

## Accumulation and distribution of some heavy metals in water, soil and rice fields along the Pradu and Phi Lok canals, Samut Songkhram province, Thailand

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### Abstract

This study aims to determine concentrations of heavy metals (Cd, Cu, and Zn) in the water, soil, and rice in paddy fields, and to compare heavy metal concentrations in the water, soil, and four parts of the rice plant (root, shoot, grain, and husk) between organic paddy fields and conventional paddy fields. The study area was the Pradu Canal (sites A and C) and the Phi Lok Canal (site B) in Phraek Nam Daeng Sub-district, Samut Songkhram Province, Thailand. Data was collected from August 2007 to December 2007. The water, soil, and rice samples were prepared for heavy metal analysis using a microwave digestion system and heavy metal concentrations were determined using an atomic absorption spectrophotometer. Physicochemical parameters of the water and soil were also studied.

The results showed the accumulation of heavy metals in the water, paddy soil, and four parts of the rice plant to be as follows (in descending order of frequency): Zn > Cu > Cd. The concentration of heavy metals in samples was also found to be as follows (greatest first): paddy soil > rice root > rice shoot > rice grain > rice husk > water. No significant difference was observed between Cd and Zn in paddy soil before ploughing and after rice harvesting at sites A, B, and C. However, Cu showed a significant difference at the level of 0.05 at site A and no significant difference at sites B and C.

It can be concluded therefore that Cd, Cu, and Zn concentrations in paddy soil and water were lower than the values permitted by the Surface Water Quality Standard and Soil Quality Standard of Thailand. Furthermore, Cd concentrations in rice seeds were lower than the values permitted by international health organizations and thus acceptable for human consumption.

**Key words:** Heavy Metal/ Organic and Conventional Paddy Field/ Rice

### 1. Introduction

Thailand is an agriculture country. The most economics and famous plant in Thailand is rice field. Rice has many important roles in Thai society from food to work. Rice uses over half of the farmable land area and labor force in Thailand. It is one of the main foods and sources of nutrition for most Thai citizens. Rice is also one of the main components of Thai exports. Increasing rice production to support the exports has

created problems of toxic heavy metal contaminations in environment. Heavy metals are impurities in all chemical fertilizers, pesticides, insecticides and fungicides used in rice growing process. These direct inputs of heavy metals through those chemicals into paddy fields can cause the accumulation of heavy metals in rice and when people consume rice, these toxic heavy metals may enter and deposit into fatty tissues. However, sources of heavy metals that enter into the paddy fields are also from all types of

water used during the growing period. The large amount of waste is dumped into rivers, canals and landfills while agriculture farm (such as paddy fields, vegetable farms, animal farms and fruit orchards) and aquaculture (such as shrimp farms and fish ponds) are irrigated using water from natural source. Heavy metals may move from soil and water to food plants through root absorption and accumulate in their tissues. In this way, heavy metals may enter the food chain and affect human health. Although adverse health effects of heavy metals have been known for a long time and exposure to heavy metals continues (Lars, 2003).

Heavy metal can accumulate in the food chain and persist in nature. The accumulation of heavy metal contaminants in the environment has become a concern due to growing health risks to the public. Because of exposure to heavy metal contamination has been found to cause kidney damage, liver damage, carcinogenic and etc. For example, at Mae Sot District, Tak Province, the paddy fields (in the Mae Tao and Mae Ku areas) were found to contain markedly elevated cadmium levels during the surveys in 2001-2004. From the exploration was found that rice

grains grown in the areas were also detected to have elevated cadmium content compared with the normal values. The studied population had much higher levels of urinary cadmium than those living in other districts of Tak Province. Most of them consumed locally produced rice that was found to contain markedly elevated cadmium levels, and therefore might increase daily intake of cadmium from food (Swaddiwudhipong et al., 2007).

The Mae Klong River, in Samut Songkhram Province, is a river that runs through three provinces in west Thailand and flows into the Gulf of Thailand and its total length is 140 km. The upper stream is mainly an agricultural area and the lower is an industrial one. The Mae Klong River supplies water for irrigation and supports water for aquaculture, i.e., shrimp farms and fish ponds and agriculture, i.e., paddy fields and fruit orchards. Land use in the study area is composed of rice fields, aquaculture, livestock, coconut grove and community (as shown in Table 1). These activities required the use of water and released waste water into the environment making the water source contaminated with all toxic substances including heavy metals.

**Table 1** Land use types and area in the year 2003

Land use	Area (rais)
Paddy fields	1,862
Permanent crop	68,655
Field crop	111
Vegetable crop, herb, flower and ornamental plant	996
Forest (planted)	77
Pasture	1
Pen	176
Aquaculture	6,059
Others	595
<b>Total</b>	<b>78,532</b>

Source: Samut Songkhram Provincial Statistical Office (2005)

The land use activities at Amphawa District consist of fish farms, paddy fields

and factories in the freshwater part while shrimp farms are in the saltwater part. A

water gate was constructed as well in order to prevent the intrusion, and this is causing conflict at Amphawa District in Samut Songkram Province, between the prawn farmers that use sea water, paddy field farmers that use freshwater and pig farmers located in the upper stream that discharge polluted water in the waterway. The water gate created the still water condition in the canal and solid suspending upon the water canal tend to settle down to the bottom. The sediment may contaminate with heavy metals from the land use activities. There is no plan for water resource management in the area. Furthermore the increasing of fertilizers, pesticides and chemicals from industries and agricultures has been discharged into the river. For this reason, heavy metals as impurities in the tributaries can widely expose into the waterway and accumulate in the food chain (aquatic and terrestrial ecosystems).

With the great concerns on health, organic farming has becoming more and more popular and practiced all around the world including Thailand. Organic paddy fields are considered very important for Thai citizens as rice is Thai major stable food. However, organic fertilizers and water used in many organic paddy fields are still contaminated with toxic heavy metals. On the other word, the organic paddy fields as understood by most

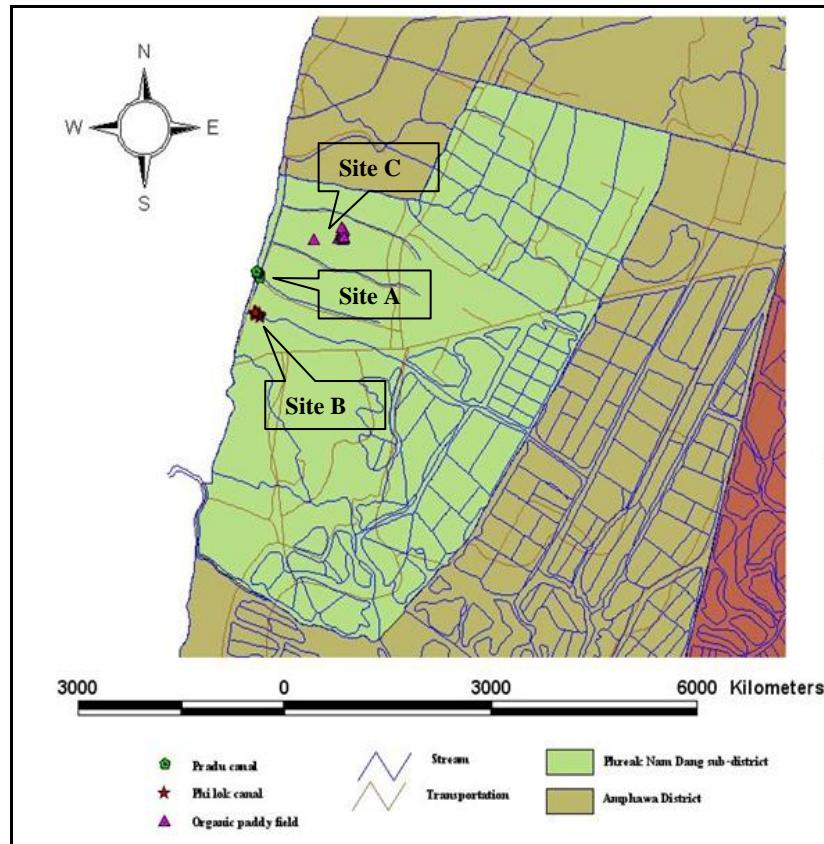
farmers are not organic as practiced somewhere else.

For the present study, conventional paddy fields and organic paddy fields located along the Phi Lok and Pradu Canals that are tributaries of the Mae Klong River, Phreak Nam Daeng Sub-district, Amphawa District in Samut Songkram Province were studied. Heavy metal concentrations, i.e., Cadmium (Cd), Copper (Cu) and Zinc (Zn), from samples of water, soil and rice (*Oryza sativa* L.) were determined.

The goal of the present study was aimed to study the accumulation and distribution of heavy metals in water, soil and rice (Chainat Rice No. 1) in the paddy field of Samut Songkram Province.

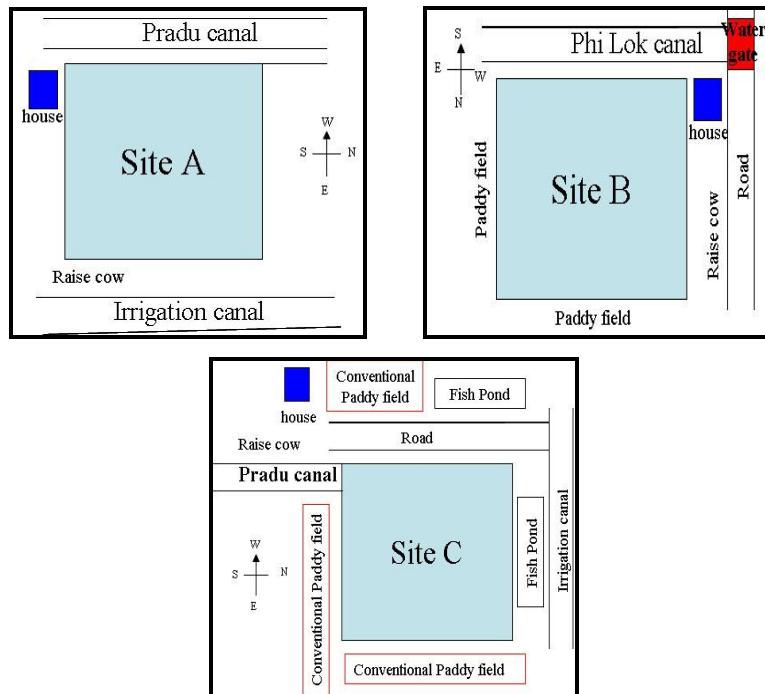
## 2. Methods

The paddy field monitoring practices were located within Preak Nam Daeng sub-district, Amphawa, Samut Songkhram province, Thailand. Irrigation is sourced from Pradu and Phi Lok canals which pass through an agricultural zone (such as pig farms, fish and shrimp ponds), an industrial zone and community zone. A paddy field monitoring was followed from October 2007 to December 2007. The study area and sampling points are shown in Figure 1.



**Figure 1** Map of the study area and sampling points at Samut Songkhram Province

The landuse activity around the paddy fields of Sites A, B and C is shown in Figure 2.



**Figure 2** The activities around the paddy fields of Sites A, B and C

## 2.1 Water, Soil and Rice plants Collected

The whole period of data collection in the field is in the seasonal paddy fields for five months of August to December 2007. Different types of samples were collected and number of sampling times was carried out as follows. Water samples were collected three times in the months of September 2007, October 2007 and November 2007. Soil samples were collected before rice growing in August 2007 and after rice harvesting in December 2007. Rice plant samples were collected about 120 days after sowing in December 2007. Soil, water and rice plant samples are collected from three sampling sites (site A, B and C). The paddy field at sites A and B were conventional agriculture but site C was organic agriculture.

Soil samples were collected at the depth level 0 to 30 centimeters, dried at room temperature, homogenized in an agate mortar, passed through a 2 mm stainless steel sieve and kept ready for physical analysis. Soil samples after passed through a 2 mm (100 g) were dried at 105 °C for 2 h for analysis heavy metal.

The root, shoot, seed and husk of rice plants were harvested separately for the determination of the metal content. The samples were washed three times with deionized water, oven dried at 70 °C and ground to pass a 2 mm stainless steel sieve and kept ready for analysis.

Collecting water samples in each study site by grab sampling. Each time, water samples are collected from pumping water from outside to the paddy field about once a month at the site of water inflow, center of water flowing and water outflow. Water samples are packed in the 1000 ml polyethylene bottle, stored at 4°C. Nitric Acid are added to water sample so that pH < 2 and stored for further analysis in the laboratory for

further analysis to find amount of heavy metal (Sirising, 2006).

## 2.2 Water, Soil and Rice plants Analysis

Materials used for experiments were soaked in 10% HNO<sub>3</sub> (v/v) for 24 hour and rinsed three times with deionized water. All reagents used in this study were analytical grade and prepared stock solutions with deionized water.

A modification method was used for the sample: 0.5 g of soil, 0.5 g of rice plant and 45 ml of water were digested with concentrated HNO<sub>3</sub> by microwave digester. All the digestion samples were analyzed the heavy metal by using atomic absorption spectrophotometer. A Varian Model SpectrAA 220FS involving direct aspiration of the aqueous solution into an air-acetylene flame, equipped with a deuterium background corrector, was used to measure copper and zinc. A Varian Model SpectrAA 220Z graphite furnace was used for absorbance measurements of cadmium.

## 3. Results

Rice growing in the study area depended on rain water and canal water. Rice growing processes in the study areas were similar in all three sites (Sites A, B and C). Sites A and B have been used chemical fertilizers and cow manures but Site C has been used only cow manures and effective microorganism (EM).

### 3.1 Physico-chemical Characteristics of Paddy Field Soils and water

The characteristics of the soils are listed in Table 2. The results could be that soil texture in paddy fields was clay.

**Table 2** Physico-chemical soil properties in paddy fields

Station	Soil particle			Texture
	% Sand	% Silt	% Clay	
Site A: Conventional paddy fields of the Pradu canal	0.56	23.90	75.54	clay
Site B: Conventional paddy fields of the Phi Lok canal	0.20	26.92	72.88	clay
Site C: Conventional paddy fields of the Ta Pet canal	1.49	23.64	74.87	clay

Chemical soil properties	pH	CEC	OM
Site A: Conventional paddy fields of the Pradu canal	6.40-6.92	27.90	1.24-2.17
Site B: Conventional paddy fields of the Phi Lok canal	5.99-6.84	21.80	1.27-1.81
Site C: Conventional paddy fields of the Ta Pet canal	6.68-7.68	25.30	1.49-1.99

The chemical properties in paddy soil are 5.99 to 7.68 for pH and 21.80 to 27.90 meq/100g for cation exchange capacity (CEC). CEC is dependent upon the amount of organic matter and clay in soils and on the types of clay.

The range of pH values varied from 7.09 to 8.33 of paddy field water and 7.05 to 7.68 of canal water at sites A, B and C that within the range of the legal values of the Thailand's surface water quality standards (1994) for class 3 of water quality at 5.00-9.00(PCD, 2004).

### 3.2 Heavy metal concentration in soil, water and rice plants

Heavy metal concentrations in soil were examined at Sites A, B and C twice,

i.e., first in the beginning of rice cultivation in August, 2007 and the second time in December, 2007 which was after harvesting rice. The results of heavy metal concentrations in paddy soil are listed in Table 3.

Heavy metal concentrations in paddy field water at Sites A, B and C and waters of the Pradu and Phi Lok canals near the three sites were analyzed. The results of heavy metal concentrations in water are listed in Table 4.

Heavy metals concentration in rice plants was determined from rice plant samples of Sites A, B and C harvested in December, 2007. The results of heavy metal concentrations in four parts of rice plants are listed in Table 5.

**Table 3** The heavy metals concentrations in paddy soil of three sampling sites (mg/kg)

Element	Time	Site A	Site B	Site C
<b>Cd</b>	Before rice cultivation	0.05±6.34	0.04±9.41	0.06±9.32
	After rice cultivation	0.05±9.17	0.04±7.67	0.05±5.36
<b>Cu</b>	Before rice cultivation	18.86±0.73	19.92±1.83	17.01±0.94
	After rice cultivation	20.20±0.62	20.96±2.47	16.38±0.24
<b>Zn</b>	Before rice cultivation	65.35±3.62	64.74±4.19	57.10±2.51
	After rice cultivation	66.77±3.59	65.77±4.46	55.23±0.84

**Table 4** Heavy Metals in canal water and paddy fields water at Samut Songkhram Province

Times	Site A		Site B		Site C	
	canal	Paddy field	canal	Paddy field	canal	Paddy field
<b>Cadmium (µg/L)</b>						
<b>September</b>	0.11	0.18±0.07	0.15	0.15±0.08	0.13	0.15±0.05
<b>October</b>	0.24	0.17±0.07	0.16	0.23±0.14	0.10	0.19±0.11
<b>November</b>	1.31	0.90±0.43	0.53	0.48±0.43	1.18	0.83±0.81
<b>Copper (µg/L)</b>						
<b>September</b>	8.42	8.42±2.49	7.37	7.37±1.69	4.92	5.75±2.00
<b>October</b>	7.36	6.24±0.75	8.52	6.85±2.20	5.39	7.68±3.50
<b>November</b>	6.12	3.80±1.45	4.77	3.34±1.23	3.06	3.22±0.85
<b>Zinc (µg/L)</b>						
<b>September</b>	35.47	28.26±14.31	28.84	20.29±8.82	13.23	20.51±5.98
<b>October</b>	42.55	15.11±7.13	59.81	26.60±12.62	5.39	18.02±4.99
<b>November</b>	29.29	17.97±3.12	26.39	17.92±5.25	12.92	15.39±4.87

**Table 5** The heavy metals concentrations in rice plants

Element	Rice plant part	Site A	Site B	Site C
<b>Cd (µg/kg)</b>	<b>Root</b>	140.45±32.17	105.32±32.04	86.32±23.06
	<b>Shoot</b>	17.04±4.58	18.94±5.68	13.74±3.93
	<b>Grain</b>	10.42±3.70	7.12±2.32	5.11±1.52
	<b>Husk</b>	6.65±2.04	5.48±1.79	3.43±1.51
<b>Cu (mg/kg)</b>	<b>Root</b>	14.62±5.44	6.21±1.75	7.50±1.31
	<b>Shoot</b>	2.44±0.66	1.73±0.85	1.84±0.76
	<b>Grain</b>	2.47±0.38	1.56±0.42	1.05±0.49
	<b>Husk</b>	1.99±0.45	1.58±0.22	1.22±0.29
<b>Zn (mg/kg)</b>	<b>Root</b>	42.91±8.61	33.96±8.74	29.36±20.45
	<b>Shoot</b>	27.48±5.19	22.79±2.96	13.44±1.81
	<b>Grain</b>	12.89±0.86	12.57±0.85	11.65±1.08
	<b>Husk</b>	10.71±1.13	9.95±0.96	8.03±1.62

The average ranges of Cd concentrations varied from 0.10 to 1.31 µg/L in canal water. The minimum Cd concentration was found in water sample of Pradu canal near site C in October while the maximum Cd concentration was found in water sample of Pradu canal near site A in November. The average ranges of Cu concentrations varied from 3.06 to 8.52 µg/L in canal

water. The minimum Cu concentration was found in water sample of Pradu canal near site C in November while the maximum Cu concentration was found in water sample of Phi Lok canal in October. The average ranges of Zn concentrations varied from 5.39 to 59.81 µg/L in canal water. The minimum Zn concentration was found in water sample of Pradu canal near site C in October while the

maximum Zn concentration was found in water sample of Phi Lok canal in October. Heavy metal concentration in canal water that can be presented in descending order as follows: Zn > Cu > Cd. This result is similar to the study of Jantarawattana (2006) on heavy metal levels in canal water of Pradu canal and Losussachan (2006) on heavy metal levels in canal water of Pradu canal. Cd, Cu and Zn concentrations in Pradu and Phi Lok canals water did not exceed Thailand's surface water quality standard for class 3 (PCD, 2004). The results of heavy metal concentrations in canal water are described as showed in Table 4.

Heavy metal concentrations in paddy field waters can be presented in descending order as follows: Zn>Cu>Cd. The important factor regulating heavy metals is the pH value of water. Decreasing the pH of water will increase heavy metal in water and plant uptake (Halwart and Gupta, 2004). From the present study, the pH values in paddy field waters and canal waters were neutral

to alkaline (7.05 to 8.33) therefore the Cd, Cu and Zn concentrations in waters were low and did not exceed the maximum values of Surface Water Quality Standards of Thailand (Class 2: 0.05 mg Cd/L, 0.1 mg Cu/L and 1.0 mg Zn/L).

#### 4. Discussions

Heavy metal concentrations between paddy soils before rice cultivation and soil after rice cultivation were compared during the rice cultivation of August to December 2007 using the paired sample T-test (as shown in Table 6). The results of the statistical analysis suggested that Cd, Cu and Zn concentrations in paddy soils before and after rice cultivation at Sites C and B are not significantly different. In case of Site A, it was found that Cd and Zn concentrations of paddy soils before and after rice cultivations are not significantly different while Cu concentrations are significantly different at the level of 0.05.

**Table 6** T-test of heavy metal concentrations in paddy soils between before and after rice cultivation

Element	Station		N	Mean	S.D.	Mean Difference	T	df	Sig 1 tailed
Cadmium	Site A	before	12	48.32	6.31		-		
		after	12	50.51	8.70	-2.19	0.706	22	0.244
	Site B	before	12	36.63	8.65		-		
		after	12	37.01	7.48	-0.38	0.115	22	0.455
	Site C	before	6	51.70	8.1				
		after	6	47.28	5.06	4.42	1.132	10	0.142
Copper	Site A	before	12	18.86	0.74		-		
		after	12	20.20	0.64	-1.34	4.729	* 22	0
	Site B	before	12	19.92	1.71		-		
		after	12	20.96	2.34	-1.04	1.242	22	0.114
	Site C	before	6	16.44	0.51				
		after	6	16.38	0.23	0.06	0.274	10	0.395
Zinc	Site A	before	12	65.35	3.57		-		
		after	12	66.77	3.29	-1.42	1.016	22	0.160
	Site B	before	12	64.74	3.93		-		
		after	12	65.77	4.10	-1.02	0.624	22	0.270
	Site C	before	6	55.71	1.71				
		after	6	55.23	0.87	0.48	0.606	10	0.279

Independent sample t-test: Correlation is significant at the 0.05 level (1-tailed)

Heavy metal accumulation in rice growing process had been studied. Rice growers of paddy fields at the three study sites, i.e., Sites A, B and C, had used the land for growing rice for more than twenty years. The conventional practice using chemical fertilizers and pesticides is still common in the study area. Site C is one of the few plots that have been changed to organic practice recently. In case of Site C the rice grower has changed for five years. However, the organic practice according to the rice grower's understanding was rather different from the international definition. Rice growers only did not apply chemical fertilizers and pesticides. Moreover, it is unfortunate that all study sites were flooded by heavy rains. Thus, rice plants at site C might be taken heavy metals from other surrounding activities including conventional paddy fields and fish pond. Sirichotiwong (2006) studied fish pond in the same area of the present study and found that heavy metal concentrations in waters of fish pond were 0.133 mg/L Cu, 0.04 mg/L Zn and 0.0011 mg/L Cd.

The fate of metals in the soil environment is dependent on both soil properties and environmental factors. The concentration of metal in soil solution is influenced by the nature of both organic (citrate, oxalate, fulvic and dissolved organic carbon) and inorganic ( $\text{H}_2\text{PO}_4^-$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$ ) ligand ions through their influence on metal sorption processes (Naidu *et. al.*, 2001). In generally, heavy metals are immobile in soil compartment (except in very sandy soils). The mobility and bioavailability of heavy metals are influenced by many factors including pH, redox potential, temperature, soil constituents, cation exchange capacity and organic compounds. The heavy metal uptake by plants and crops depends on the uptake by root surface and hence transfer of the metal from root surface into the root; the

concentration and speciation of the metal in soil solution; and its translocation from root to the shoot (Mukherjee, 2001).

From the study it was found that heavy metal concentrations accumulated in paddy soils among the three sites can be presented as follows: Cd concentration in paddy soils before rice cultivation at Sites C > A > B and paddy soils after rice cultivation at Sites A > C > B and Cd concentrations of Sites A, B increased while those of Site C decreased. It was found that Cu concentrations in paddy soils at Sites B > A > C and Zn concentrations in paddy soils at Sites A > B > C. Cd concentrations at Site C were higher than those of other sites. As described earlier, rice grower of Site C used effective microorganism (EM) and cow manures. EM was used to improve physical, chemical and biological environments of soil and suppressed soil borne pathogens and pests. Also, it can increase the efficacy of organic matters as fertilizers and promote germination, flowering and ripening in plants. The average pH values of EM are 3.3 to 6.2 depending on substrates such as plant, snail, fish (Department of Agriculture, 2004). Cadmium has the stable complexes form in clay soil and long half-lives in nature (1.3 to  $9.3 \times 10^{15}$  years dependent on Cd isotope) (Argonne National Laboratory, 2005). The most importance factors influencing heavy metals accumulation in soils are clay mineral, CEC, metal oxides/ oxyhydroxides (Fe and Mn oxide) and humic substance associated with natural organic meter (Chuangcham *et.al.*, 2008).

## 5. Conclusions

From the present study, the accumulation and distribution of some heavy metals in parts of rice plant may be determined in three parts such as root, shoot and grain (seed and husk) because

the values of heavy metal concentration in seed and husk were like. Most of the heavy metals studied were found to accumulate mainly in roots of rice plant and paddy soil, while other parts including the shoot, grain, husk and water contained low levels. The enough number of rice plant samples is important for analysis. When compare the heavy metal concentration in conventional paddy field and organic paddy field, it was found that heavy metal concentration in conventional paddy field was higher than organic paddy field, thus farmer should be using organic agriculture practice for produce the product food safety. The heavy metal accumulated in shoot may be transferred to higher tropic levels by human or animal consumed and decomposed to the food chain such as animal feed, a straw mushroom spore and straw-covered vegetable. Toxic heavy metal (Cd) concentration in rice seed samples was lower than the permissible levels for human consumption.

Cadmium concentrations studied in chemical or organic fertilizer are rare, cow manure and herbicides are unavailable data in Thailand. But that used in paddy field activity.

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