Assessment of Agricultural Pesticide Contamination and Health Risk in Main Rivers of Thailand

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Abstract

Agricultural production has been one of the major economic resources of Thailand. Maintaining agricultural yields is vital to economic growth and stability. Hence, the application of pesticides in agriculture has been employed to boost yields and avoid unanticipated consequences. This study was conducted to assess pesticide contamination in the Chao Phraya, Pa Sak, and Tha Chin rivers, Thailand. Water samples of these three rivers were collected in both dry and wet seasons from 2009 to 2016. Gas chromatography was used for pesticide analyses of organochlorine, organophosphate, carbamate, pyrethroid, and triazine. The results revealed that organochlorine (endosulfan), organophosphate (chlorpyrifos, malathion, pirimiphos methyl, EPN and diazinon), carbamate (carbaryl), and triazine (ametryn and atrazine) pesticides were detected in the range of 0.01 to 13.40 μg/L in these three rivers. Atrazine of the triazine group was the most frequently detected herbicide. The highest concentration of 13.40 μg/L was detected in the Pa Sak river in the wet season, right after pesticide application. However, the detected levels did not exceed the maximum allowable concentration of guidelines by the World Health Organization or U.S. Environmental Protection Agency. The detected pesticides were non-carcinogenic. The hazard quotient estimate was <1 showing a low health risk using these waters for drinking.

Keywords: Agriculture; pesticide; river; contamination; health risk

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1. Introduction

Thailand, a major food-producing country, has gained much income from exporting agricultural commodities. In 2014, agricultural products with a value of $39,766 million were exported to foreign countries, accounting for 17.9% of total exports (OAE, 2015). The demand for increased agricultural productivity has caused a rapid increase in pesticide use to protect products lost to weeds, a multitude of pests, and diseases. The reasons for the extensive use of pesticides in Thailand are the wide variation in climate and crops. In Thailand, most pesticides used in the country are imported, and their amount has increased significantly over time. The Office of Agriculture Regulation (OAR) reported that pesticide use increased four-fold in the last decade, with nearly 100,000 tons of active ingredients imported into Thailand between 2004 and 2015. Herbicides made up the most significant proportion of imported pesticides (OAR, 2016).

The Chao Phraya, Pa Sak, and Tha Chin are the three main rivers in central Thailand. The pollution in these rivers is mainly caused by discharge from agriculture, aquaculture, and industries located along these rivers. Wastes from agricultural discharge are deposited in various environmental components of the aquatic ecosystem, particularly in the water column. Such pollutants are derived mainly from amounts of applied pesticides washed off from fields and transported to surface water through surface runoff. Generally, the pesticide residues that are detected in water bodies are closely related to the agricultural practices of the nearby area. Pesticides seep into soils and groundwater, which can end up in drinking water (Jaipieam *et al*., 2009). Therefore, pesticides can enter the human body through oral intake of contaminated water.

Exposure or consumption of pesticides may cause long-term adverse effects. For example, organochlorines are widely known to damage the central nervous system (Costa *et al*., 2008), thyroid and bladder, while chlorpyrifos induces neurotoxicity (Steenland *et al*., 2000). Endosulfan blocks chloride uptake induced by GABA in primary cultures of cortical neurons (Pomes *et al*., 1994; Vale *et al*., 2003). Atrazine induces aromatase and promotes the conversion of testosterone to estrogen, which could risk impaired sexual development in mammals (Haye *et al*., 2002). Several studies have monitored the presence of pesticide contamination in the water of the Chao Phraya, Pa Sak, and Tha Chin rivers (Chulintorn *et al*., 2002; Chatrasuntiprapa *et al*., 2002). More recent studies of this area are still limited so far. Therefore, this study aims to evaluate the agricultural pesticide contamination in three main Thai rivers, i.e. Chao Phraya, Pa Sak, and Tha Chin rivers, and assess its potential non-carcinogenic and cancer risk for human health.

2. Methods

2.1 Study area and sampling sites

The sampling was conducted from the Chao Phraya, Pa Sak, and Tha Chin Rivers in the dry and wet seasons. A total of 46 and 22 water samples were collected in the Chao Phraya river in 2009 and 2016, respectively. In Pa Sak, a total of 52 and 52 water samples were collected in 2010 and 2014, respectively. At the same time, a total of 48 and 48 water samples were collected from the Tha Chin river in 2011 and 2015, respectively. Four one liter (1-L) water samples for composite sampling were collected in high density polyethylene (HDPE) bottles at every sampling site. Water samples were collected using a Kemmerer sampler at a depth of 50-100 cm below the water surface. All samples were stored in an icebox at 4 $^{\circ}$C until transferred to the laboratory for analysis. The number and geographic location of the sampling points were based on the total length of each river and focused on locations in nearby agricultural areas in the river's basin. A global positioning system (GPS) was used to indicate the sampling locations, as shown in Figure 1.

2.2 Analytical procedure

Pesticide contaminates in water samples were extracted by liquid-liquid extraction and analyzed by Gas Chromatography. Pesticides analyzed included those of the organochlorines group: aldrin, α- hexachlorocyclohexane, α-endosulfan, β- endosulfan, dieldrin, endosulfan sulfate, endrin, γ- hexachlorocyclohexane, heptachlor, heptachlor epoxide, *o,p'*-DDE, *o,p'*-DDT, *o,p'*-TDE, *p,p'*-DDE, *p,p'*-DDT, and *p,p'*-TDE; of the organophosphates group: chlorpyrifos, diazinon, *O*-Ethyl *O*-(4-nitrophenyl) phenylphosphonothioate (EPN), ethion, malathion, methidathion, parathion methyl, pirimiphos methyl, profenofos, and triazophos; of the carbamate group: carbaryl, carbofuran, Isoprocarb, fenobucarb, metolcarb, and promecarb; of the pyrethroids group: bifenthrin, cypermethrin, cyfluthrin, λ-cyhalothrin, δ-methrin, fenvalerate, and permethrin; and of the triazine group: ametryn, atrazine, and metribuzin (George *et al*., 2016).

2.3 Extraction of water samples

Pesticide contamination of water samples was analyzed by gas chromatography. A volume of 800 ml water sample was placed in a 1000 ml Erlenmeyer flask to extract the pesticide concentration. A solution of 100 ml of n-hexane was added to each sample, which was shaken for 3 minutes by a separatory funnel shaker. The extract was dropped through a glass funnel containing filter paper No. 1, and 5 grams of sodium sulfate anhydrous was added to eliminate water residue. The extraction was repeated twice with 50 ml n-hexane, and the extracts were combined. The extracted sample was evaporated to dryness using a vacuum at 40 $^{\circ}$C. The concentration in the evaporated flask was then re-dissolved in 1 ml of n-hexane. The filtrate was kept frozen at -20 $^{\circ}$C before the analysis was made.

2.4 Quality assurance of analysis

A reagent blank was analyzed for each batch of samples to determine if there was any unforeseen contamination. The accuracy of the analytical process was confirmed using 3 replicates of each recovery. The mean recovery of pesticides through the analytical procedures used was between 70 and 109 %. The limits of detection

(LOD) of all pesticides were in the range of 0.001 to 0.03 µg/L, and the limits of quantitation (LOQ) were in a range from 0.005 to 0.05 µg/L. Working standard solutions of 5 points in each pesticide were used, and the concentration levels were between 0.01 and 2.50 µg/mL. The correlation coefficients (R^2) of all the calibration curves of the pesticides were in the range of 0.995 - 0.999 (CIPAC, 2003).

2.5 Assessment of health risk

The non-carcinogenic and carcinogenic risks of detected pesticides to adults and children were calculated using a model described by U.S. EPA, 2011. The hazard quotient (HQ) was used to calculate the non-carcinogenic risk for humans by direct ingestion of surface water as drinking water (U.S. EPA, 2005a). When HQ for a pesticide is higher than 1, potential adverse health effects are possible, whereas when HQ is lower than 1, adverse health effects are not likely. As for carcinogenic concerns, carcinogenic risk (R) is calculated according to the equation described by the U.S. EPA, 2000. It is an acceptable risk if R is between 10^{-6} and 10^{-4}. When R exceeds 10^{-4}, potential carcinogenic effects are possible (U.S. EPA, 2005a).

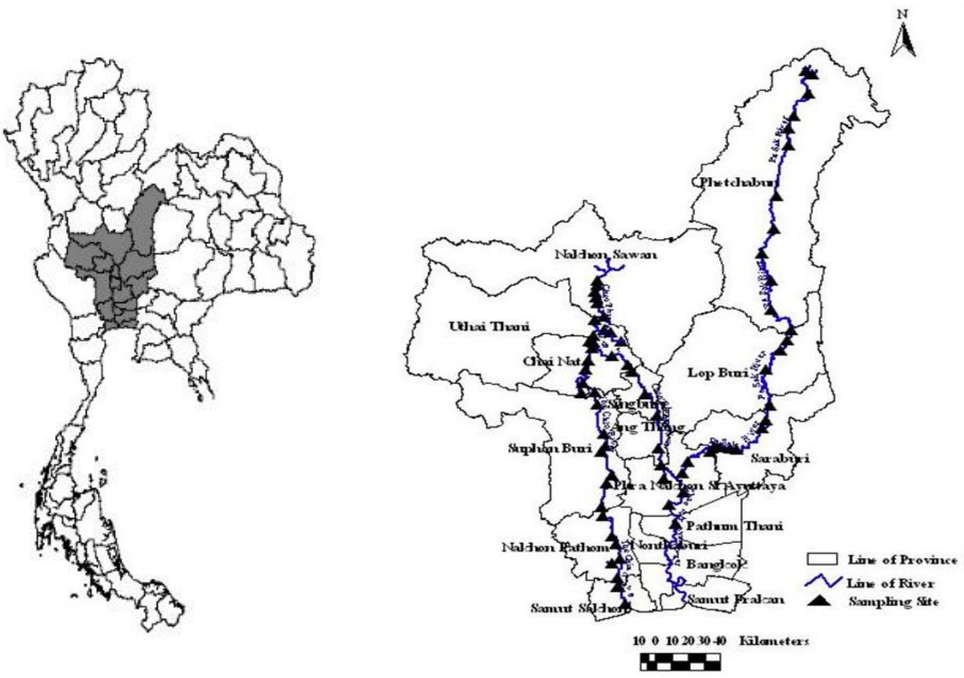

Figure 1 Sampling locations of water collected from the Chao Phraya, Pa Sak, and Tha Chin rivers.

3. Results and Discussions

3.1 Pesticide in the Chao Phraya River

Chlorpyrifos and pirimiphos methyl were detected in both dry and wet seasons in 2009 (Table 1). Endosulfan, EPN, and malation were detected only in the dry season. Pyrethroid, carbamate, and triazine were not detected in any sample or season. Most insecticides were detected in the dry season because they were used for dry crops (for example, corn, maize, tobacco, and soybean). In addition, the volume of river water is low in the dry season. Similar results have been reported in previous studies (Chulintorn *et al*., 2002). In 2016, there were two pesticides detected, atrazine and ametryn. Atrazine was the most frequently detected pesticide with a detection frequency of 100% in both the dry and wet seasons. Atrazine concentrations in dry and wet seasons ranged from 0.13 to 0.28 µg/L and 0.16 to 0.40 µg/L, respectively. The highest concentration of atrazine was found at Nakhon Sawan and Sing Buri in the dry season and Sing Buri in the wet season. Ametryn was detected only in the wet season. The concentrations were detected in the range of 0.01 to 0.04 µg/L. Sugarcane plantations predominant in the area with nearly 10,000 rai, having intensive use of atrazine herbicide to control weeds, pre-emergent in the fields (DOAE, 2017). The values for water solubility and log K_{ow} of atrazine and ametryn are 35 mg/L (log K_{ow} = 2.7) and 200 mg/L (log K_{ow} = 2.63), respectively (IUPAC, 2017); as such they are weakly absorbed into soil. Therefore, the concentration in water must have resulted from pesticide contamination in the soil after herbicide application to crops that might be leached out from the soil via runoff. Previous studies of the Chao Phraya River in 1995-1996 revealed pesticide contamination from insecticides (organochlorines, organophosphate and carbamate) (Chulintorn *et al*., 2002); while in this present study, there was no detection of insecticides organochlorine, organophosphate, carbamate, or pyrethroid in any seasons. This might be due to the fact that the year 2004 was declared as the year of food safety in Thailand. This resulted in a policy to reduce the amount of pesticide in plantations and monitoring pesticide residue in the market (FAO, 2014). As a result, pesticides use in plantations decreased. In addition, some organochlorines pesticides (endosulfan) were banned in 2004 in Thailand (Notification of MOI, 2004). Pesticide contamination results in 2009 were all insecticides (endosulfan, chlorpyrifos, EPN, malathion, pirimiphos methyl) and chlorpyrifos was the most frequently detected. In 2016, only herbicides (ametryn, atrazine) were found in both seasons due to large amounts applied to crops (OAR, 2016).

3.2 Pesticides in Pa Sak River

Results from the Pa Sak river in 2010 showed that most pesticides were detected in the wet season. Atrazine was the most frequently detected pesticide in both dry and wet seasons. A detection frequency of 98% was found in both seasons. The highest concentrations detected in the dry and wet seasons were 0.22 and 3.23 µg/L, respectively. Carbaryl was detected at high concentrations in the wet season. In 2014, ametryn and atrazine were the most frequently detected pesticides in the wet seasons with a detection frequency of 52% and 67%, respectively. The highest concentration of atrazine was detected in the wet season at 13.40 µg/L. Because the Pa Sak river has a sufficient volume of water throughout the year, cultivation along it is year round without rotating crops throughout the year. Since pesticides are continuously applied to the crops, they remain in soil and runoff by rain to nearby waters. Similar pesticide results have been reported in previous studies (Chulintorn *et al.*, 2002).

3.3 Pesticides in Tha Chin River

Atrazine was the most frequently detected pesticide in both seasons in 2011. Endosulfan is the Persistent Organic Pollutants (POPs) pesticide that was detected in both seasons, with a detection frequency of 46%. Similar results have been reported in previous studies (Chatrasuntiprapa and Harutaitanasunti, 2002). In 2015, ametryn was only detected in the dry season. Organochlorine, organophosphate, carbamate, and pyrethroid were not detected in any season. Most plantations in this area were for horticulture and vegetables. The variety of pesticides used in this area included insecticides, herbicides, fungicides, and plant growth regulators.

Herbicides make up the most significant proportion of imported pesticides in Thailand (OAR, 2016), and they are intensively used in plantations. The most frequently detected pesticides were herbicides, particularly, atrazine which was detected in all rivers and in both seasons. It was detected in all sampling sites of the Chao Phraya river in 2016. It is used to control broadleaf weeds predominantly in corn, sorghum, and sugar cane during May to June, with persistence associated with rainfall events when concentrations in the rivers result from pesticides in the soil after pesticide applications on crops leach out from the ground via runoff (Kristin *et al.*, 2012). Pesticide contamination in water depends on several factors, such as the closeness of crop field to surface water, characteristics of surrounding fields (soil, slope, and distance to water bodies), and climate conditions (temperature, humidity, wind, and precipitation) (Edwin, 1996). In this current study, pesticides were detected in higher concentrations in the wet season than in the dry season because of the outbreak of pests due to the high humidity (Karuppuchamy and Venugopal, 2016). Thus, most pesticides are detected in the wet season, while pesticides are detected in low concentrations in the dry season due to some pesticides accumulated in sediment (Poolpak *et al.*, 2008). However, this study did not analyze sediments that require more complicated sampling. In addition, the water sampling from each river was not conducted in the same year, so results between rivers are not comparable.

In Thailand, endosulfan was banned in 2004 (Notification of MOI, 2004), but it remained in the river due to its persistence in the environment (Stockholm Convention on POPs, 2008). Similar results have been reported in previous studies (Abrantes *et al.*, 2010; Zheng *et al.*, 2016). However, endosulfan was not detected in the current year. Observed pesticide residue concentrations in surface water correlate with current pesticide quantities purchased and applied. The organophosphate pesticide most frequently detected was chlorpyrifos because it makes up the largest proportion of insecticide imported to Thailand (OAR, 2016).

Table 1. The concentration of pesticides detected in water samples from the Chao Phraya, Pa Sak, and Tha Chin rivers

Year	Frequency (%)		Pesticides	HQ			
				Dry season		Wet season	
				Min.	Max.	Min.	Max.
2009 (N = 46)	1	(2.2)	Endosulfan	0.62	0.62	ND	ND
	6	(13.4)	Chlorpyrifos	0.05	0.06	0.01	0.02
	1	(2.2)	EPN	0.06	0.06	ND	ND
	1	(2.2)	Malathion	0.05	0.05	ND	ND
	1	(2.2)	Pirimiphos methy	0.10	0.10	0.09	0.09
2016 (N = 22)	5	(22.7)	Ametryn	ND	ND	0.01	0.04
	22	(100)	Atrazine	0.13	0.28	0.16	0.40
2010 (N = 52)	2	(3.8)	Endosulfan	0.02	0.02	ND	ND
	1	(1.9)	Chlorpyrifos	0.04	0.04	ND	ND
	1	(1.9)	Diazinon	0.06	0.06	ND	ND
	1	(1.9)	Ethion	0.05	0.05	ND	ND
	3	(5.7)	Carbaryl	ND	ND	0.19	0.44
	28	(53.8)	Ametryn	0.01	0.06	0.08	8.04
	51	(98.1)	Atrazine	0.02	0.22	0.05	3.23
2014 (N = 52)	27	(51.9)	Ametryn	0.04	0.83	0.02	0.47
	35	(67.3)	Atrazine	0.19	3.28	0.03	13.40
2011 (N = 48)	22	(45.8)	Endosulfan	0.02	0.03	0.01	0.02
	2	(4.2)	Chlorpyrifos	ND	ND	0.03	0.03
	22	(45.8)	Ametryn	ND	ND	0.22	0.79
	39	(81.3)	Atrazine	0.06	0.29	0.24	0.42
2015 (N = 48)	5	(10.4)	Ametryn	0.04	0.06	ND	ND
	25	(52.1)	Atrazine	0.06	0.09	0.06	0.22

N = number of sample, ND= not detected, and frequency = number with detected amounts (percentage with positive samples).

Chlorpyrifos is used primarily to control foliage and soil-borne insect pests on various food and feed crops. Besides, chlorpyrifos is unstable in water, and the rate at which it is hydrolyzed increases with temperature. In water at pH 7.0 and 25 □C, it has a half-life of 35 to 78 days (Howard, 1989).Even though pesticides have a certain half-life, their continuous application may accumulate in an area. However, none of the pesticides in the three rivers tested exceeded the maximum allowable concentrations of standard guidelines, as shown in Table 2.

3.4 Health Risk Assessment for Children and Adults in Thailand

All pesticides detected at low concentrations in the three rivers: organochlorine (endosulfan), organophosphate (chlorpyrifos, diazinon, ethion, EPN, malathion, pirimiphos methyl), and carbamate (carbaryl), triazine (ametryn, atrazine) are non-carcinogenic compounds according to the Integrated Risk Information System assessment of the U.S. EPA, 2015. Non-carcinogenic detected pesticides are assessed for potential health risks using the hazard quotient (HQ) value. In order to evaluate the situation for Thailand, the amount of water consumption according to the classification of age group and body weight in Thailand (ACFS, 2016) is used (Table 3).

Table 2 Guidelines of allowable levels of pesticides in drinking water (μg/L)

Pesticides	WHO[a]	EPA[b]	Canada[c]	Australia[d]
Ametryn	N/A	N/A	N/A	70
Atrazine	100	3	5	20
Carbaryl	50	N/A	90	30
Chlorpyrifos	30	N/A	90	10
Diazinon	N/A	N/A	20	4
Endosulfan	20	N/A	N/A	20
Ethion	N/A	N/A	N/A	4
Malathion	N/A	N/A	190	70
Pirimiphos methyl	N/A	N/A	N/A	90

N/A = not available

Source: [a]World Health Organization; WHO (2017)

[b]United States Environmental Protection Agency; EPA (2012)

[c]Health Canada (2017)

[d]National Health and Medical Research Council; NHMRC, Natural Resource Management Ministerial Council; NRMMC (2011)

Children are usually subject to higher risk when compared to adults, which has been reported in previous studies (Emmaluel *et a*l., 2015; Wu *et al.*, 2014). Children (<6 years) are more vulnerable than adults to chemicals found in the environment (U.S. EPA, 2005b). The Hazard quotient (HQ) values were calculated on two groups of the U.S. EPA, 2010 (children, <3 to 5.9 years and adults, 6 to over 65 years) by dividing the estimated amount of ingested concentration pesticide per kilogram body weight of 13.90 kg and 55.84 kg for children and adults, respectively. The water consumption rate was 0.81 L/day for children and 1.01 L/day for adults. The HQ value <1 for children and adults was calculated for all detected pesticides. These show low potential risk for humans, implying that no adverse health effects occur in children and adults. The results are summarized in Table 4.

Table 3 Amount of water consumption in each age group in Thailand

Age group (year)	Body weight (kg)	Amount of water consumption (liters/day)
< 3	10.55	1.105
3 to 5.9	17.25	0.515
6 to 12.9	33.38	0.745
13 to 17.9	53.42	0.996
18 to 34.9	63.12	1.171
35 to 64.9	63.53	1.217
65 years and over	55.77	0.943

Source: National Bureau of Agricultural Commodity and Food Standard (ACFS, 2016).

Pesticides can affect human health and the environment, depending on how much chemical is present, the length and frequency of exposure, and the pesticide's toxicity. Effects also depend on the age and health of a person and the condition of the environment when exposure occurs (U.S. EPA, 2016). The risk assessment of pesticides on human health is not an easy and remarkably accurate process because of differences in the periods and levels of exposure, the types of pesticides used, and the environmental characteristics of the areas where pesticides are usually applied. Regarding the adverse effects on human health depend on the physical properties of the pesticide, the measures are taken during its application, the amount used, the adsorption on soil colloids, the weather conditions prevailing after application, and how long the pesticide persists in the environment (Christos and Ilias, 2011).

4. Conclusions

Herbicides comprised the vast majority of detected pesticides in the three rivers tested. Atrazine was detected in all rivers with different concentrations. Characterizing pesticide fate and transportation in the environment is important in understanding human exposures. Pesticide fate is determined by chemical properties and environmental conditions.

Table 4. HQ for non-carcinogenic detected pesticides in the Chao Phraya, Pa Sak, and Tha Chin rivers for children and adults in Thailand

Year	Pesticides	HQ			
		Dry season		Wet season	
		Child	Adult	Child	Adult
The Chao Phraya River					
2009	Endosulfan	5.77×10^{-3}	1.80×10^{-3}	NA	NA
	Chlorpyrifos	1.12×10^{-2}	3.48×10^{-3}	3.73×10^{-3}	1.16×10^{-3}
	EPN	3.35×10^{-1}	1.05×10^{-1}	NA	NA
	Malathion	1.40×10^{-4}	4.35×10^{-5}	NA	NA
	Pirimiphos methyl	5.59×10^{-4}	1.74×10^{-4}	5.03×10^{-4}	1.57×10^{-4}
2016	Ametryn	NA	NA	2.48×10^{-4}	7.74×10^{-5}
	Atrazine	4.47×10^{-4}	1.39×10^{-4}	6.39×10^{-4}	1.99×10^{-4}
Pa Sak River					
2010	Endosulfan	1.86×10^{-4}	5.81×10^{-5}	NA	NA
	Chlorpyrifos	7.45×10^{-3}	2.32×10^{-3}	NA	NA
	Diazinon	1.34×10^{-3}	4.18×10^{-4}	NA	NA
	Ethion	5.59×10^{-3}	1.74×10^{-3}	NA	NA
	Carbaryl	NA	NA	2.46×10^{-4}	7.66×10^{-5}
	Ametryn	3.73×10^{-4}	1.16×10^{-4}	4.99×10^{-2}	1.56×10^{-2}
	Atrazine	3.51×10^{-4}	1.09×10^{-4}	5.16×10^{-3}	1.61×10^{-3}
2014	Ametryn	5.15×10^{-3}	1.61×10^{-3}	2.92×10^{-3}	9.10×10^{-4}
	Atrazine	5.24×10^{-3}	1.63×10^{-3}	2.14×10^{-2}	6.67×10^{-3}
Tha Chin River					
2011	Endosulfan	2.79×10^{-4}	8.71×10^{-5}	1.86×10^{-4}	5.81×10^{-5}
	Chlorpyrifos	NA	NA	5.59×10^{-4}	1.74×10^{-4}
	Ametryn	NA	NA	4.90×10^{-3}	1.53×10^{-3}
	Atrazine	4.63×10^{-4}	1.44×10^{-4}	6.71×10^{-4}	2.09×10^{-4}
2015	Ametryn	3.73×10^{-4}	1.16×10^{-4}	NA	NA
	Atrazine	1.44×10^{-4}	4.48×10^{-5}	3.51×10^{-4}	1.09×10^{-4}

NA = not applicable.

In the Chao Phraya river, the herbicides found in higher concentrations in water samples were ametryn and atrazine. Comparison of the results obtained in the present study with those from a previous study conducted in 2009 indicates a decreasing trend in the contamination of water by endosulfan, chlorpyrifos, EPN, malathion, and pirimiphos methyl and an increasing trend in the contamination by atrazine and ametryn. These results may, at least partially, be due to changes in agriculture practice. None of the pesticides measured exceeded the maximum allowable concentrations established by WHO (WHO, 2017) and Australia (NHMRC, 2011).

The Pa Sak river also had ametryn and atrazine herbicides in higher concentrations from its tested water samples. Comparison of the results obtained in the present study with those in a previous study conducted in 2010 indicates a decreasing trend in the contaminations by endosulfan, chlorpyrifos, diazinon, ethion, and carbaryl and an increasing trend in the contamination from atrazine and ametryn. The seasonal and spatial variations of pesticide distribution were related to land use patterns, especially of field crops. The herbicides ametryn and atrazine are widely used in the wet season.

In the Tha Chin river, insecticides and herbicides were detected in both seasons in 2011, while primarily herbicides were detected in 2015. There is a decreasing trend in contamination from endosulfan and chlorpyrifos, and a continuing trend of contamination from atrazine and ametryn.

In terms of potential health risk, all pesticides detected were non- carcinogenic pesticides in the Chao Phraya, Pa Sak, and Tha Chin Rivers. The HQ of water revealed that the water in the three rivers was rated low hazard (HQ <1) and had a low potential risk through human ingestion of drinking water.

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