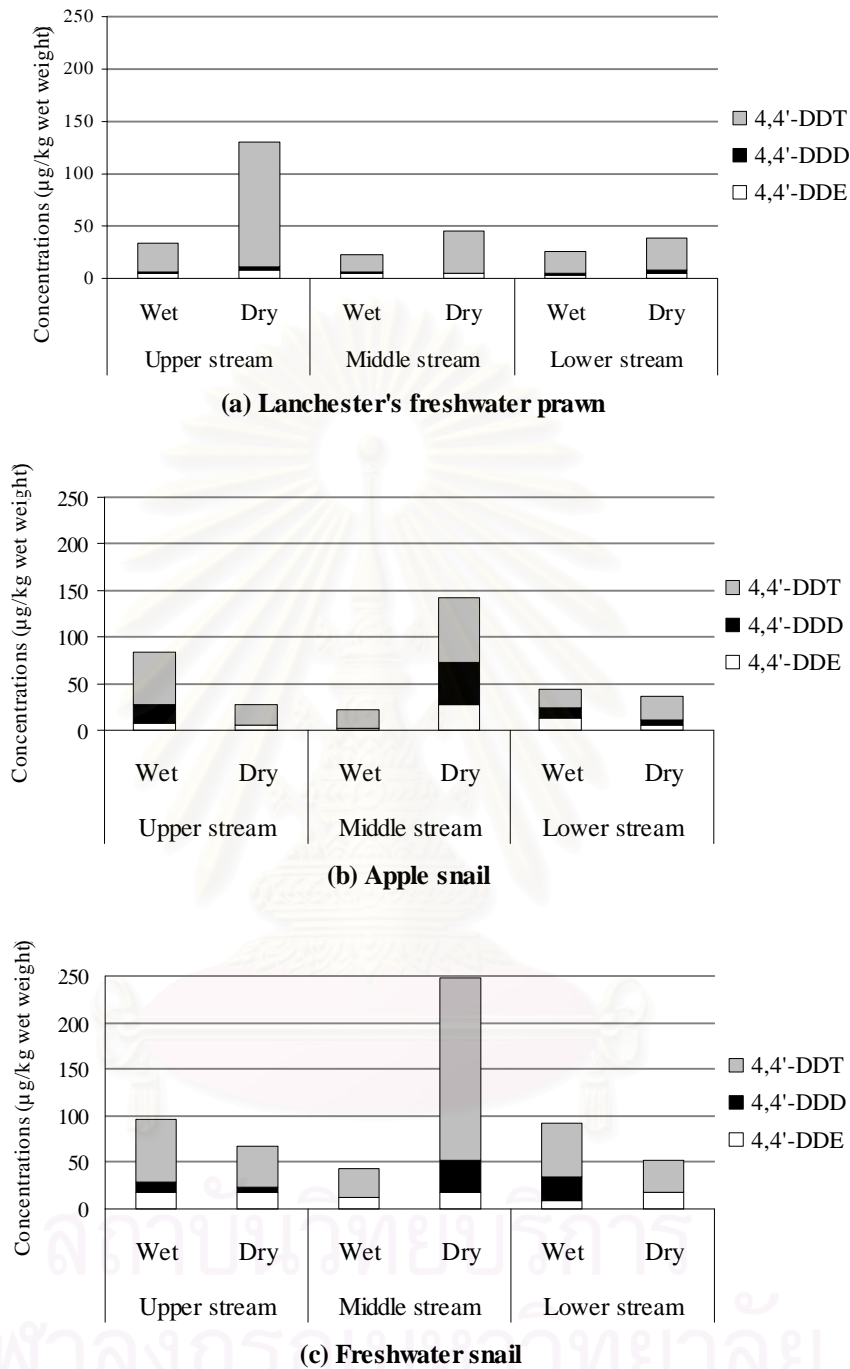
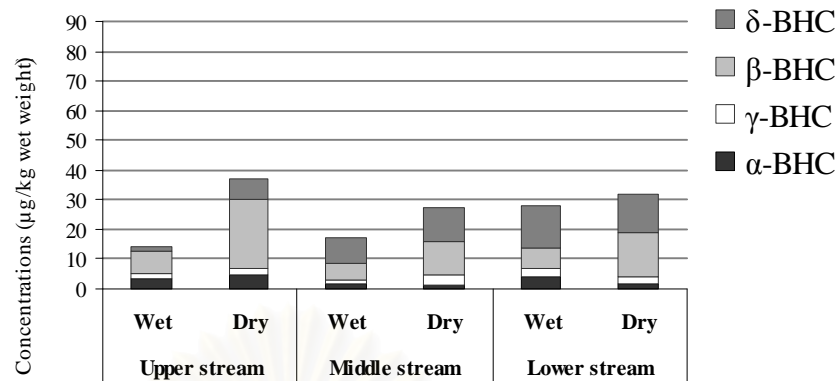
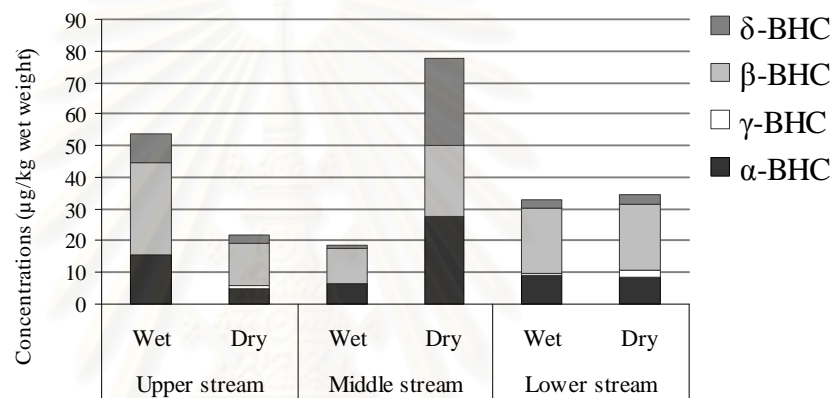


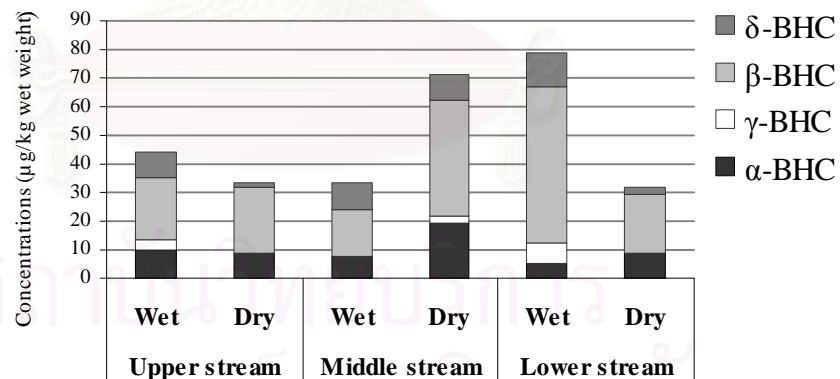
Figure 4.8 Compositions of Σ DDTs concentrations including 4,4'-DDT, 4,4'-DDD, and 4,4'-DDE in (a) Lanchester's freshwater prawn, (b) apple snail, and (c) freshwater snail samples between wet and dry season among upper stream, middle stream, and lower stream at Khlong 7, Rangsit agricultural area in Pathum Thani province collected once a month from June 2004 to May 2005.



(a) Lanchester's freshwater prawn

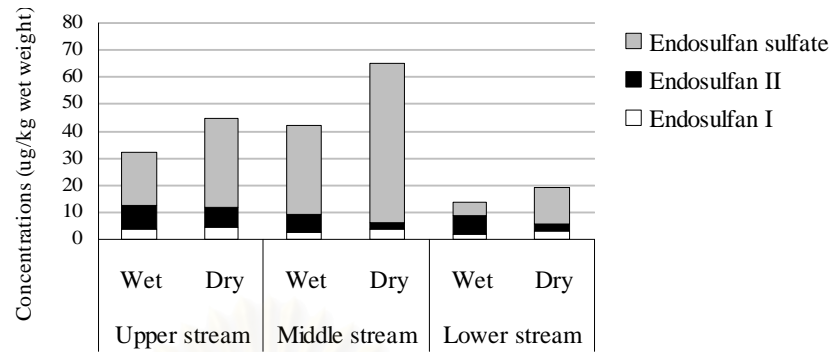


(b) Apple snail

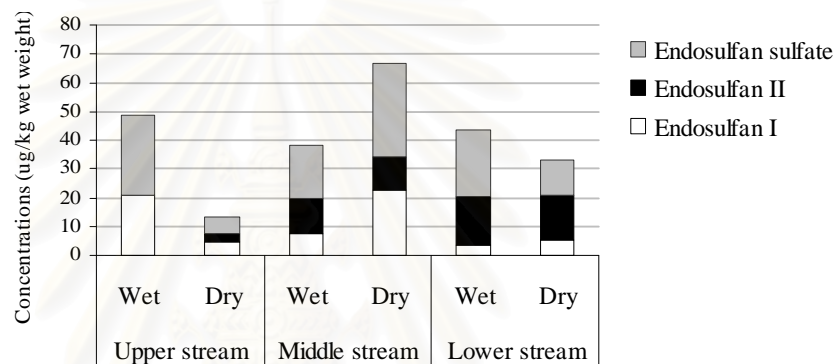


(c) Freshwater snail

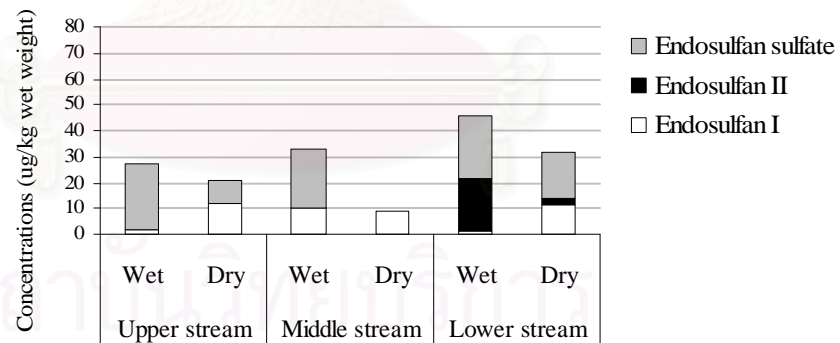
Figure 4.9 Compositions of Σ BHCs concentrations including α -BHC, β -BHC, γ -BHC, and δ -BHC in (a) Lanchester's freshwater prawn, (b) apple snail, and (c) freshwater snail samples between wet and dry season among upper stream, middle stream, and lower stream at Khlong 7, Rangsit agricultural area in Pathum Thani province collected once a month from June 2004 to May 2005.



(a) Lanchester's freshwater prawn



(b) Apple snail



(c) Freshwater snail

Figure 4.10 Compositions of Σ Endosulfans concentrations including endosulfan sulfate, endosulfan I, and endosulfan II in (a) Lanchester's freshwater prawn, (b) apple snail, and (c) freshwater snail samples between wet and dry season among upper stream, middle stream, and lower stream at Khlong 7, Rangsit agricultural area in Pathum Thani province collected once a month from June 2004 to May 2005.

4.4 Accumulation of OCPRs

The average concentrations of OCPRs in the five group samples were summarized in Table 4.10. In this study, the magnitude concentrations ratios of sediment and water samples were determined to explain the increase of OCPRs concentrations from water to sediment and from both water and sediment to aquatic invertebrates (Table 4.11). In Khlong 7 sub-canal, all OCPRs concentrations detected in sediment samples were greatly higher than those in water samples, ranging from 38 orders of magnitude for methoxychlor to 2,163 orders of magnitude for Σ Heptachlor. The results reflected the influence of hydrophobic property of organochlorine pesticides in the water compartment, so the concentrations of OCPRs were detected much more in sediment samples than in water samples. A majority source of OCPRs in the water system was probably by the runoff water from a number of agricultural farms surrounding along the Khlong 7 sub-canal. The increase of OCPRs from water to sediment was also found in the recent study of Awofolu and Fatoki (2003) in the Eastern Cape, South Africa. They suggested that the source of OCPRs was from the runoff from agricultural areas through water systems. Due to the persistence and slow degradation, these OCPRs were sorbed onto solids, and some dissolved in the surface water.

The results in this study showed that all kinds of OCPRs concentrations in the three aquatic invertebrate species such as Lanchester's freshwater prawn, apple snail, and freshwater snail were higher than in sediment and much higher than in water (Tamhane's T2, $P < 0.05$). Therefore, the results indicated that bioaccumulation was greater in the aquatic organisms. Due to the high lipid composition in aquatic organism bodies, the nonpolar OCPRs were highly detected in aquatic organism tissues more than both in water and sediment. Besides, agricultural activity might be the major source of OCPRs and these organochlorine pesticides entered the aquatic environment through atmospheric deposition, surface runoff or leaching and frequently accumulate in sediments, including aquatic organisms (Miles and Pfeuffer, 1997; Kreuger *et al.*, 1999).

In this study, Σ DDTs were the major contaminants in aquatic invertebrate samples with the average concentrations of 79.61 $\mu\text{g}/\text{kg}$ wet weight in freshwater snail samples, 53.04 $\mu\text{g}/\text{kg}$ wet weight in Lanchester's freshwater prawn samples, and 47.83 $\mu\text{g}/\text{kg}$ wet weight in apple snail samples. The magnitude concentrations ratios of Σ DDTs from both water and sediment to the three kinds of aquatic invertebrates were shown in Figure 4.11. The concentrations of Σ DDTs were more than 2,000 orders of magnitude from water to Lanchester's freshwater prawn and apple snail, and up to more than 4,000 orders of magnitude from water to freshwater snail samples. In contrast, the increase of Σ DDTs concentrations from sediment samples were from 4, 4.4, and 6.6 orders of magnitude to apple snail, Lanchester's freshwater prawn, and freshwater snail, respectively (Figure 4.12).

OCPRs are incorporated in the Khlong 7 ecosystem. These compounds have persisted in the environment and accumulated in the fatty tissues of aquatic animals because of their lipophilic properties and the difficulty breaking down in the animal body (Allsopp and Johnston, 2000). Even though the OCPRs concentrations were low in environments such as water and sediment, they increased to high levels in the body tissues of aquatic animals. As mentioned above, the accumulation of OCPRs from water and sediment could be transferred through aquatic animals of Khlong 7 ecosystem. Figure 4.13 and 4.14 showed the accumulated order of magnitude of the two dominant OCPRs of Σ BHCs and Σ Endosulfate, respectively, from water and sediment through aquatic invertebrates at Khlong 7, Rangsit agricultural area in Pathum Thani province. Bioaccumulation depends not only on the feeding behavior of animal species, but also on a number of different factors such as ages, sexes, and stages in the annual breeding cycle (Pérez-Ruzafa *et al.*, 2000). Lipid content of aquatic organisms is an important factor determining the bioaccumulation of organic contaminants (Landrum and Fisher, 1998). Two ways of pesticide exposure to benthic organisms are, firstly, the absorption from pore water and overlaying water which penetrate through body walls and respiratory surfaces and, secondly, the ingestion of contaminated sediment particles (Power and Chapman, 1992). Also, Swartz and Lee (1980) suggested that the major uptake routes of pollutants in aquatic organisms are through the epidermis, gill epithelium, and gastrointestinal tract and the minor route is through ingestion. Therefore, the bioaccumulations of lipophilic OCPRs in aquatic invertebrates were taken up by passive diffusion as above explanations.

Accumulations of OCPRs were mostly found in freshwater snail more than in apple snail and Lanchester's freshwater prawn. The higher OCPRs accumulation in freshwater snail might be explained by its feeding behavior as a scavenger and its inhabitation on the floor at the bottom of the canal as a benthic fauna. As a result, freshwater snail tends to expose to pesticides, by both absorption and ingestion, higher than apple snail and Lanchester's freshwater prawn which prefer to live mainly at the littoral zone.

This result of accumulation was related to the study of Bouchot *et al.* (1995) who reported that OCPRs in the Palizada river, Mexico were usually found in biological organisms such as shrimp, oysters, and mussels higher than in the sediments. Along the food web in the Mar Menor lagoon in the southeast of Spain, the highest OCPRs concentrations were also in a green algae *Chaetomorpha linum* and an isopod *Idotea basteri*, following by sediments and water, respectively (Pérez-Ruzafa *et al.*, 2000). Also, the other similar results were reported in several areas, for example, in the Göksu delta in Turkey (Ayas *et al.*, 1997), in Paranoá lake of Brasilia, Brazil (Caldas *et al.*, 1999), in Los Padres pond watershed, Argentina (Miglioranza *et al.*, 1999) in the Hanoi region (Nhan *et al.*, 2001), and in the coastal area of Dar es Salaam city, Tanzania (Mwevura, 2002).

Table 4.10 Comparison of overall means of OCPRs concentrations ^a in water, sediment, Lanchester's freshwater prawn, apple snail, and freshwater snail samples at Khlong 7, Rangsit agricultural area, Pathum Thani province collected once a month from June 2004 to May 2005.

Organochlorine Pesticides Residues (OCPRs)	Concentration of OCPRs in each sample (mean ± SE)				
	Water (n = 108) (µg/L)	Sediment (n = 108) (µg/kg dry weight)	Lanchester's freshwater prawn (n = 93) (µg/kg wet weight)	Apple snail (n = 72) (µg/kg wet weight)	Freshwater snail (n = 57) (µg/kg wet weight)
α-BHC	0.00173 ± 0.00021 ^a	0.26 ± 0.07 ^b	2.77 ± 0.52 ^c	10.02 ± 1.09 ^d	9.24 ± 0.71 ^d
γ-BHC	0.00509 ± 0.00029 ^a	5.13 ± 0.08 ^b	2.40 ± 0.34 ^c	0.73 ± 0.29 ^a	1.52 ± 0.56 ^{ac}
β-BHC	0.00696 ± 0.00178 ^a	3.32 ± 0.19 ^b	12.31 ± 1.13 ^c	18.29 ± 1.38 ^{cd}	25.57 ± 2.89 ^d
δ-BHC	0.00054 ± 0.00022 ^a	0.65 ± 0.09 ^b	9.60 ± 1.86 ^c	5.30 ± 1.35 ^{bc}	5.94 ± 1.24 ^c
∑BHCs	0.01432 ± 0.00190 ^a	9.35 ± 0.27 ^b	27.08 ± 2.13 ^c	34.35 ± 2.64 ^{cd}	42.27 ± 4.59 ^d
Heptachlor	0.00626 ± 0.00088 ^a	13.38 ± 0.42 ^b	12.44 ± 0.76 ^b	15.41 ± 1.34 ^b	14.12 ± 1.24 ^b
Heptachlor epoxide	0.00052 ± 0.00011 ^a	1.29 ± 0.09 ^b	2.09 ± 0.37 ^{bc}	3.61 ± 0.54 ^{cd}	4.80 ± 1.78 ^d
∑Heptachlor	0.00678 ± 0.00090 ^a	14.67 ± 0.48 ^b	14.52 ± 0.85 ^b	19.02 ± 1.54 ^b	18.92 ± 2.30 ^b
Aldrin	0.00275 ± 0.00026 ^a	0.81 ± 0.06 ^b	3.52 ± 0.28 ^c	6.73 ± 0.63 ^d	6.93 ± 0.53 ^d
Dieldrin	0.00449 ± 0.00080 ^a	2.15 ± 0.18 ^b	2.36 ± 0.38 ^b	8.97 ± 1.41 ^c	11.40 ± 1.24 ^c
∑Aldrin	0.00720 ± 0.00096 ^a	2.96 ± 0.23 ^a	5.88 ± 0.46 ^a	15.70 ± 1.73 ^b	18.33 ± 1.58 ^b
4,4'-DDE	0.00044 ± 0.00011 ^a	3.03 ± 0.08 ^b	5.04 ± 0.41 ^c	7.84 ± 1.55 ^c	15.93 ± 1.14 ^d
4,4'-DDD	0.00078 ± 0.00017 ^a	1.66 ± 0.05 ^b	2.18 ± 0.55 ^{bc}	9.40 ± 2.04 ^d	8.44 ± 2.25 ^{cd}
4,4'-DDT	0.01803 ± 0.00105 ^a	7.36 ± 0.24 ^b	45.82 ± 7.73 ^{cd}	30.59 ± 2.62 ^c	55.24 ± 6.56 ^d
∑DDTs	0.01926 ± 0.00110 ^a	12.05 ± 0.30 ^b	53.04 ± 7.85 ^c	47.83 ± 5.07 ^c	79.61 ± 8.82 ^c
Endosulfan I	0.00524 ± 0.00083 ^a	0.87 ± 0.03 ^b	3.34 ± 0.32 ^c	8.74 ± 1.79 ^d	8.10 ± 0.98 ^d
Endosulfan II	0.00726 ± 0.00156 ^a	2.30 ± 0.13 ^b	5.41 ± 0.73 ^{cd}	10.08 ± 1.73 ^d	2.62 ± 1.24 ^{bc}
Endosulfan sulfate	0.00726 ± 0.00156 ^a	3.20 ± 0.18 ^b	27.94 ± 5.63 ^c	17.69 ± 1.73 ^c	17.15 ± 2.80 ^c
∑Endosulfans	0.08252 ± 0.01281 ^a	6.36 ± 0.25 ^b	36.69 ± 5.71 ^c	36.51 ± 3.97 ^c	27.87 ± 3.44 ^c
Endrin	0.00379 ± 0.00052 ^a	0.55 ± 0.10 ^b	3.17 ± 0.72 ^c	9.50 ± 2.52 ^{cd}	8.65 ± 1.57 ^d
Endrin aldehyde	0.00097 ± 0.00019 ^a	0.23 ± 0.07 ^b	2.65 ± 0.73 ^c	7.26 ± 1.22 ^d	0.68 ± 0.68 ^{abc}
∑Endrin	0.00476 ± 0.00056 ^a	0.78 ± 0.12 ^b	5.82 ± 1.23 ^c	16.76 ± 3.15 ^d	9.34 ± 1.79 ^{cd}
Methoxychlor	0.00407 ± 0.00127 ^a	0.16 ± 0.06 ^a	2.08 ± 0.59 ^b	6.77 ± 1.08 ^c	5.39 ± 1.06 ^{bc}

^a means with the same letter in the same row are not significantly different, $p \leq 0.05$

n: number of sampling

Table 4.11 Magnitude concentrations ratio of sediment, water, and aquatic invertebrates (Lanchester's freshwater prawn, apple snail, and freshwater snail samples) at Khlong 7, Rangsit agricultural area, Pathum Thani province collected from June 2004 to May 2005.

OCPRs	The magnitude concentrations ratio between sediment and water (C _S /C _W)	The magnitude concentrations ratio between aquatic invertebrates and water			The magnitude concentrations ratio between aquatic invertebrates and sediment		
		C _L /C _W	C _A /C _W	C _F /C _W	C _L /C _S	C _A /C _S	C _F /C _S
∑BHCs	653	1,891	2,399	2,952	2.9	3.7	4.5
∑Heptachlor	2,163	2,142	2,805	2,790	1.0	1.3	1.3
∑Aldrin	412	817	2,181	2,546	2.0	5.3	6.2
∑DDTs	626	2,754	2,483	4,133	4.4	4.0	6.6
∑Endosulfans	77	445	442	338	5.8	5.7	4.4
∑Endrin	163	1,223	3521	1,961	7.5	21.6	12.0
Methoxychlor	38	511	1,663	1,324	13.3	43.4	34.6

C_S = concentrations in sediment samples, C_W = concentrations in water samples, C_L = concentrations in Lanchester's freshwater prawn, C_A = concentrations in apple snail samples and C_F = concentrations in freshwater snail samples

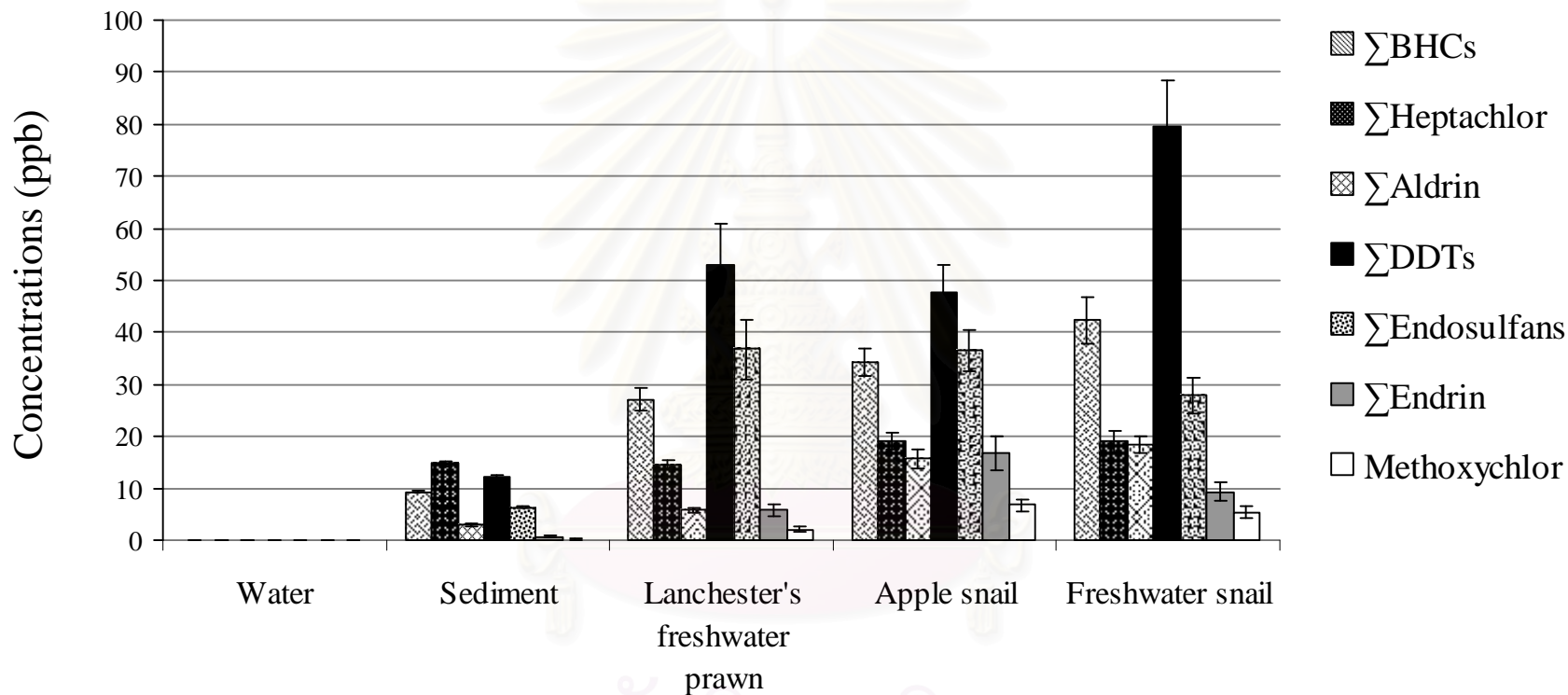


Figure 4.11 Comparisons of overall means of the OCPs concentrations (\pm SE) in water, sediment, Lanchester's freshwater prawn, apple snail, and freshwater snail samples collected once a month from June 2004 to May 2005 at Khlong 7, Rangsit agricultural area in Pathum Thani province.

Figure 4.12 Accumulated orders of magnitude of Σ DDTs from water and sediment through aquatic invertebrates at Khlong 7, Rangsit agricultural area in Pathum Thani province collected from June 2004 to May 2005.

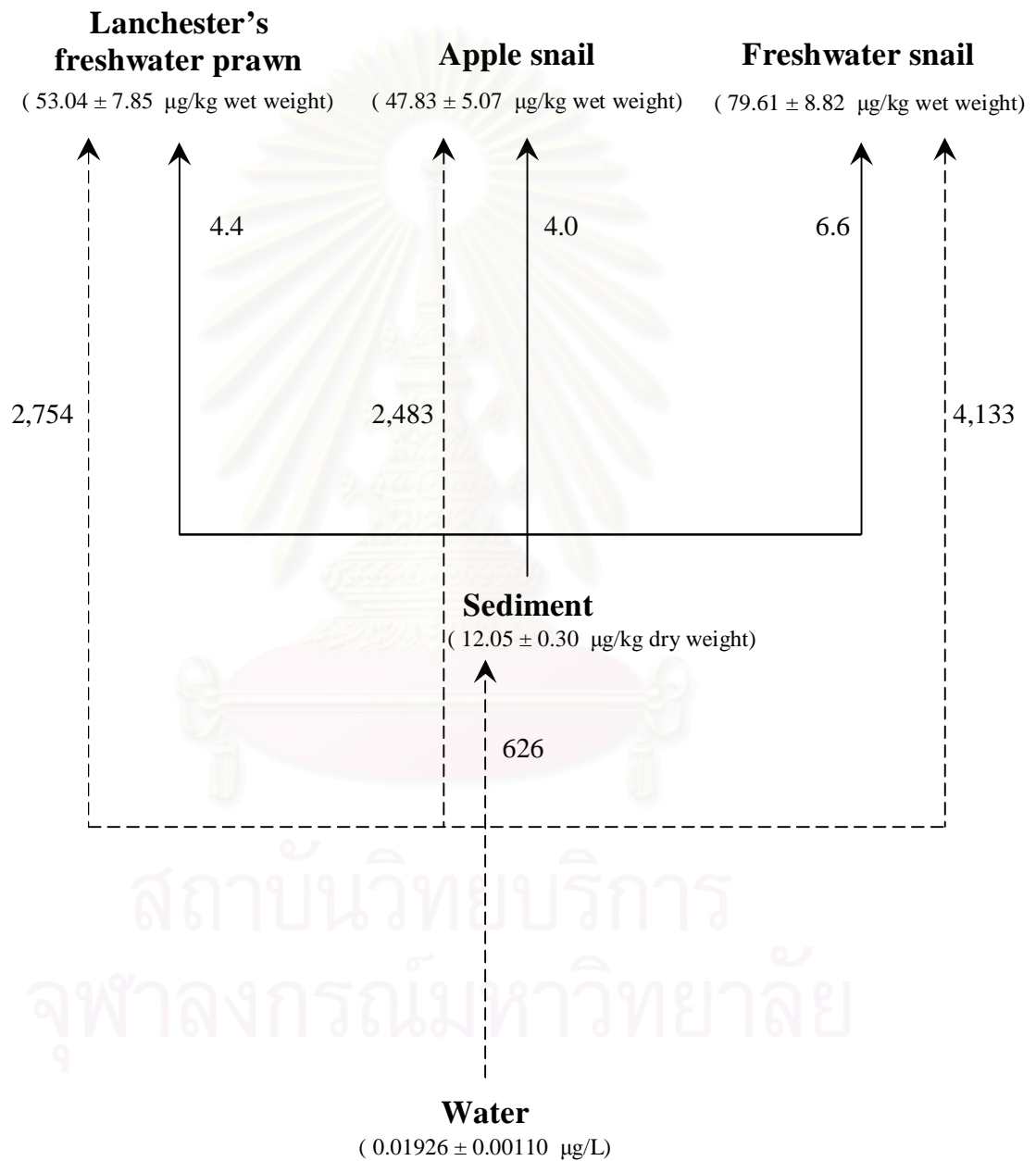


Figure 4.13 Accumulated orders of magnitude of Σ BHCs from water and sediment through aquatic invertebrates at Khlong 7, Rangsit agricultural area in Pathum Thani province collected from June 2004 to May 2005.

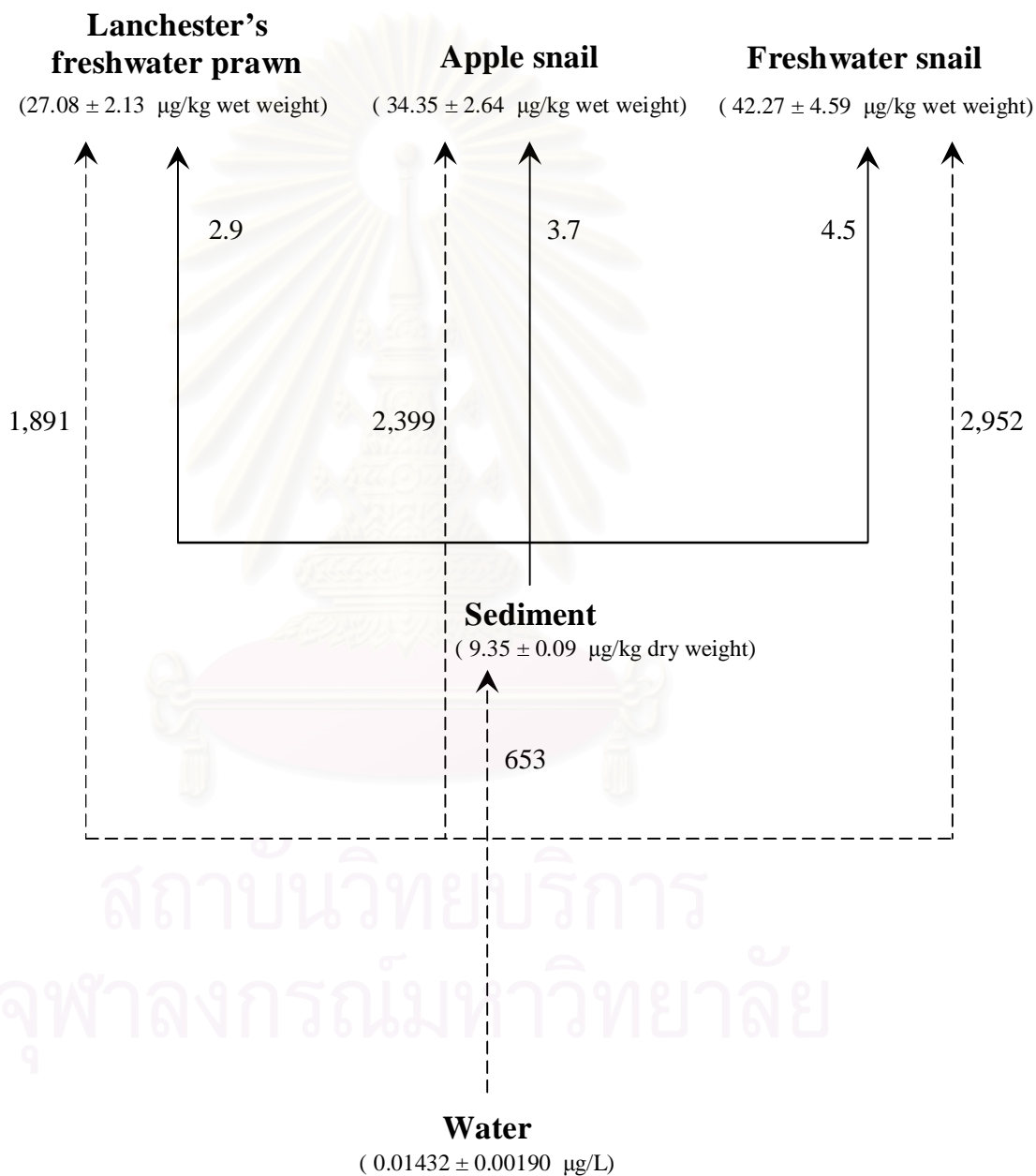
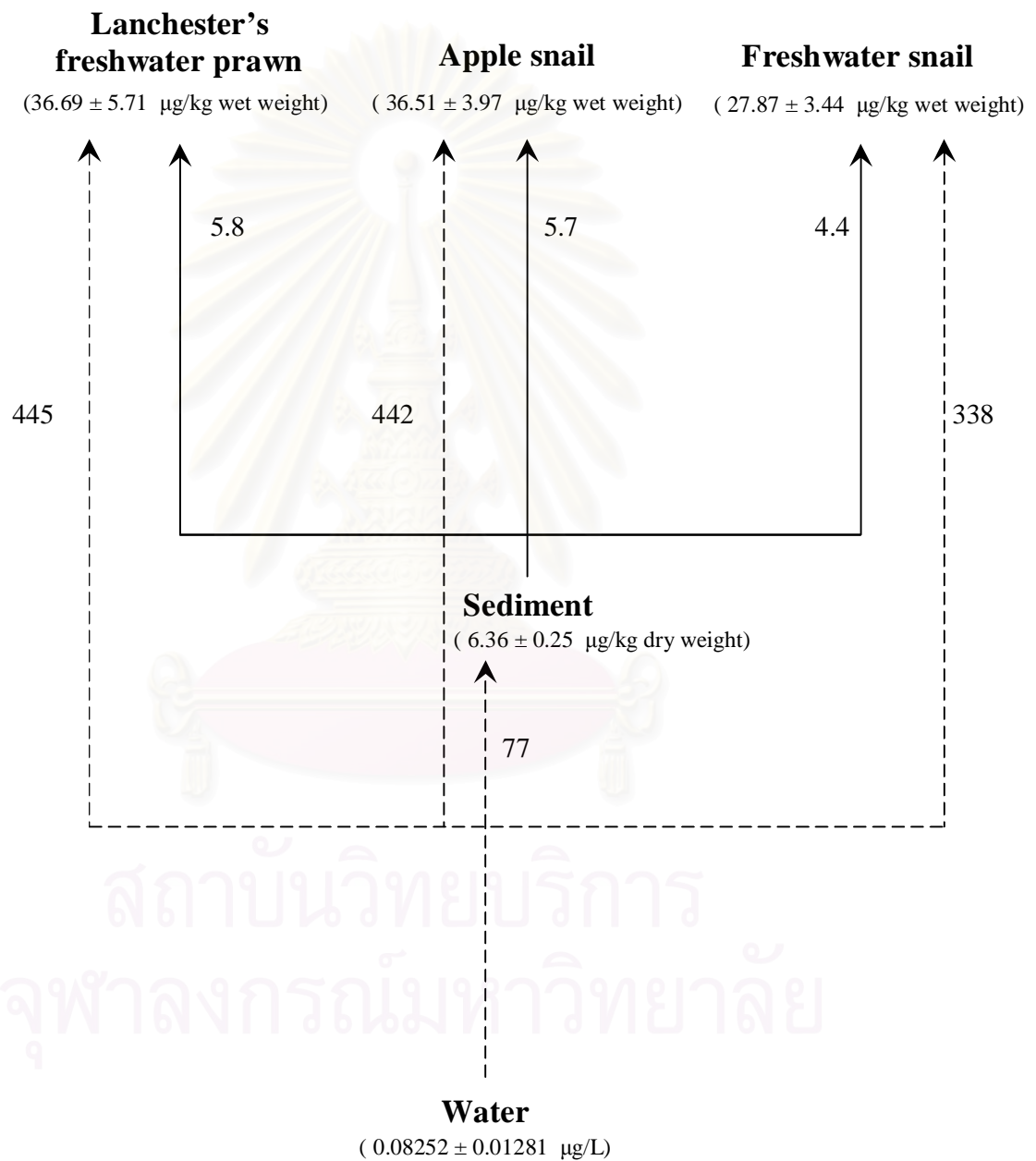


Figure 4.14 Accumulated orders of magnitude of of Σ Endosulfans from water and sediment through aquatic invertebrates at Khlong 7, Rangsit agricultural area in Pathum Thani province collected from June 2004 to May 2005.



4.5 Risk evaluation

Generally, the organochlorine pesticides are hydrophobic substances with low water solubility. Most of them have octanol-water partition coefficients (K_{ow}) with log K_{ow} between 3.5 and 6 and, thus, are very soluble in lipids. As a consequence, these pesticides are highly concentrated by living organisms and concentrations can biomagnify along the food chain (Noble, 1993; Spacie *et al.*, 1995; Carvalho *et al.*, 1999). Their toxicity is mainly the effect on the nervous system. They are relatively unreactive stable compounds and are characterized by their long-lasting residual effects.

Invertebrate species such as Lanchester's freshwater prawn *Macrobrachium lanchesteri*, apple snail *Pomacea* sp., and freshwater snail *Filopaludina martensi* that reside in the Khlong 7 sub-canal are components in this ecosystem, serving as primary consumers as well as preys for some fish species. According to this study, bioaccumulation was happened in aquatic invertebrates. Therefore, the amount of OCPRs may have adverse effect on human health by direct consumption of these invertebrates and by indirect consumption of numerous fish species.

In this present study, the average OCPRs concentrations in aquatic biota were compared with the maximum residue limits (MRLs) which is the maximum concentration of a pesticide residue expressed as mg/kg, recommended by the Codex Alimentarius Commission and the Ministry of Public Health in Thailand. From Table 4.12, the levels of organochlorine pesticide residues in Lanchester's freshwater prawn, apple snail, and freshwater snail samples did not exceed the MRLs for aquatic animals as recommended by both the Codex Alimentarius Commission and Ministry of Public Health in Thailand. Consequently, the concentrations in aquatic invertebrates at Khlong 7, Rangsit agricultural area in Pathum Thani province, particularly Σ DDTs, Σ Endosulfans, Σ Heptachlor, Σ BHCs, Σ Endrin, dieldrin, aldrin, and methoxychlor were below the levels that were suggested to cause adverse effects in human and wildlife in this area. The average concentrations of OCPRs in this study were compared to a study of Siriwong (1991) who reported the levels of OCPRs in green mussel *Perna viridis* samples from the Gulf of Thailand. Similarly, it may be

concluded that the levels of OCPRs in this study did not exceed the MRLs as recommended by the Ministry of Public Health.

However, the issue that should be concerned is the accumulation of pesticides from aquatic invertebrates to human by food consumption because aquatic invertebrates are the primary consumers in the food chain. The interesting point is that these OCPRs may produce a chronic effect to the farmers including the local people who expose to OCPRs for a long period.

Table 4.12 OCPRs concentrations in each aquatic invertebrate samples (mg/kg) compared to the maximum residue limits (MRLs in mg/kg) by the Codex Alimentarius Commission and the Ministry of Public Health in Thailand.

OCPRs	MRLs ^a (mg/kg)	MRLs ^b (mg/kg)	OCPRs concentrations in each aquatic invertebrates (mg/kg)		
			Lanchester's freshwater prawn	Apple snail	Freshwater snail
∑BHCs	-	0.5	0.03	0.03	0.04
∑Heptachlor	0.3	0.3	0.01	0.02	0.02
Aldrin	0.3	0.3	0.004	0.007	0.007
Dieldrin	0.3	0.3	0.002	0.009	0.01
∑DDTs	5.0	5.0	0.05	0.05	0.08
∑Endosulfans	0.3	0.3	0.04	0.04	0.03
∑Endrin	-	0.3	0.01	0.02	0.01
Methoxychlor	5.0	5.0	0.002	0.01	0.01

Source: ^a MRLs from the Notification of Codex Alimentarius Commission

^b MRLs from the Ministry of Public Health No.71 (B.E.2525) issued under Food Act B.E.2522 (1979), published in the Royal Government Gazette (Special issued) Vol. 169, Part 168, dated November 1982.

CHAPTER V

CONCLUSIONS AND SUGGESTIONS FOR FUTURE WORKS

Rangsit agricultural area has 14 irrigation canals which support various agricultural lands especially paddy fields. This area has been contaminated by organochlorine pesticide residues (OCPRs) which are lipophilic with slow chemical and biological degradation. Khlong 7 sub-canal was selected as a study area, since the canal is located at the middle of Rangsit irrigation system and surrounded by large area of paddy fields. The rice growing system in this area required pump-in water from the canal to paddy fields at the beginning of cultivation and water was released to the canal before harvesting. Therefore, organochlorine pesticides used during the cultivations contaminated the canal and its components via the released water. Moreover, each paddy field can be cultivated three times a year with continuous use of pesticides. Although most organochlorine pesticides have been banned for several years, OCPRs are still detected in water, sediment, and some aquatic invertebrate samples such as Lanchester's freshwater prawn, apple snail, and freshwater snail.

5.1 OCPRs in water, sediment, and aquatic invertebrates the samples

The OCPRs in water, sediment, and three aquatic invertebrate samples such as Lanchester's freshwater prawn, apple snail, and freshwater snail at Khlong 7, Rangsit agricultural area in Pathumthani Province collected from June 2004 to May 2005 were investigated. The predominant concentrations of OCPRs in water samples were Σ Endosulfans, following by Σ DDTs, Σ BHCs, Σ Aldrin, Σ Heptachlor, Σ Endrin, and methoxychlor, respectively. The comparisons of OCPRs between wet and dry seasons revealed that Σ Endosulfans were significantly higher in wet season than in dry season. For sediment samples, the highest OCPRs were Σ Heptachlor, following by Σ DDTs, Σ BHCs, Σ Endosulfans, Σ Aldrin, Σ Endrin, and methoxychlor. The average concentrations of Σ DDTs were predominant in Lanchester's freshwater prawn, apple snail, and freshwater snail. Due to the lipophilic, persistent, and slowly degradable properties of organochlorine pesticides, bioaccumulations of OCPRs were observed in

aquatic invertebrate samples. The main routes of OCPRs accumulation in biological samples may be due to absorption and ingestion from environments such as water and sediment through their tissues (Swartz and Lee, 1980; Power and Chapman, 1992).

In this study, the average OCPRs concentrations in aquatic invertebrates were compared with the maximum residue limits (MRLs) which recommended by the Codex Alimentarius Commission and the Ministry of Public Health in Thailand. The levels of organochlorine pesticide residues in Lanchester's freshwater prawn, apple snail, and freshwater snail samples did not exceed the MRLs for aquatic animals.

5.2 Suggestion for management

Even though the levels of OCPRs in aquatic animals did not exceed the MRLs, the issue that should be concerned is that some OCPs have been illegally used in this area. The use of banned pesticides should be more restricted. The local administration sectors should give the people awareness and education of the correct pesticide practices. Group communication between local people and the popular wisdom such as biological control, using duck to get rid of apple snail instead of chemical pesticides especially endosulfan, should be performed. In the case of by-product from industry such as heptachlor epoxide, and some persistent metabolite of pesticides such DDT and derivatives, BHC and derivatives, etc., the government should be routine monitoring.

5.3 Suggestion for future works

To complete the food chain and food web of Klong 7 aquatic community, the concentration of OCPRs in aquatic plants, other aquatic invertebrates, and aquatic vertebrates should be investigated. Furthermore, OCPRs in soil and rice surrounding the canal should be investigated. Also, the biomagnification through the complex food web would be demonstrated. Moreover, to check risk of consumers, risk assessment should be done.

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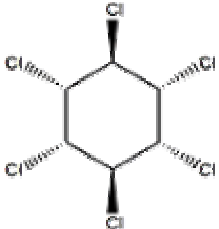
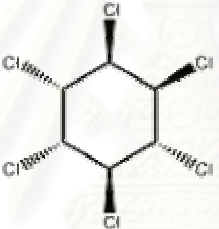
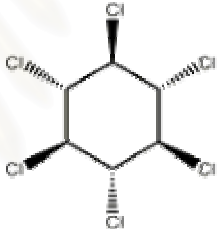
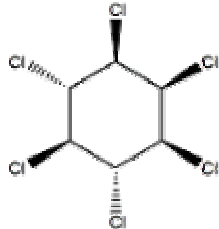
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Appendix

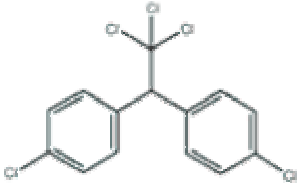
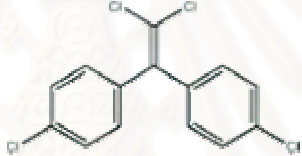
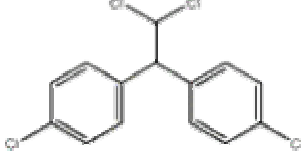
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Table A-1 Physical and chemical properties of benzenehexachloride (BHC) isomers

Characteristic	γ -BHC	α -BHC	β -BHC	δ -BHC
Synonym(s)	Gamma-benzenehexachloride; gamma-hexachlorocyclohexane; lindane	Alpha-benzenehexachloride; alpha-1,2,3,4,5,6-hexachlorocyclohexane; alpha-hexachlorane	Beta-benzenehexachloride; beta-hexachlorobenzene; beta-1,2,3, 4,5,6-hexachlorocyclohexane	Delta-benzenehexachloride; delta-1,2,3,4,5,6-hexachlorocyclohexane; delta-lindane
Chemical formula	$C_6H_6Cl_6$	$C_6H_6Cl_6$	$C_6H_6Cl_6$	$C_6H_6Cl_6$
Chemical structure				
Melting point	112.5 °C	159-160 °C	314-315 °C	141-142 °C
Boiling point	323.4 °C at 760 mmHg	288 °C at 760 mmHg	60 °C at 0.5 mmHg	60 °C at 0.36 mmHg
Water solubility	17 ppm; insoluble in water	10 ppm; 69.5 ppm at 28°C	5 ppm	10 ppm
Partition coefficients:				
Log K_{ow}	3.72	3.8	3.78	4.14
Log K_{oc}	3.0, 3.57	3.57	3.57	3.8
Vapor pressure	4.2×10^{-5} mmHg at 20 °C	4.5×10^{-5} mmHg at 25 °C	3.6×10^{-7} mmHg at 20 °C	3.5×10^{-5} mmHg at 25 °C

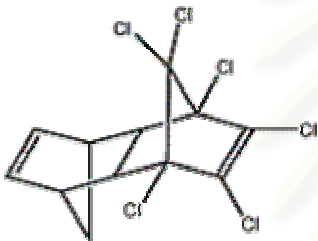
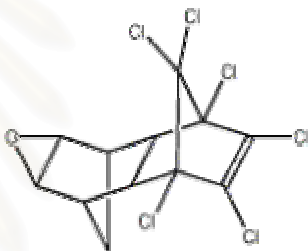
Source: ATSDR, 2005

Table A-2 Physical and chemical properties of DDT and derivatives

Characteristic	4,4'-DDT	4,4'-DDE	4,4'-DDD
Synonym(s)	1,1,1-trichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; dichlorodiphenyltrichloro ethane; DDT; 1,1'-(2,2,2-trichloroethylidene)bis(4-chloro-benzene)	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethylene; dichlorodiphenyldichloro ethane; 1,1'-(2,2-dichloroethylidene)bis(4-chlorobenzene)	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane; 1,1-bis(4-chlorophenyl)-2,2-dichloroethane; TDE; tetrachlorodiphenylethane
Chemical formula	C ₁₄ H ₉ Cl ₅	C ₁₄ H ₈ Cl ₄	C ₁₄ H ₁₀ Cl ₄
Chemical structure			
Melting point	109 °C	89 °C	109-110 °C
Boiling point	Decomposes	336 °C	350 °C
Water solubility	0.025 mg/L at 25 °C	0.12 mg/L at 25°C	0.090 mg/L at 25 °C
Partition coefficients:			
Log K _{ow}	6.91	6.51	6.02
Log K _{oc}	5.18	4.70	5.18
Vapor pressure	1.60×10 ⁻⁷ mmHg at 20 °C	6.0×10 ⁻⁶ mmHg at 25 °C	1.35×10 ⁻⁶ mmHg at 25 °C

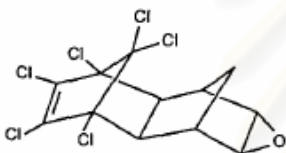
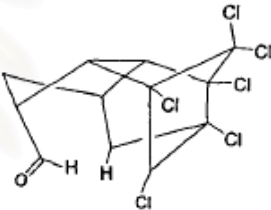
Source: ATSDR, 2002

Table A-3 Physical and chemical properties of aldrin and dieldrin

Characteristic	Aldrin	Dieldrin
Synonym(s)	1,2,3,4,10,10-hexachloro-1,4,4 α 5,8,8 α -hexahydro-exo-1,4-endo-5,8-dimethano-naphthalene	1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4 α ,5,6,7,8,8 α -octa-hydro-1,4-endo,exo-5,8-dimethanonaphthalene
Chemical formula	C ₁₂ H ₈ Cl ₆	C ₁₂ H ₈ Cl ₆ O
Chemical structure		
Melting point	104-105.5 °C 49-60 °C (technical grade)	176-177 °C 95 °C (technical grade)
Boiling point	Decomposes	Decomposes
Water solubility	0.011 mg/L at 20 °C	0.110 mg/L at 20°C
Partition coefficients:		
Log K _{ow}	6.50	6.2
Log K _{oc}	7.67	6.67
Vapor pressure	7.5×10 ⁻⁵ mmHg at 20 °C, 1.2×10 ⁻⁴ mmHg at 25 °C	3.1×10 ⁻⁶ mmHg at 20 °C, 5.89×10 ⁻⁶ mmHg at 25 °C

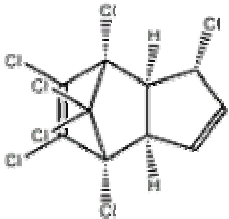
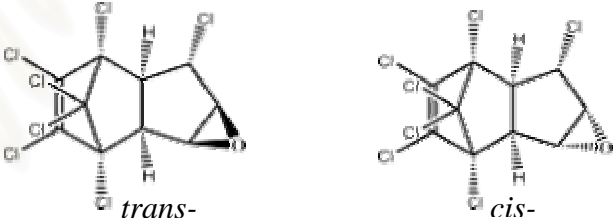
Source: ATSDR, 2002

Table A-4 Physical and chemical properties of endrin and endrin aldehyde

Characteristic	Endrin	Endrin aldehyde
Synonym(s)	1,2,3,4,10,10-hexachloro-6,7-epoxy- 1,4,4A,5,6,7,8,8A-octahydro-endo,endo-1,4:5,8- dimethanonaphthalene	1,2,4-methanecyclopenta(c,d)pentalene-5- carboxaldehyde,2,2a,3,3,4,7-hexachlorodecahydro
Chemical formula	C ₁₂ H ₈ Cl ₆ O	C ₁₂ H ₈ Cl ₆ O
Chemical structure		
Melting point	235 °C, 226-230 °C 49-60 °C (technical grade)	176-177 °C 95 °C (technical grade)
Boiling point	Decomposes at 245 °C Decomposes above 200 °C	No data
Water solubility	200 µg/L at 25 °C	50 µg/L, 0.25-0.26 ppm at 25°C
Partition coefficients:		
Log K _{ow}	5.6, 5.34, 5.45 (calculated)	3.146, 4.7, 5.6 (calculated)
Log K _{oc}	4.532 (calculated), 5.195 (±0.005)	4.80, 3.929-4.653 (calculated)
Vapor pressure	2.0×10 ⁻⁷ mmHg at 25 °C	2.0×10 ⁻⁷ mmHg at 25 °C

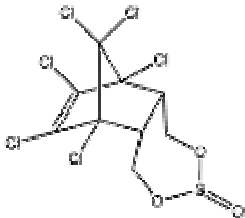
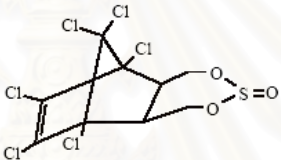
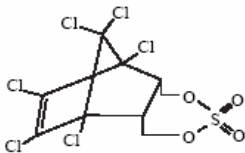
Source: ATSDR, 1996

Table A-5 Physical and chemical properties of heptachlor and heptachlor epoxide

Characteristic	Heptachlor	Heptachlor epoxide
Synonym(s)	3-Chlorochlordene; 1,4,5,6,7,8,8a-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindane; 1,4,5,6,7,8,8-heptachloro-3A,4,5,5a tetrahydro; alpha-dicyclopentadiene, 3,4,5,6,8,8a-heptachloro, and others	Epoxyheptachlor; 1,4,5,6,7,8,8a-hepta-chloro-2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindene; 4,7-methanoindan,1,4,5,6,7,7-heptachloro-1a,1b,5,5a,6,6a-hexahydro
Chemical formula	$C_{12}H_8Cl_6O$	$C_{12}H_8Cl_6O$
Chemical structure		
Melting point	95-96 °C (pure); 46-74 °C (technical grade)	160-161.5 °C
Boiling point	145 °C	No data
Water solubility	0.05 mg/L at 25 °C	0.275 mg/L at 25°C
Partition coefficients:		
Log K_{ow}	6.10	5.40
Log K_{oc}	4.34	3.34-4.37
Vapor pressure	3×10^{-4} mmHg at 20 °C and 25 °C	2.6×10^{-6} mmHg at 25 °C

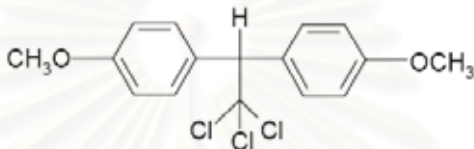
Source: ATSDR, 2005

Table A-6 Physical and chemical properties of endosulfan and derivative

Characteristic	Endosulfan I (α -endosulfan)	Endosulfan II (β -endosulfan)	Endosulfan sulfate
Synonym(s)	6,9-methano-2,4,3- benzodioxathiepin-6,7,8,9,10,10-hexachloro-1,5,5a,6,9, 9a-hexahydro-3-oxide	6,7,9,10,10-hexachloro-1,5,5a,6,9, 9a-hexahydro-6,9-methano-2,4,3- benzodioxathiepin-3-oxide	6,7,8,9,10,10-hexachloro-1,5,5a,6,9, 9a-hexahydro-6,9-methano-2,4,3- benzodioxodioxathiepin-3,3-dioxide
Chemical formula	$C_9H_8Cl_6O_3S$	$C_9H_8Cl_6O_3S$	$C_9H_6Cl_6O_4S$
Chemical structure			
Melting point	108-110 °C	207-212 °C	181 °C, 198-201 °C
Boiling point	No data	No data	No data
Water solubility	0.53 mg/L at 25 °C	0.28 mg/L at 25°C	0.22 mg/L at 25 °C
Partition coefficients:			
Log K_{ow}	3.83	3.52	3.66
Log K_{oc}	3.55	No data	No data
Vapor pressure	1×10^{-5} mmHg at 25 °C	1×10^{-5} mmHg at 25 °C	1×10^{-5} mmHg at 25 °C

Source: ATSDR, 2000

Table A-7 Physical and chemical properties of methoxychlor

Characteristic	Methoxychlor
Synonym(s)	2,2-bis(p-methoxyphenyl)-1,1,1-trichloroethane; 1,1,1-trichloro-2,2-bis(4-methoxyphenyl)ethane; methoxy-DDT; 1,1-(2,2,2-trichloroethylidene)-bis(4-methoxybenzene)
Chemical formula	C ₁₆ H ₁₅ Cl ₃ O ₂
Chemical structure	
Melting point	89 °C, 77 °C (technical grade)
Boiling point	No data (decomposes)
Water solubility	0.045 mg/L at 25 °C 0.02 mg/L at 15 °C 0.04 mg/L at 24 °C 0.095 mg/L at 35 °C 0.185 mg/L at 45 °C
Partition coefficients:	
Log K _{ow}	4.68-5.08
Log K _{oc}	4.9
Vapor pressure	1.4×10 ⁻⁶ mmHg at 25 °C

Source: ATSDR, 2002

Table A-8 Organochlorine pesticides status in Thailand

Common name	Chemical name	Molecular formula	Imported year	Banned year
Aldrin	1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo,exo-5,8-dimethanonaphthalene	C ₁₂ H ₈ Cl ₆	1975	1988
Benzene hexachloride (BHC) or Hexachlorocyclohexane (HCH)	Hexachlorocyclohexane	C ₆ H ₆ Cl ₆	1975	2001
Chlordane	1, 2, 4, 5, 6, 7, 8, 8-octachloro – 2, 3, 3a, 4, 7, 7a – hexahydro – 4, 7 - methanoindene	C ₁₀ H ₆ Cl ₈	1974	2000
DDD	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane	DDT derivative	No record	2001
DDE	1,1-dichloro-2,2-bis(<i>p</i> -chlorophenyl)ethylene	C ₁₄ H ₈ Cl ₄	No record	-
DDT	1,1,1-trichloro-2,2-bis(<i>p</i> -chlorophenyl)ethane	C ₁₄ H ₉ Cl ₅	1978	1983
Dieldrin	1,2,3,4,10,10-hexachloro-1,4,4a,5,6,8,8a-hexahydro-6,7-epoxy-1, 4:5, 8-dimethanonaphthalene	C ₁₂ H ₈ Cl ₆ O	1975	1988
Endosulfan	6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepin-3-oxide	C ₉ H ₆ Cl ₆ O ₃ S	1975	2004
Endrin	(1a,2,2a,3,6,6a,7,7a)-3,4,5,6,9,9-hexachloro-1a,2,2a,3,6,6a,7,7a-octahydro-2,7:3,6-dimethanonaphth[2,3-b]oxirene	C ₁₂ H ₈ Cl ₆ O	1978	1981
Heptachlor	1, 4, 5, 6, 7, 8, 8 - heptachloro - 3a, 4, 7, 7a – tetrahydro - 4, 7 - methanoindene	C ₁₀ H ₅ Cl ₇	1978	1988
Methoxychlor	1,1,1-trichloro-2,2-di-(<i>p</i> -methoxyphenyl) ethane	C ₁₆ H ₁₅ O ₂ Cl ₃	No record	Available

Source: Thirakhupt, 2006

BIOGRAPHY

Premkamol Thongkongwm was born on the 23 April, 1981, in Chanthaburi province. She received a Bachelor of Science degree in Zoology in 2002 from the Faculty of Science, Chulalongkorn University. She continued her study for a Master of Science in Environmental Management, the Graduate School, Chulalongkorn University in 2003 and completed the program in 2005.



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