

HEALTH RISK ASSESSMENT FOR HEAVY METALS IN CHAO PHRAYA RIVER BASIN



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จุฬาลงกรณ์มหาวิทยาลัย
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การประเมินความเสี่ยงทางสุขภาพของโลหะหนักในกลุ่มแม่น้ำเจ้าพระยา



วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรมหาบัณฑิต
สาขาวิชาพิษวิทยาอุตสาหกรรมและการประเมินความเสี่ยง ภาควิชาวิทยาศาสตร์สิ่งแวดล้อม
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ณัฐธินิชา นทีกุลเจริญ : การประเมินความเสี่ยงทางสุขภาพของโลหะหนักในลุ่มแม่น้ำ
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การปนเปื้อนของโลหะหนัก ได้กลายเป็นปัญหาที่สำคัญ เนื่องจากความเป็นพิษของโลหะหนัก ที่มีผลต่อสิ่งแวดล้อมและยังส่งผลกระทบต่อสุขภาพของมนุษย์ ลุ่มแม่น้ำเจ้าพระยา เป็นพื้นที่ที่สำคัญในการค้า อุตสาหกรรม รวมถึงเป็นที่อยู่อาศัยของประเทศไทย การศึกษานี้มีเป้าหมายในการประเมินผลกระทบของโลหะหนักต่อสุขภาพของมนุษย์และวิเคราะห์ความสัมพันธ์ระหว่างการใช้น้ำและการปนเปื้อนของโลหะหนักในลุ่มแม่น้ำเจ้าพระยา การศึกษานี้ได้ใช้สหสัมพันธ์แบบเพียร์สันในการวิเคราะห์ความสัมพันธ์ระหว่างการปนเปื้อนของโลหะหนักและค่าพารามิเตอร์ของน้ำ ผลที่ได้แสดงว่าในระหว่างปี 2552 ถึงปี 2556 ค่าความเข้มข้นของเหล็กมีความสัมพันธ์เชิงบวกต่อค่าความขุ่น (0.640), ค่าฟอสเฟตโดยรวม (0.622), ค่าของแข็งแขวนลอย (0.542) และค่าความเข้มข้นของนิเกิล (0.513) ในแม่น้ำ แต่มีค่าความสัมพันธ์เชิงลบต่อค่าความเข้มข้นของสังกะสี (-0.517) โดยมีค่านัยสำคัญทางสถิติที่ $P < 0.01$ ในขณะช่วงปี 2557 ถึงปี 2561 ผลลัพธ์แสดงว่าค่าความเข้มข้นของเหล็กมีความสัมพันธ์เชิงบวกต่อค่าความขุ่น (0.900), ค่าของแข็งแขวนลอย (0.671), ค่าความเข้มข้นของแมงกานีส (0.607) และ ค่าความเข้มข้นของนิเกิล (0.512) โดยมีค่านัยสำคัญทางสถิติที่ $P < 0.01$ รวมถึง ค่าความเข้มข้นของแคดเมียมก็มีความสัมพันธ์เชิงบวกต่อ ค่าความเข้มข้นของโครเมียม (0.509) นอกเหนือจากนี้ผลลัพธ์ยังแสดงว่าในช่วงปี 2552 ถึงปี 2554 ความเสี่ยงทางสุขภาพของมนุษย์ในบางสถานีในจังหวัดลำปาง เชียงใหม่ และสุโขทัยยังอยู่ในเกณฑ์ที่เป็นที่ยอมรับได้ และได้มีการเปลี่ยนแปลงเป็น มีความเสี่ยงต่อสุขภาพของมนุษย์อย่างมากในช่วงปี 2558 ถึงปี 2561 ค่าดัชนีมลพิษจากโลหะหนักได้ถูกคำนวณและเปรียบเทียบกับข้อมูลการใช้น้ำที่ผลลัพธ์ที่ได้แสดงถึงการเปลี่ยนแปลงของพื้นที่ป่าไม้เป็นพื้นที่เกษตรกรรม โดยเฉพาะในพื้นที่บริเวณแม่น้ำยมที่อาจเป็นสาเหตุของการปนเปื้อนของโลหะหนักจากปุ๋ย ซึ่งข้อมูลทั้งหมดถูกสร้างในรูปแบบแผนที่เพื่อให้เข้าใจได้ง่าย

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Heavy metal contamination has become a serious concern due to its toxicity not only on ecosystems but also directly on human health. The Chao Phraya River Basin is the main area important for commerce, industries, and residence in Thailand. This study aimed to evaluate the impact on human health due to heavy metal contamination and identify the relationship between land use and heavy metal contamination in the Chao Phraya River. Pearson correlation was applied to analyze the relationship between heavy metal concentration and water parameters. The results indicated that during 2009-2013, Fe concentration positively correlated to turbidity (0.640), total phosphate (0.622), suspended solids (0.542), and Ni level (0.513) in the river but negatively correlated to Zn level (-0.517) at $P < 0.01$. From 2014 to 2018, the results revealed that Fe concentration positively correlated to turbidity (0.900), suspended solids (0.671), Mn (0.607), and Ni level (0.512) at $P < 0.01$, and Cd concentration also positively correlated to Cr level (0.509). The results showed that human health risks in some stations in Lampang, Chiang Mai, and Sukhothai province were acceptable from 2009 to 2011 and changed to very high risk on human health from 2015 to 2018. The heavy metal pollution index was calculated and compared to land use data. The results revealed that the change from deforestation into agricultural land, especially in the Yom River area, could be the cause of the heavy metal contamination by fertilizer. All of the data was created to map for easy understanding.

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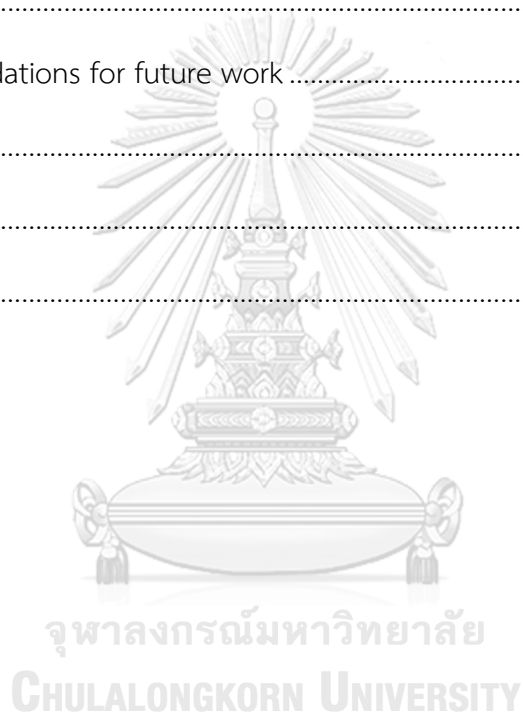
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CHAPTER 1

INTRODUCTION

1.1 Background of the study

Environmental degradation such as global warming, climate change, and pollution has affected human's daily life and raised people's awareness of Environmental knowledge.

Heavy metal contamination has become a serious societal concern due to its abundance, persistence, long-lasting availability, and toxicity (Siddiqui & Pandey, 2019). Heavy metals have the potential to produce a negative effect not only on human health but also on ecosystems (Wang et al., 2017).

Heavy metal contamination occurred from natural processes (such as atmospheric deposition, erosion, and mineral wearing) as well as anthropogenic activities (such as urban and industrial development and agriculture) (Liu et al., 2016; Yang et al., 2015). This study focuses on the water quality and heavy metal concentration in Chao Phraya River Basin.

Chao Phraya River Basin is located in the center of the northern part of Thailand. The Chao Phraya River Basin was mainly agricultural and forest land with an area of 20,523.42 km² and had the Chao Phraya River as the main river, which is 379 kilometer-long. The Chao Phraya River supports 13 million people in various ways, including drinking water and irrigation, and flows into the Gulf of Thailand. As a confluent of the Ping, Wang, Yom, and Nan Rivers, the Chao Phraya River's water quality are greatly affected by upstream activities (Simachaya, 2003).

The Pollution Control Department (PCD) has carried out regular water quality monitoring for Thailand's rivers. Since 1994, the overall water quality of the rivers was found to be polluted or degrading, particularly near urban centers. The historical data showed that the water quality index has not covered every river in Thailand and is complicated for public use.

According to the reason above, there are few studies about health risk assessment in Thailand, especially from the water-quality analysis. This study might

provide the results that are uncomplicated and useful for the inhabitants to have some insight into formulating a protective plan to reduce heavy metal contamination.

1.2 Objectives

1.2.1. To evaluate the data to find the impact on human health due to the heavy metal contamination

1.2.2. To identify the relationship between land use and heavy metal contamination.

1.3 Hypotheses

1.3.1. The health risk from some heavy metals in the Chao Phraya River basin is high.

1.3.2. The concentration of some heavy metals in the Chao Phraya River basin associated with increasing urbanization.

1.4 Scope of the study

1.4.1 The observed data of water quality parameters including (pH, turbidity (tur), conductivity (con), salinity (sal), Dissolved Oxygen (DO), Biochemical Oxygen Demand (BOD), Total phosphorus (TP), ammonia (NH₃), Suspended Solids (SS), Total Dissolve Solid (TDS) and heavy metal concentration (Fe, Cd, Cr, Mn, Ni, Pb, Zn, Cu, Hg, and As) of 5 rivers (Ping, Wang, Yom, Nan, Chao Phraya River) from 65 stations from 2009-2018 are supported by the Pollution Control Department, Ministry of Natural Resource and Environment.

1.4.2 The Land Development Department supported the land use data during 2009-2018, Ministry of Agriculture and Cooperatives.

1.4.3 The water quality data would be verified and calculated for health risk assessment.

1.4.4 All files would be added with semantic and spatiotemporal metadata such as category, location, date, and description for each database on visualizing map (ArcMap 10.4.1).

1.5 Expected outcome

1.5.1 Health risk assessment of heavy metal contamination in the Chao Phraya River basin was clarified.

1.5.2 The correlation between land use and heavy metal concentration in the Chao Phraya River basin over ten years was evaluated.



CHAPTER 2

LITERATURE REVIEW

2.1 Sources and effects of the heavy metal

Heavy metals are naturally occurring elements with a high atomic weight and density at least five times greater than water. Their multiple industrial, domestic, agricultural, medical, and technological applications have led to their wide distribution in the environment, raising concerns over their potential effects on human health and the environment. Their toxicity depends on several factors, including the dose, route of exposure, chemical species, and the age, gender, genetics, and nutritional status of exposed individuals. Because of their high degree of toxicity, arsenic, cadmium, chromium, lead, and mercury rank among the priority metals of public health significance. These metallic elements are considered systemic toxicants and are known to induce multiple organ damage, even at lower levels of exposure. They are also classified as human carcinogens (known or probable) according to the U.S. Environmental Protection Agency and the International Agency for Research on Cancer.

Although heavy metals are naturally occurring elements that are found throughout the earth's crust, most environmental contamination and human exposure result from anthropogenic activities such as mining and smelting operations, industrial production and use, and domestic and agricultural use of metals and metal-containing compounds (Tchounwou et al., 2012).

Organic species of metals may be more or less toxic than inorganic forms. These compounds occur naturally at low levels in drinking water, so they must be carefully regulated (USEPA, 2014).

Arsenic is a toxic heavy metal that poses severe and ecological health hazards to humans. Arsenic has two primary forms: arsenite As (III) and arsenate As (V). Both forms cause acute and chronic toxicity to various organisms, including humans. Exposure to inorganic arsenic can cause various health effects, such as irritation of the stomach and intestines, decreased production of red and white blood cells, abdominal pain, muscular pain, skin changes, and lung irritation, as well as other skin changes such as hyperkeratosis and pigmentation changes. Increased risks of lung and

bladder cancer and arsenic-associated skin lesions have been reported to be related to ingestion of drinking water having arsenic concentrations of 50 ppb. Effects on the cardiovascular system were observed in children consuming arsenic-contaminated water (mean concentration 0.6 mg/l) for an average of 7 years (World Health Organization, 2017).

Cadmium accumulates primarily in the kidneys and has a long biological half-life in humans of 10–35 years. There is evidence that cadmium is carcinogenic by the inhalation route, and IARC (International Agency for Research on Cancer) has classified cadmium and cadmium compounds in Group 2A (probably carcinogenic to humans). However, there is no evidence of carcinogenicity by the oral route and no clear evidence for the genotoxicity of cadmium. The kidney is the main target organ for cadmium toxicity (World Health Organization, 2017). After acute ingesting cadmium, symptoms such as abdominal pain, burning sensation, nausea, vomiting, salivation, muscle cramps, vertigo, shock, loss of consciousness, and convulsions usually appear within 15 to 30 min and may cause osteomalacia, anemia, and teeth discoloration (Charles et al., 2018; Tchounwou et al., 2012).

Any route poorly absorbs chromium (III). The toxicity of chromium is mainly attributable to the Cr (VI) form that can pass through cell membranes and its subsequent intracellular reduction to reactive intermediates. The gastrointestinal tract can absorb it. Cr (VI) caused severe progressive proteinuria, urea nitrogen, and creatinine, elevated serum alanine aminotransferase activity, and hepatic lipid peroxide formation (Tchounwou et al., 2012). IARC has classified Cr (VI) in Group 1 (human carcinogen) and Cr (III) in Group 3 (not classifiable as to its carcinogenicity to humans). Cr (VI) compounds are active in a wide range of in vitro and in vivo genotoxicity tests, whereas Cr (III) compounds are not (World Health Organization, 2017).

Lead is associated with various neurodevelopmental effects, mortality (mainly due to cardiovascular diseases), impaired renal function, hypertension, impaired fertility, and adverse pregnancy outcomes. Impaired neurodevelopment in children is generally associated with lower blood lead concentrations than the other effects. For adults, the adverse effect associated with the lowest blood lead concentrations for

which the weight of evidence is most significant and most consistent is a lead-associated increase in systolic blood pressure (World Health Organization, 2017).

Mercury is mainly toxic in the kidney in humans and laboratory animals following short-term and long-term exposure. In humans, acute oral poisoning results primarily in hemorrhagic gastritis and colitis; the ultimate damage is to the kidney. The overall weight of evidence is that mercury (II) chloride can potentially increase the incidence of some benign tumors at sites where tissue damage is apparent and that it possesses weak genotoxic activity but does not cause point mutations (World Health Organization, 2017).

Nickel can cause severe damage to gastrointestinal distress (e.g., nausea, vomiting, diarrhea), renal edema, and neurological effects (USEPA, 1992). IARC concluded that inhaled nickel compounds are carcinogenic to humans (Group 1) and that metallic nickel is possibly carcinogenic (Group 2B). However, there is no evidence of carcinogenic risk from oral exposure to nickel (World Health Organization, 2017).

Manganese is possible adverse neurological effects at doses not far from the range of essential and can cause weakness/fatigue, gait disturbances, tremors, and dystonia (USEPA, 1995).

Copper and Zinc are essential to maintain physiological processes and functions in humans. They play an important role in defense mechanisms against free radical damage, mainly through Zn/Cu superoxide dismutase. A high Cu dose can induce hepatic and renal lesions and overexposure to Zn. Zn may cause vomiting, nausea, and stomach cramps. They are also associated with hematotoxicity, pancreatic and adrenal abnormalities, and impaired immune function (Charles et al., 2018; Lin et al., 2017). Recent studies have delineated the threshold for the effects of copper in drinking water on the gastrointestinal tract. However, there is still some uncertainty regarding the long-term effects of copper on sensitive populations, such as carriers of the gene for Wilson's disease and other metabolic disorders of copper homeostasis (World Health Organization, 2017).

Iron as free iron can penetrate the heart, liver, and brain cells. Due to the disruption of oxidative phosphorylation by free iron, the ferrous iron is converted to ferric iron that releases hydrogen ions, thus increasing metabolic acidity. Free iron

can also lead to lipid peroxidation, severely damaging mitochondria, microsomes, and other cellular organelles (Jaishankar et al., 2014).

2.2. Heavy metal contamination in the river

Data on the dissolved concentration of heavy metal species (Cd, Co, Cu, Ca, Mn, Mo, Ni, Pb, Hg, As, Fe, and Zn) in typical rivers; Bangshi River, Turag River (Bangladesh), Tama River (Japan), Aras River (Iran), and Shenjia River, Wei River (China) showed that some heavy metals are higher than WHO drinking water guideline.

Sakata et al. (2010) studied heavy metal concentrations in the Tama River and the sources of the heavy metals. The experiments compared the measured heavy metal concentration and water flow rates from published data and the measured heavy metal concentration of each source. Results showed that Cd, Cu, Mn, Ni, and Pb concentrations under high flow rate conditions were higher than the WHO reference dose. The heavy metal concentration was strongly affected by the discharge of treated water from sewage treatment plants located along the Tama River catchment.

Nasehi et al. (2012) studied heavy metal concentration in the Aras River. The experiments measured heavy metal concentration and analyzed the data using cluster analysis. Results showed that some stations' Cu, Fe, Zn, Ni, Cd, and Pb concentrations were higher than the WHO reference dose. The season and temperature affect the density of some heavy metal concentrations. Ni, Pb, and Fe density were high in the rainy season, and sources were soil erosion and river bed dissolution. In spring, the density of Cu and other fungicides was high due to river water reduction. However, in winter, the pollution load was reduced because the decrease in the temperature also decreased metal dissolving in water.

Rahman et al. (2013) studied ecological risk assessment of heavy metal contamination in sediment and water body in the Bangshi River. The experiments measured water parameters and heavy metal concentration. Results showed that The Pb concentration was higher than the WHO reference dose, which may be due to the flushing of the metal from rather immobilized deposits like domestic and industrial sludge in the rainy season. The Cd, Ni, Cr, As, and Mn concentration was also higher

than the WHO reference dose. The source of these heavy metals was wastewater discharge from various industrial processes. In contrast, Zn concentration was higher than the value in some seasons.

Yang et al. (2015) studied Heavy metal pollution and health risk assessment in the Wei River. The experiment measured heavy metal concentration and evaluated the data with heavy metal pollution index (HPI) and Health risk assessment. Results showed that during 2008-2012, the heavy metal pollution index (HPI) of Hg, Cd, Cr (VI), Pb, and As in the Wei river fluctuated wildly in 2008 and then declined gradually with time. This general reduction trend in HPI appears from the continued improvement in heavy metal pollution control strategies. However, a health risk assessment of As was higher than an acceptable value, indicating potentially adverse health risks for the local population.

Arefin et al. (2016) studied heavy metal contamination in surface water used for irrigation in the Turag River. The experiment measured heavy metal concentration and some water parameters. Results showed that Fe, Cu, Mn, Cr, and Pb concentrations were higher than the WHO reference dose in some samples. The heavy metal concentrations were strongly affected by the discharge of treated water from the pharmaceutical, tannery, dyeing, and textile industries.

Wang et al. (2017) studied heavy metals' spatial distribution, sources, and ecological risk assessment in the Shenjia River watershed. The experiment measured heavy metal concentration and used ARCGIS 9.3 software to generate heavy metal concentration spatial distribution maps. Results showed that the spatial distribution pattern of As, Cd, Cr, Cu, Ni, Pb, and Zn maps and ecological risk index map were compared with land use map to describe the sources of Cd and other heavy metals, which were more significant than the threshold. The heavy metal concentrations were strongly affected by industrial and domestic sources, fertilizers, and traffic sources.

2.3 Chao Phraya River basin

The Chao Phraya River basin has a mouth river bound to the northern part of the Gulf of Thailand that is important in terms of ecosystem, agriculture, water

consumption, industrial, tourism, and such. The abundance of the Chao Phraya River basin is the primary variable for agriculture alongside the Chao Phraya River, including Thailand's economy. The water resources projects have been developed for water use for over 100-years (Mekapreuksawong & Nakeesin, 2018; Wongsa, 2021).

The Chao Phraya River basin, the largest basin in Thailand with an area of 20,523.42 km², is located in the center of the northern part of the country. It consists of Nakhon Sawan, Phichit, Kamphaeng Phet, Uttaradit, Chainat, Singburi, Lopburi, Ang Thong, Suphanburi, Nakhon Pathom, Nakhon Nayok, Ayuttaya, Saraburi, Pathum Thani, Nontaburi, Samut Prakarn Province and Bangkok. The Chao Phraya River basin has Chao Phraya River as the main river. Coordinates at 13°30'- 16°05'N and 99°30'- 101°00'E (Hydro-Infomatics Institute, 2012) (Figure 1).

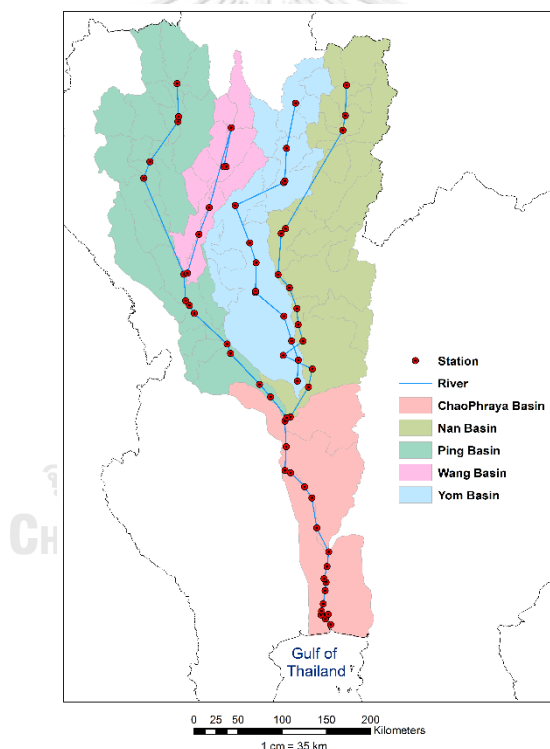


Figure 1 The Chao Phraya River Basin

2.3.1. Chao Phraya River

The Chao Phraya River is the largest river located in the northern and central parts of Thailand. Coordinates at 13°N 100°E. It accounts for about half of the river flow to the Gulf of Thailand. The river and estuary are the main sea artery to Bangkok and are influenced by domestic and industrial activities before interacting

with the gulf of Thailand, a shallow arm of the South China Sea. Agricultural and marine cultural activities in the basin also contribute to nutrient loading. The river dramatically influences the population in Thailand's Central Plain for agriculture, industries, and livelihood (Stansfield & Garrett, 1997). The headwaters of the Chao Phraya River consist of four large tributaries: the Ping, Wang, Yom, and Nan Rivers. The four tributaries flow southward to meet at Nakhon Sawan Province and form the Chao Phraya River (Sharma & Babel, 2013) (Figure 1).

2.3.2. Ping River

The Ping River passes Kamphaeng Phet Province, confluence with Nan River and became the Chao Phraya River. The Ping River is 658 km-long and coordinates at $19^{\circ}48'45''\text{N}$ $98^{\circ}50'20''\text{E}$. It originates in Chiang Mai Province and flows through Lamphun Province, then confluence with Wang River at Tak Province. The Bhumibol Dam was built on the Ping River with a maximum storage capacity of about 13.4 billion m^3 (Figure 2).

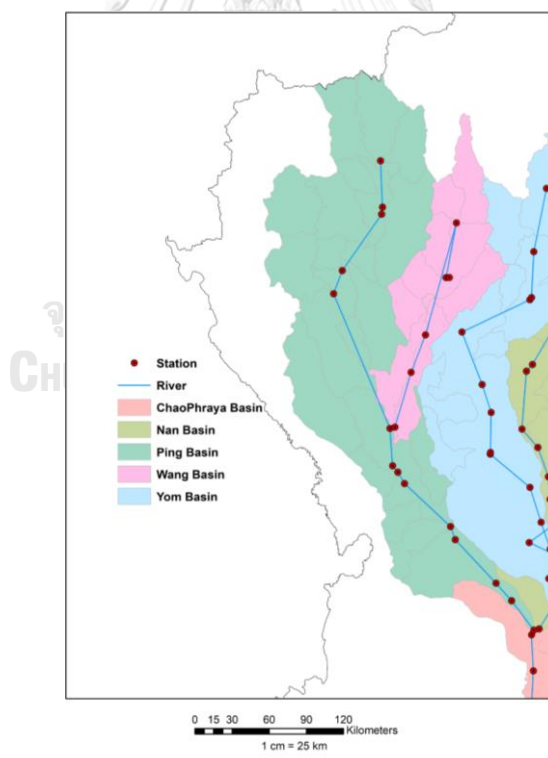


Figure 2 The Ping River Basin and Wang River Basin

2.3.3 Wang River

The Wang River is 392 km-long. Its source is in Chiang Rai Province. The river flows southwards, passing Lampang into Tak Province, then joins the Ping River. Coordinates at 17°7'23"N 99°3'37"E (Figure 2).

2.3.4 Nan River

The Nan River joins the Yom River in Nakhon Sawan Province. Its confluence with the Yom River become the Chao Phraya River. Nan River originates in Nan Province. The Provinces along the River after Nan Province are Uttaradit, Phitsanulok, and Phichit. Nan River is 740 km-long and coordinates at 19°20'0"N 101°12'0"E. The Sirikit Dam was built on the Nan River with a maximum storage capacity of about 9.5 billion m³ (Figure 3).

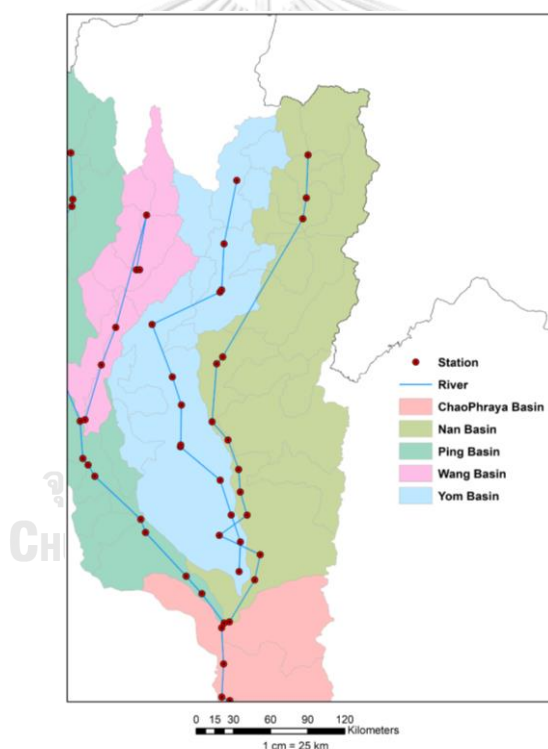


Figure 3 The Nan River Basin and Yom River Basin

2.3.5 Yom River

The Yom River is 787 km-long. Its source in Phayao Province flows through Phrae and Sukhothai as the primary water resource of both Provinces before it joins the Nan River. Coordinates at 19°23'24"N 100°27'18"E (Figure 3).

2.4. Health risk assessment of heavy metal

A human health risk assessment is estimating the nature and probability of adverse health effects in humans who may be exposed to chemicals in contaminated environmental media, now or in the future (USEPA, 2019).

2.4.1 Risk assessment in this study includes

- (1) Hazard identification; heavy metals.
- (2) Dose-response assessment.
- (3) Exposure assessment; Exposure of the human body to heavy metal occurs typically via three main pathways, ingestion, inhalation, and dermal. However, ingestion and dermal are considered human health risks from heavy metals in water environments.
- (4) Risk characterization; data analysis.

According to the International Agency for Research on Cancer (IARC), the Integrated Risk Information System (IRIS) database of EPA, and the classification system of the World Health Organization (WHO), heavy metals are grouped by their carcinogenicity to humans.

Table 1 Group of heavy metal, reference dose (RfD), WHO permissible value, and desirable value.

Heavy metal	Group	RfD ingestion ($\mu\text{g}/\text{kg}/\text{day}$)	RfD dermal ($\mu\text{g}/\text{kg}/\text{day}$)	WHO permissible value* (mg/L)	WHO desirable value* (mg/L)
As	1	0.3	0.285	1.0×10^{-2}	1.0×10^{-2}
Cd	1	0.5	0.025	3.0×10^{-3}	2.0×10^{-3}
Cr	1	3	0.075	5.0×10^{-2}	1.5×10^{-3}
Ni	2B	20	0.8	7.0×10^{-2}	2.0×10^{-2}
Pb	2B	1.4	0.42	1.0×10^{-2}	0
Hg	3	3×10^{-4}	0.0285	6.0×10^{-3}	1.0×10^{-3}
Mn	3	24	0.96	4.0×10^{-1}	1.0×10^{-1}

Cu	3	40	8	2.0	1.0
Zn	3	300	60	3.0	3.0
Fe	3	0.7	1.1×10^{-4}	2.0	3.0×10^{-1}

Group 1 is carcinogenic to humans, Group 2B is possibly carcinogenic to humans, and Group 3 is not classifiable as to its carcinogenicity to humans (USEPA, 2019).

*(World Health Organization, 2017).

2.5. Heavy metal Pollution Index (HPI)

HPI signifies water quality concerning heavy metals, based on the unit weighted arithmetic mean technique. In this index, the unit weightage of each heavy metal is assigned based on its relative significance in influencing water quality for human consumption, which is derived from making values inversely proportional to standard values for the corresponding metal to rule out any discrepancy. In the present study, HPI is calculated with the help of the following equations (Cengiz et al., 2017; Horton, 1965; Patel et al., 2018; Prasad & Bose, 2001):

$$Q_i = \sum_{i=1}^n \frac{[M_i(-)I_i]}{S_i - I_i} \times 100,$$

$$HPI = \frac{\sum_{i=1}^n W_i Q_i}{\sum_{i=1}^n W_i}$$

where M_i = measured value of i th heavy metal, I_i = ideal value (WHO desirable value), S_i = standard value or permissible limit (WHO, 2017), Q_i = sub index, W_i = unit weightage ($W_i = 1/S_i$) (Milivojevic et al., 2016), and n = number of metals used in calculation. (-) = numerical difference of two values, ignoring algebraic sign if any.

The use of different metals' desired and permissible limit values has been employed here to understand the extent of contamination level in river water. The value of $HPI \leq 100$ is safe for drinking water for human consumption, and $HPI > 100$ is unsafe for potable use.

2.6 Human health risks of heavy metals

Because of heavy metal pollution, the current international human health risk assessment model is divided into the carcinogenic chemical risk assessment model and the non-carcinogenic chemical risk assessment model.

The model of health risk from the chemical carcinogens was as follows (Wang et al., 2017; Wang et al., 2019):

$$Rc = \sum_{i=1}^k Rc_i$$

$$Rc_i = [1 - \exp(-D_i \cdot Q_i)]/70$$

Where Rc = health risks from chemical carcinogens, Rc_i = average annual cancer risk for an individual through drinking water channels of chemical carcinogen, D_i (mg/(kg•d)) = the personal exposure dose of 1 U/kg every day through drinking water channels of chemical carcinogens, Q_i (mg/(kg•d)) = strength coefficient of carcinogenic effect through drinking water channels of chemical carcinogen, and 70 was the average life span for human. D_i can be performed as follows:

$$D_i = 2.2C_i/60$$

where 2.2 L= amount of water intake of an adult, C_i = concentration of chemical carcinogens, and 60 kg was the average weight of an adult in Thailand (Wongsasuluk et al., 2018).

The model of health risk from the chemical non-carcinogens was as follows:

$$Rn = \sum_{i=1}^k Rn_i$$

$$Rn_i = (D_i \times 10^{-6}/RfD_i)/70$$

where Rn = the health risks from chemical non-carcinogens, Rn_i = average annual cancer risk for an individual through drinking water channels of chemical non-carcinogens, RfD_i (mg/(kg•d)) = reference dose through drinking water channels of chemical carcinogen, and 70 was the average life span for human. We assumed that all poisonous materials' harmful actions on human health were dependent.

Therefore, the total health risk (R_s) in the water environment was calculated according to the following equation:

$$R_s = R_c + R_n$$

Humans are considered exposed to no health risks if the value of $R_s < 10^{-6}$. The health risk is considered not obvious if $10^{-4} > R_s > 10^{-6}$ and very high if $R_s > 10^{-4}$. As health risk from cancer risk and irradiation are similar, the International Commission on Radiological Protection (ICRP) recommended that the acceptable health risk level for humans is 5×10^{-5} (Wang et al., 2019).



CHAPTER 3

METHODOLOGY

3.1 Sites Information

The observed data of water quality and heavy metal concentration over ten years (2009-2018) monitored by the Pollution Control Department, Ministry of Natural Resource and Environment were used in this study. Each year contained four-quarter data (January, May, July, and November) of 5 rivers (Ping, Wang, Yom, Nan, and Chao Phraya River) from 65 stations (Chao Phraya 18 stations, Nan 14 stations, Ping 14 stations, Yom 13 stations, Wang 6 stations) (Figure 1).

3.2 Water quality analysis

The data was verified for an error for further use. Pearson correlation analysis in the SPSS software for window Version 22 was used to determine correlations between each water quality parameter (pH, tur, con, sal, DO, BOD, TP, NH₃, SS, TDS) and each heavy metal species (Fe, Cd, Cr, Mn, Ni, Pb, Zn, Cu, Hg, and As)

3.3 Health risk assessment of heavy metal

3.3.1. Heavy metal Pollution Index (HPI)

$$Q_i = \sum_{i=1}^n \frac{[M_i(-)I_i]}{S_i - I_i} \times 100,$$
$$HPI = \frac{\sum_{i=1}^n W_i Q_i}{\sum_{i=1}^n W_i} \quad (1)$$

Equation (1) was used to calculate the HPI of each heavy metal species. The value of HPI ≤ 100 is safe for consumed water for human consumption, and HPI > 100 is unsafe for potable use.

3.3.2 Health risks of heavy metals

$$Rc = \sum_{i=1}^k Rc_i$$
$$Rc_i = [1 - \exp(-D_i \cdot Q_i)]/70 \quad (2)$$

$$D_i = 2.2C_i/60 \quad (3)$$

Equations (2) and (3) were used to calculate the health risks of carcinogenic heavy metals.

$$Rn = \sum_{i=1}^k Rn_i$$

$$Rn_i = (D_i \times 10^{-6}/RfD_i)/70 \quad (4)$$

Equations (3) and (4) were used to calculate the health risks of non-carcinogenic heavy metals.

$$Rs = Rc + Rn \quad (5)$$

Equation (5) was used to calculate the total Health risk in the water environment. Humans are considered exposed to no health risks if the value of $R_s < 10^{-6}$. The health risk is considered not obvious if $10^{-4} > R_s > 10^{-6}$ and very high if $R_s > 10^{-4}$. Recommended the acceptable health risk level for humans is 5×10^{-5} .

Each heavy metal species' human health risk assessment was applied to ArcMap 10.4.1 to visualize maps.

3.4 Visualize Map

3.4.1 Prepare layer

The Thailand map data (supported by Associate Professor Dr. Pasicha Chaikaew) would be set to the Display area in ArcMap 10.4.1 and managed to be WGS_1984_UTM_Zone_47N polygon-based vector format, then selected each of the basin areas that we wanted to focus and exported data output to be shapefile for further use.

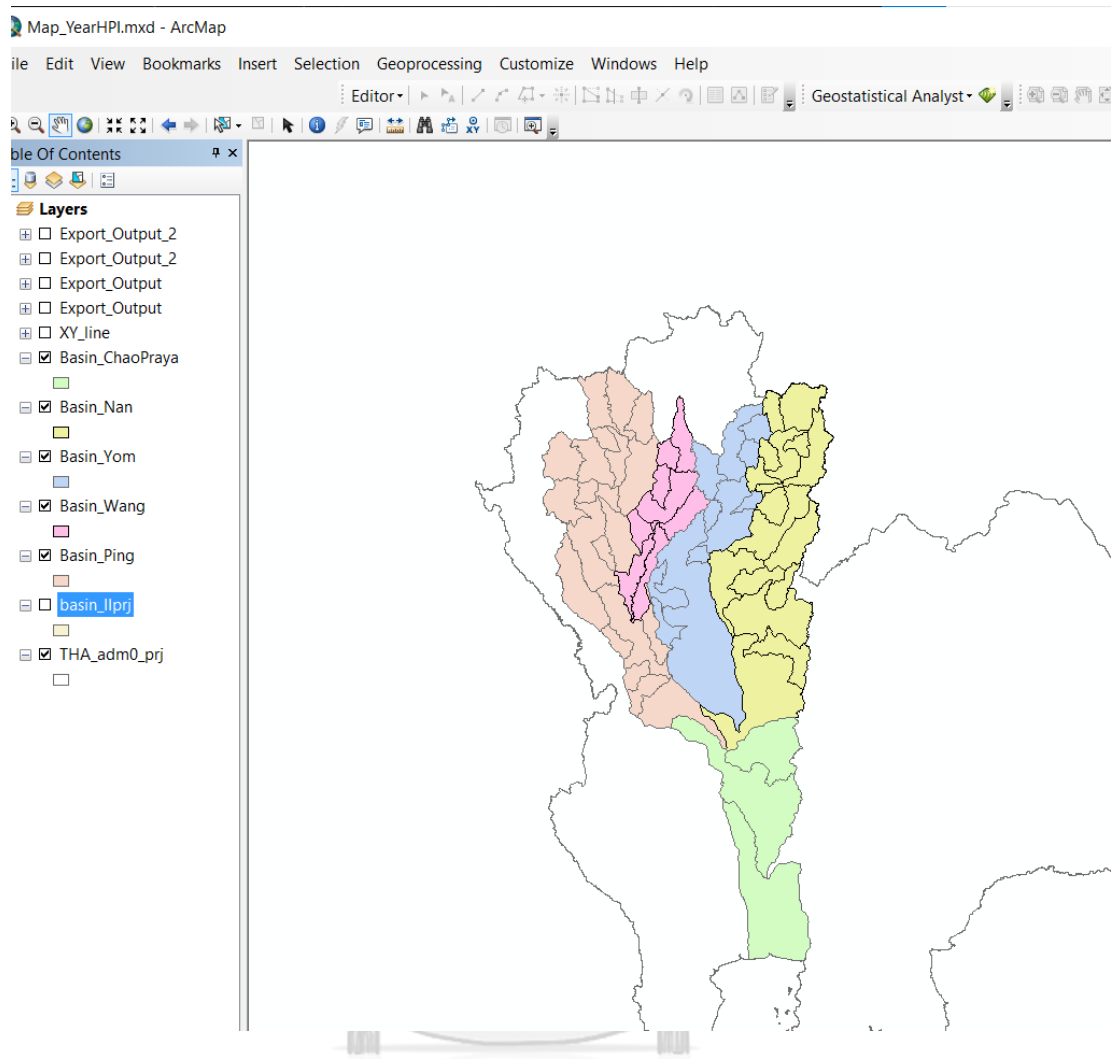


Figure 4 Polygon data of Chao Phraya River Basin

3.4.2 Stations plotting

The calculated data were used as map sources by plotting the HPI and Health Risks data with coordinates as the point on the prepared shapefile. Allocation of the weight of data according to the expert opinions (Safe, and Unsafe for HPI, and no health risks, considered not obvious, acceptable health risk level to humans, and very high for Health Risks).

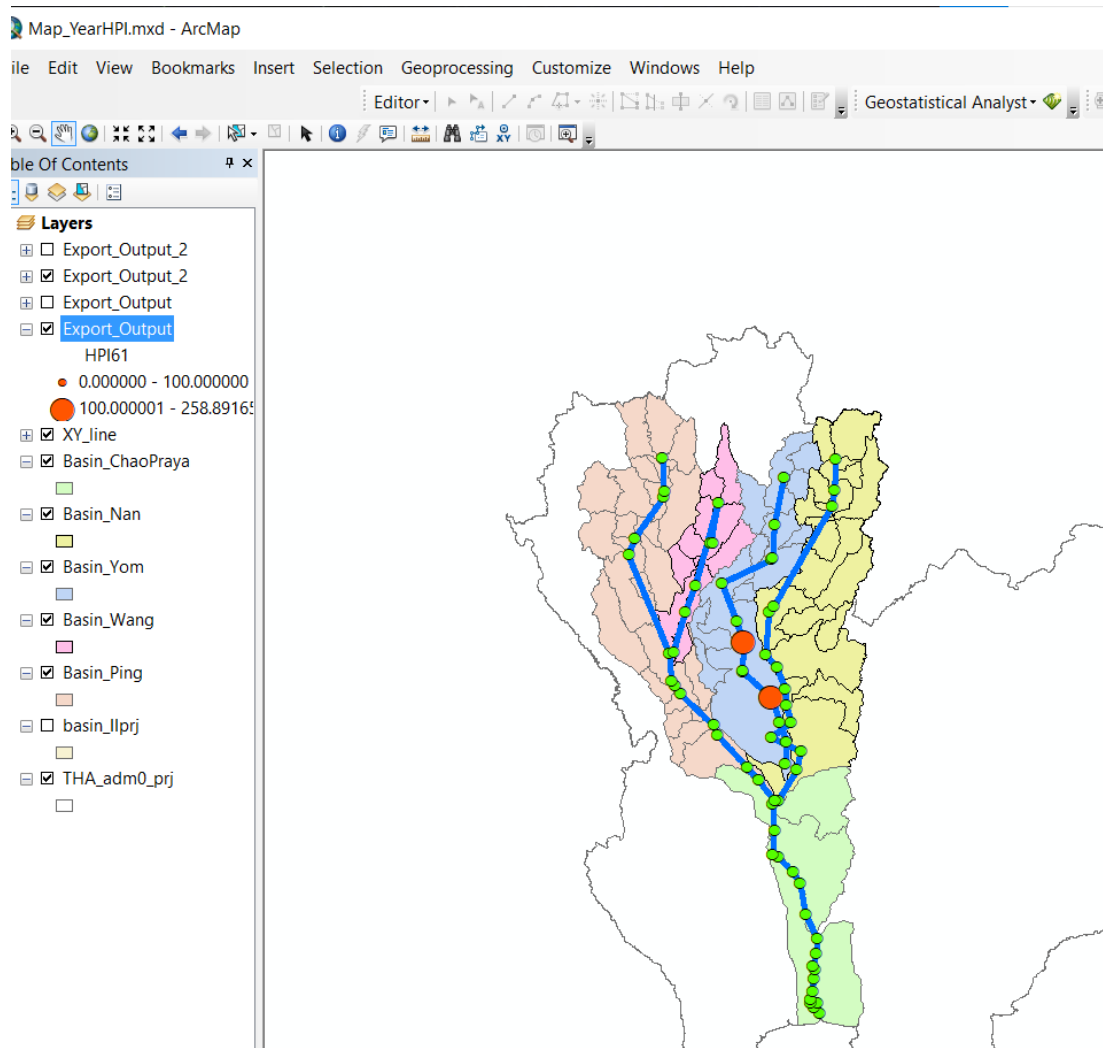


Figure 5 Plotted map

3.4.3 Land use

Land use maps and data supported by the Land Development Department, Ministry of Agriculture and Cooperatives during 2009, 2010-2013, 2015-2016, and 2017-2018. The category type was simplified to 6 categories (Agricultural land, Aquacultural land, Forest land, Miscellaneous land, Water body, and Urban and built-up land)

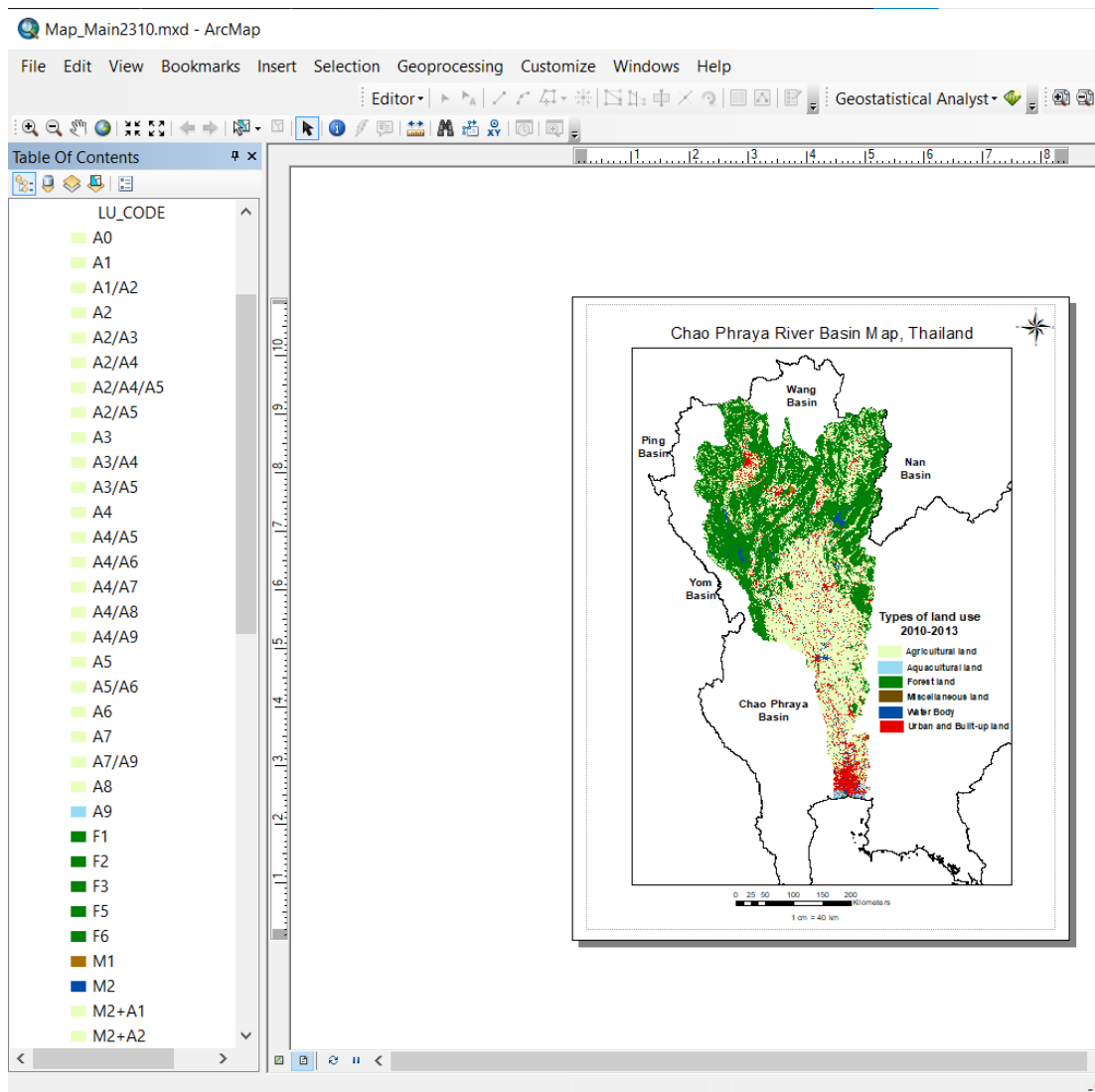


Figure 6 Simplified land use map

3.5 Changed area

The land use data were compared between 4 periods (2009, 2010-2013, 2015-2016, and 2017-2018) and calculated the changing area.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Relationship between heavy metal concentration and water parameter in Chao Phraya River

The concentration of heavy metals in a river depends on many factors, including pH, temperature, and turbidity. In this study, 2-tailed Pearson correlation was applied to analyze the relationship between heavy metal concentration and water parameters in Chao Phraya River during two periods: 2009-2013 and 2014-2018. The results showed that during 2009-2013, Fe concentration positively correlated to turbidity (0.640), total phosphate (0.622), suspended solids (0.542), and Ni level (0.513) in the river but negatively correlated to Zn level (-0.517) at $P < 0.01$ (Figure 7).

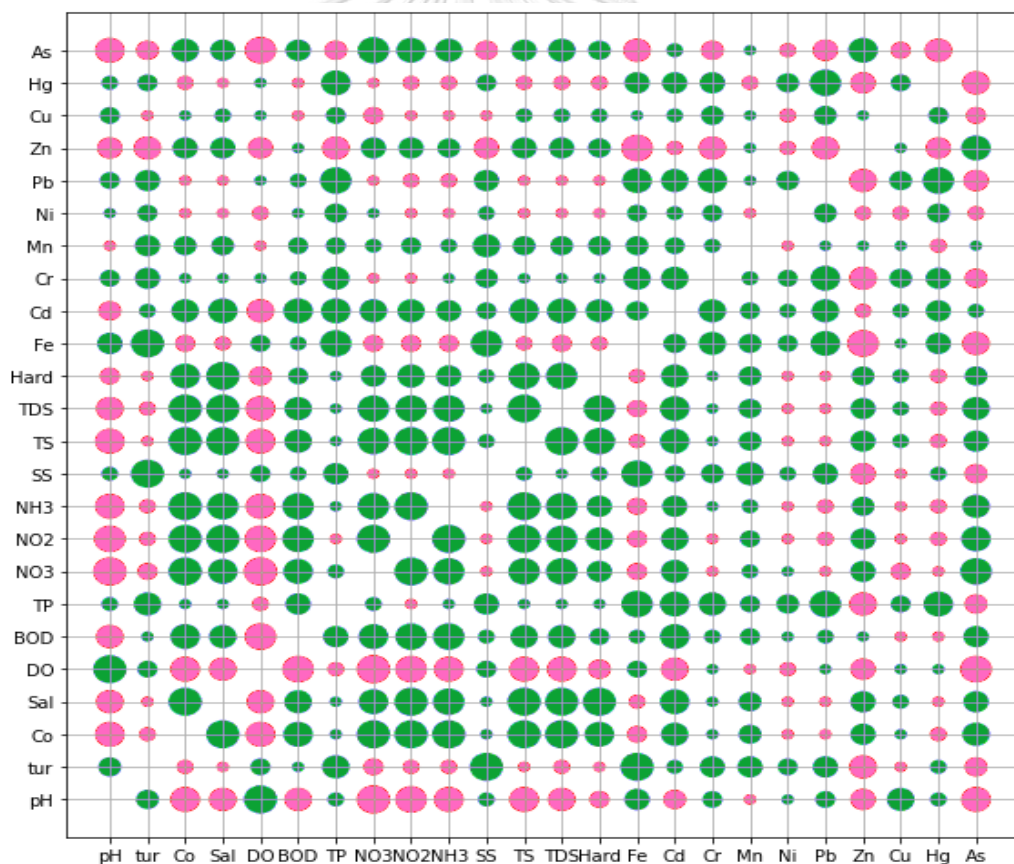


Figure 7 Correlation between heavy metal concentration and water parameters in the Chao Phraya River during 2009-2013

From 2014 to 2018, the results revealed that Fe concentration positively correlated to turbidity (0.900), SS (0.671), Mn (0.607), and Ni level (0.512) at $P < 0.01$

(Figure 8). The experiment results on the river Etsu by (Garg R. K., 2010) also showed high turbidity during the rainy season. During the rainy season, silt, clay, and other suspended particles contribute to the turbidity values. Several mineral deposits have been found in Thailand. Water erosion may be caused by the dissolution of some heavy metals in the river.

Cd contamination was also positively correlated to Cr level (0.509). Cd is a common impurity in phosphate fertilizer, with the increasing use of fertilizers in agriculture. The other sources include landfill leachates from Ni-Cd-based battery dumps in urban and rural communities (Silas II & AU, 2018).

Since some of the parameters had missing data or measured to be very less, some of the correlation might be a false positive or false negative and made the correlation compared between two periods unstable and inaccurate in some parameters.

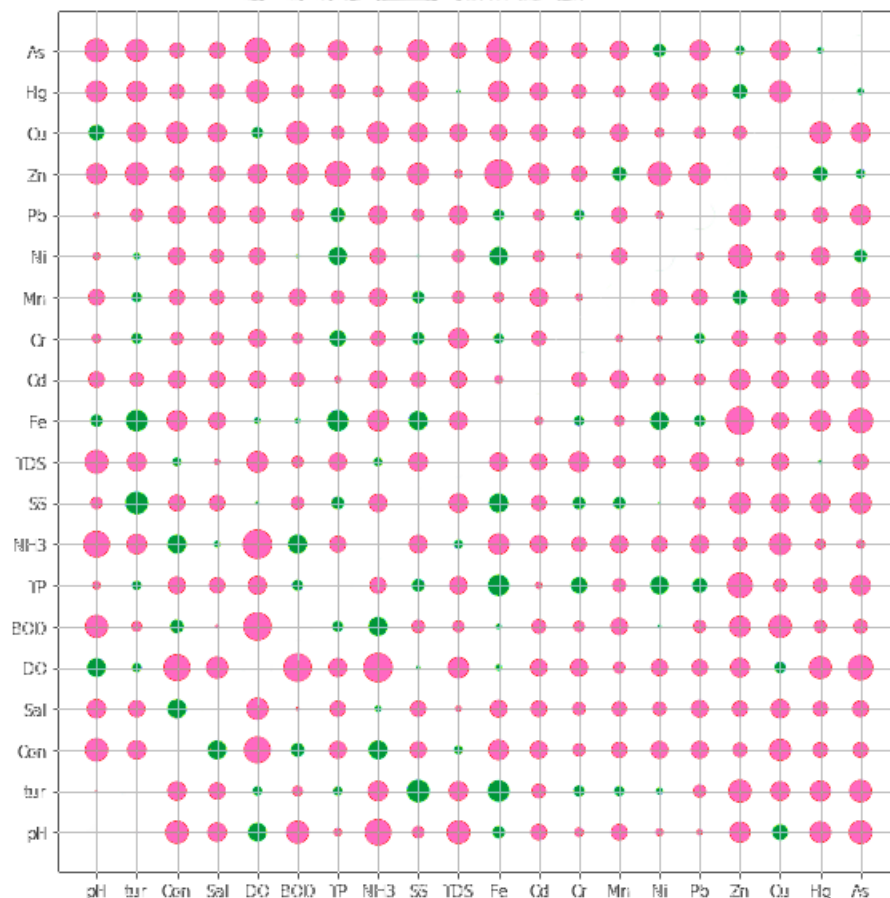


Figure 8 Correlation between heavy metal concentration and water parameters in the Chao Phraya River during 2014-2018

4.2 Heavy metal pollution index in Chao Phraya River Basin during 2009-2018

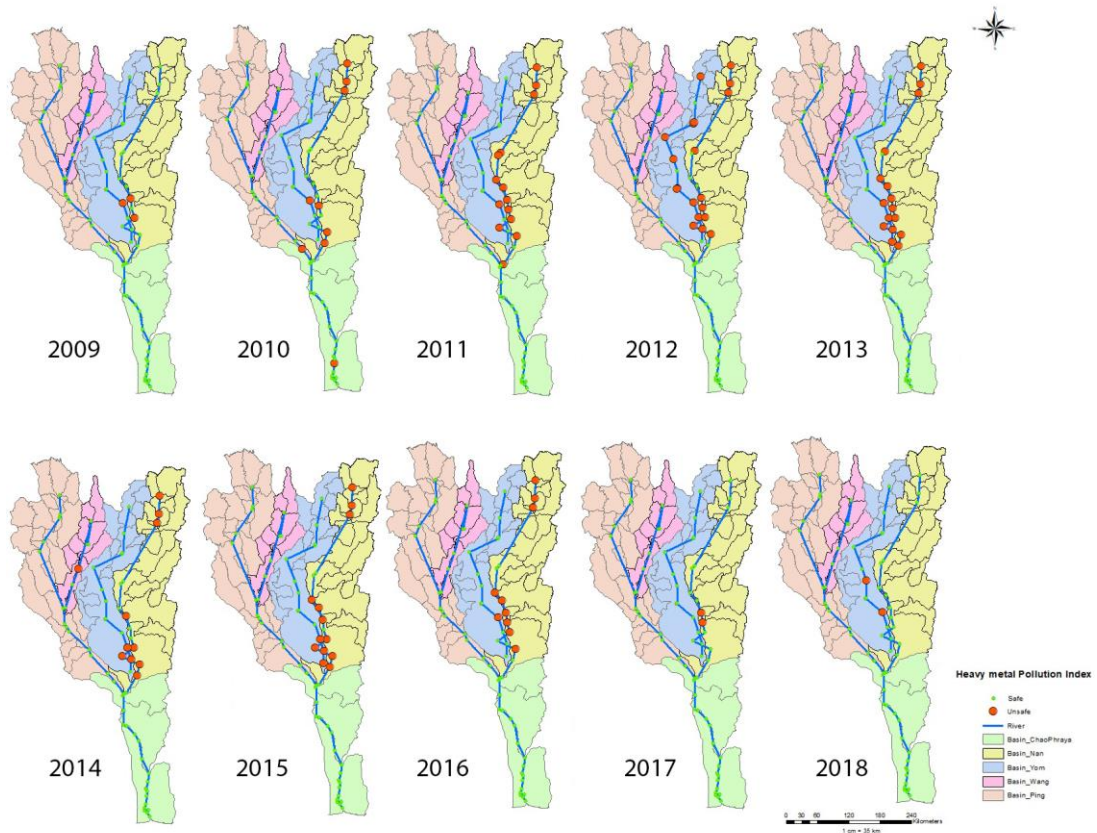


Figure 9 HPI of Chao Phraya River Basin from 2009 to 2018

This study classified the heavy metal pollution index into two categories. HPI below 100 assumes as safe or acceptable for consumed water. However, if HPI was more significant than 100, the water was unacceptable for consumed water. HPI was calculated separately for each station of each year.

HPI values of all stations during ten years ranged from 15.456 to 1084.832, with an average value of 71.382.

From the calculation, the HPI value depended on the concentration of Cd, Cr, Ni, and Pb even though these heavy metals had low concentrations but caused the high values in HPI calculations.

Table 2 The water sampling station

Station	River	Area	Amphoe	Province
CH01	Chao Phraya	Phra Samut Chedi	Meuang	Samut Prakan
CH03	Chao Phraya	District office	Phra Pradaeng	Samut Prakan
CH06	Chao Phraya	Bangkok Port	Yan Nawa	Bangkok
CH08	Chao Phraya	Krung Thep Bridge	-	Bangkok
CH10	Chao Phraya	Phra Phuttha Yodfa Bridge	Samphanthawong	Bangkok
CH12	Chao Phraya	Rama VI Bridge	Bang Kruai	Nonthaburi
CH15	Chao Phraya	Nonthaburi Bridge	Meuang	Nonthaburi
CH16.1	Chao Phraya	Samlae Raw water pump station	Meuang	Pathum Thani
CH17	Chao Phraya	Bridge	Sam Khok	Pathum Thani
CH18	Chao Phraya	Bang Pa-in Paper mill industry	Bang Pa-in	Ayutthaya
CH20	Chao Phraya	Pomphet	Phra Nakhon Si Ayutthaya	Ayutthaya
CH21	Chao Phraya	Bridge over the Chao Phraya river	Meuang	Ang Thong
CH24	Chao Phraya	Bridge over the Chao Phraya river	Meuang	Sing Buri
CH25	Chao Phraya	Under the market area	In Buri	Sing Buri
CH27	Chao Phraya	Chao Phraya Dam	Sappaya	Chai Nat
CH28	Chao Phraya	Town hall	Meuang	Chai Nat
CH30	Chao Phraya	Somdet Phra Wannarat Bridge	Phayuha Khiri	Nakhon Sawan
CH32	Chao Phraya	Dechatiwong Bridge	Meuang	Nakhon Sawan
NA01	Nan	Wat Kriangkrai Tai	Meuang	Nakhon Sawan
NA02	Nan	Bridge	Bang Mun Nak	Phichit
NA03	Nan	Ratratransan Bridge	Taphan Hin	Phichit
NA04	Nan	Wat Tha Luang	Meuang	Phichit
NA05	Nan	Phichit-Noen Maprang Bridge	Meuang	Phichit
NA06	Nan	Wat Sawang Arom Bridge	Meuang	Phitsanulok
NA07	Nan	Wat Phothisyan water pump station	Meuang	Phitsanulok
NA08	Nan	Naresuan Dam	Phrom Phiram	Phitsanulok
NA09	Nan	Pichai Bridge	Phichai	Uttaradit
NA10	Nan	North Development 13 Bridge	Meuang	Uttaradit
NA11	Nan	Ban Wang Kong Bridge	Meuang	Uttaradit
NA12	Nan	Ban Don Sri Serm	Meuang	Nan
NA13	Nan	Wiang Sa water pumping station	Wiang Sa	Nan
NA14	Nan	Tha Wang Pha water pumping station	Tha Wang Pha	Nan
PI01	Ping	Phitsanulok Bridge	Meuang	Nakhon Sawan
PI02	Ping	Ban Thong Kung	Banphot Phisai	Nakhon Sawan
PI03	Ping	Ban Saen Tor Bridge	Khanu Woraklaksaburi	Nakhon Sawan
PI04	Ping	Ban Wang Yang Bridge	Meuang	Kamphaeng Phet
PI05	Ping	Kamphaeng Phet Bridge	Meuang	Kamphaeng Phet

Station	River	Area	Amphoe	Province
PI06	Ping	Wat Tha Takhro	Meuang	Tak
PI07	Ping	Kittikachorn Bridge	Meuang	Tak
PI08	Ping	Suspension Bridge	Meuang	Tak
PI09	Ping	Bridge	Ban Tak	Tak
PI10	Ping	Kong Hin Hydrological center	Hot	Chiang Mai
PI11	Ping	Nong Pla Swai Bridge	Chomthong	Chiang Mai
PI12	Ping	Provincial Police Station 5 Bridge	Meuang	Chiang Mai
PI13	Ping	Ban Wang Sing Kum Bridge	Meuang	Chiang Mai
PI14	Ping	Cho Lae Bridge	Mae Taeng	Chiang Mai
WA01	Wang	Ban Wang Mun Bridge	Sam Ngao	Tak
WA02	Wang	Thong Sawat Bridge	Thoen	Lampang
WA03	Wang	Sobphrab raw water pumping station	Sop Prap	Lampang
WA06	Wang	Soi River confluence	Chae Hom	Lampang
WA4.1	Wang	Yang weir	Meuang	Lampang
WA5.1	Wang	Setuwaree Bridge	Meuang	Lampang
YO01	Yom	Pho Thale Bridge	Pho Thale	Phichit
YO02	Yom	Wat Tha Bua Thong	Pho Prathap Chang	Phichit
YO03	Yom	Ban Sam Ngam Bridge	Sam Ngam	Phichit
YO04	Yom	Yom River Bridge	Bang Rakam	Phitsanulok
YO05	Yom	Phra Ruang Bridge	Meuang	Sukhothai
YO06	Yom	Ban Wang Hin Patthana	Meuang	Sukhothai
YO07	Yom	Waterside	Sawankhalok	Sukhothai
YO08	Yom	Si Satchanalai Bridge	Si Satchanalai	Sukhothai
YO09	Yom	Wang Chin Bridge	Wang Chin	Phrae
YO10	Yom	North Development 8 Bridge	Meuang	Phrae
YO11	Yom	Klong Pho Bridge	Meuang	Phrae
YO12	Yom	Mae Yom weir	Song	Phrae
YO13	Yom	Highway 1091 Bridge	Chiang Muan	Phayao

Table 3 The number of stations of each category of HPI

Year	HPI			
	Safe	Unsafe		
	HPI <100	Mild (101-400)	Moderate (401-800)	Severe (801-1200)
2009	62	3	0	0
2010	57	8	1	0
2011	51	14	0	0
2012	47	18	1	0
2013	50	15	1	0
2014	56	9	1	1
2015	52	13	0	0
2016	56	9	1	0
2017	63	2	0	0
2018	63	2	0	0

This study divided the HPI into two categories: safe and unsafe. As the value exceeded the standard value, unsafe was divided into three ranges; mild, moderate, and severe, depending on how high the value was.

The result indicated that the HPI value was lowest at the CH06 station (15.46) in 2018 and highest at the NA14 station (1084.83) in 2014.

HPI value was high (unsafe) around Nan River and Yom River area in Pichit, Pitsanulok, and Sukhothai province, and some station was nearly raw water pumping station. In 2010, the HPI started to increase in Nan, Uttaradit, and Nonthaburi provinces and decreased in 2017 (Table 3).

In Yamuna river, India, also being as one of the major rivers in India facing the same heavy metal problem. The river had a very high HPI above 497.96 that much worse when compared with the HPI of Chao Phraya River Basin in our study (Bhardwaj et al., 2019).

4.3 Human Health risk assessment in Chao Phraya River Basin in 2009-2018

Human Health risk assessment is an essential method for evaluating the magnitude of adverse effects in humans who are exposed to contaminants in water environments. In this research, the health risk assessment from the dermal absorption and ingestion were calculated because they are the significant pathways of exposure to the river. The toxicity to human health is related to their daily intake (Mohammadi et al., 2019).

In this study, the human health risk was divided into four categories; the health risk is considered no health risk if the value of $R_s < 10^{-6}$, and the health risk is considered acceptable health risk level to humans if $10^{-6} < R_s < 5 \times 10^{-5}$, the health risk is considered not obvious if $5 \times 10^{-5} < R_s < 10^{-4}$ and very high if $R_s > 10^{-4}$.

The results showed that the health risk values of all stations were in the range of 7.14×10^{-5} to 7.56×10^{-4} , with an average value of 2.37×10^{-4} .

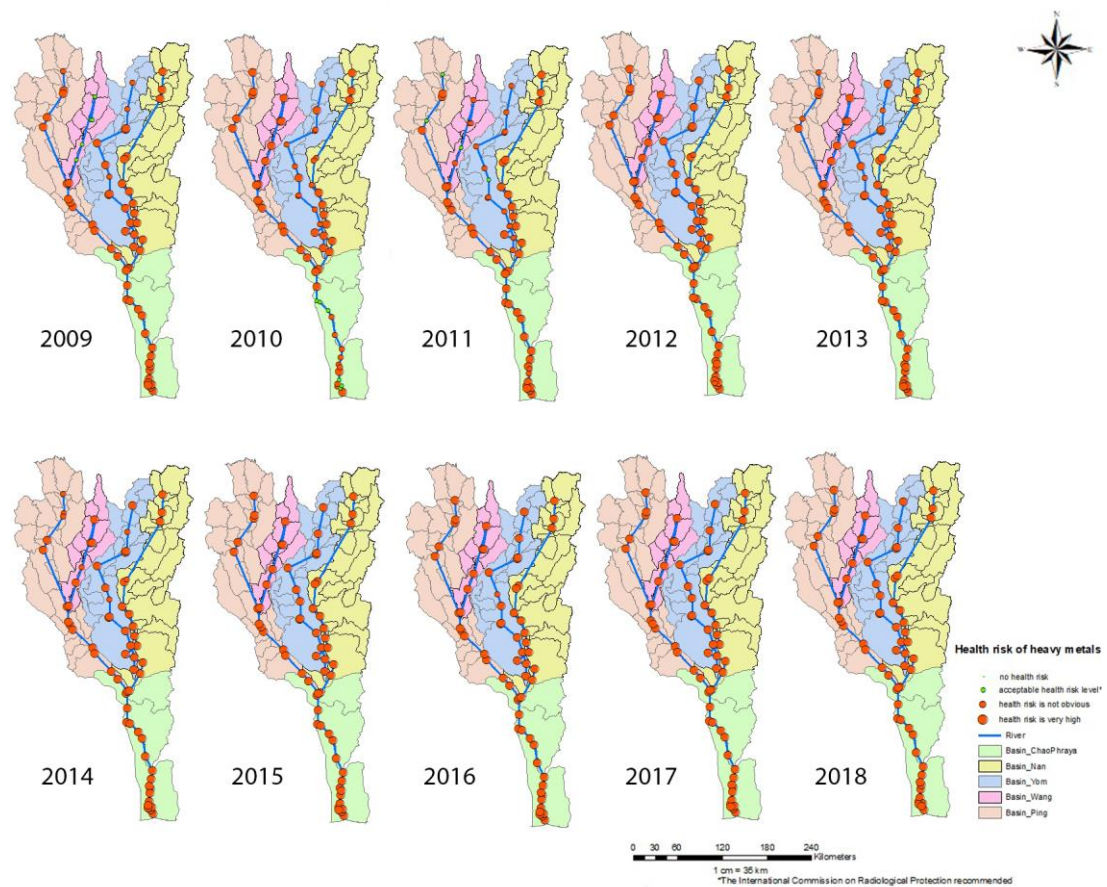


Figure 10 Health risk assessment of Chao Phraya River Basin in 2009-2018

Likewise, health risk assessment uses only heavy metal parameters to calculate the risk. After calculating the health risk assessment for each heavy metal in water at each site, the highest risk in water consumption depended on the high concentration of Fe, Cd, Cr, Ni, and Pb, especially the concentration of Cd and Cr that both almost exceed from the standard value.

Table 4 The number of stations in each category of health risk assessment

Year	health risk assessment			
	no health risk	acceptable health risk	health risk is not obvious	health risk is high
2009	0	5	4	56
2010	0	6	20	39
2011	0	4	22	39
2012	0	0	2	63
2013	0	0	4	61
2014	0	0	7	58
2015	0	0	0	65
2016	0	0	1	64
2017	0	0	0	65
2018	0	0	0	65

This study revealed that from 2009 to 2011, some stations in Lampang, Chiang Mai, and Sukhothai province were at acceptable health risk (Table 4). However, from 2012 to 2018, there was no acceptable health risk categories station. Moreover, in 2015, 2017, and 2018, all stations were classified as very high risk.

4.4 Change of land use in Chao Phraya River Basin during 2009-2018

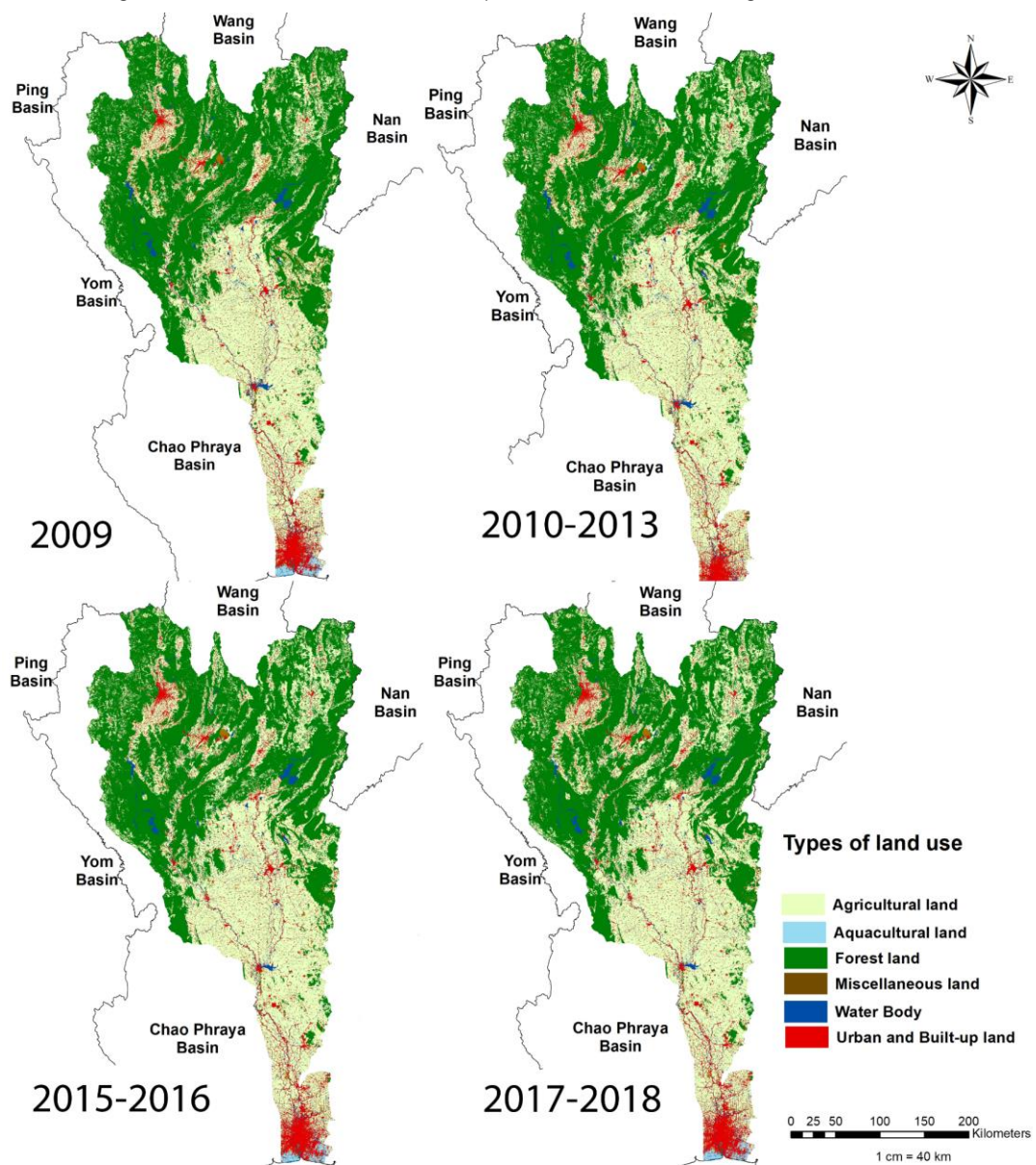


Figure 11 Change of land use in Chao Phraya River Basin during 2009-2018

The results for space-acquired images for Chao Phraya River Basin, a classification was carried out to land cover categories. Six categories were identified: agricultural land, aquacultural land, forest land, miscellaneous land, water body, and urban and built-up land (Figure 11).

In 2009, the forest land, agricultural land, and urban and built-up land were 49.67%, 39.68%, and 5.74%, respectively. In 2018, the land use structure was changed. The results indicated that in 2018, the area of forest land decreased to

45.85%. On the other hand, agricultural land and urban and built-up land were increased to 43.14% and 6.51%, respectively. However, aquacultural land area decreased from 0.61% in 2009 to 0.56% in 2015 and slightly increased to 0.57% in 2018.

The highest rate of deforestation was 2.40% in the 2009-2010 period, also found to have the most significant expansion of agricultural land at 2.27%.

Table 5 Change in area of lands of different categories (km²) in Chao Phraya River basin from 2009 to 2018

Category of Area	Area in Each year (km ²)			
	2009	2010	2015	2018
Agricultural land	49361.41	52185.58	53545.66	53661.14
Aquacultural land	752.860	739.767	692.729	704.537
Forest land	61783.36	58797.69	57318.04	57029.53
Miscellaneous land	2708.766	2441.692	2076.652	1969.559
water body	2647.286	2821.122	2827.473	2935.866
Urban	7145.355	7407.451	7932.720	8092.648
Total	124399.046	124393.302	124393.274	124393.293

Table 6 Total Area in Category of Thailand Area between 2009-2018

Category/Area (km ²)	% Change		
	2009-2010	2010-2015	2015-2018
Agricultural land	2.270	1.093	0.093
Aquacultural land	-0.011	-0.039	0.009
Forest land	-2.400	-1.189	-0.232
Miscellaneous land	-0.215	-0.293	-0.086
water body	0.140	0.005	0.087
Urban	0.211	0.422	0.129

4.4 The relationship between land use and heavy metal contamination

In this study, the Heavy metal Pollution Index (HPI) was applied as the indicator of heavy metal contamination because it was calculated using heavy metal concentrations and unit weightage of heavy metal from typical values. From limited land use data, this study was divided into four periods: 2009, 2010-2013, 2015-2016, and 2017-2018.

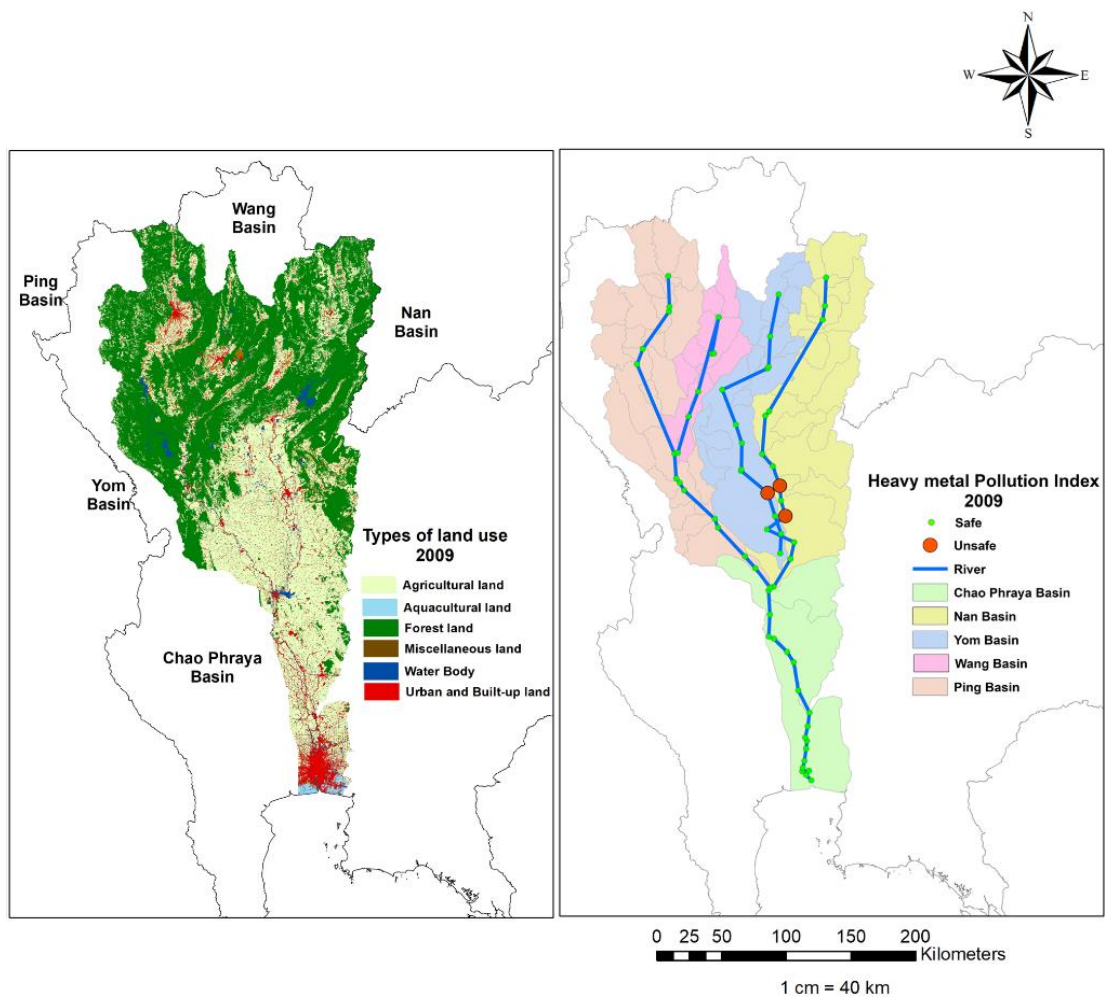


Figure 12 Relationship of HPI and change in land use in 2009

HPI values of all stations were in the range of 27.246 to 356.299, with an average value of 64.871. Since 2000, the lower Yom Basin has gradually changed from mainly forest land to agricultural lands and urban areas due to commercial and urban growth (Chotpantararat & Boonkaewwan, 2018). HPI value in this study also showed that the area is unsafe.

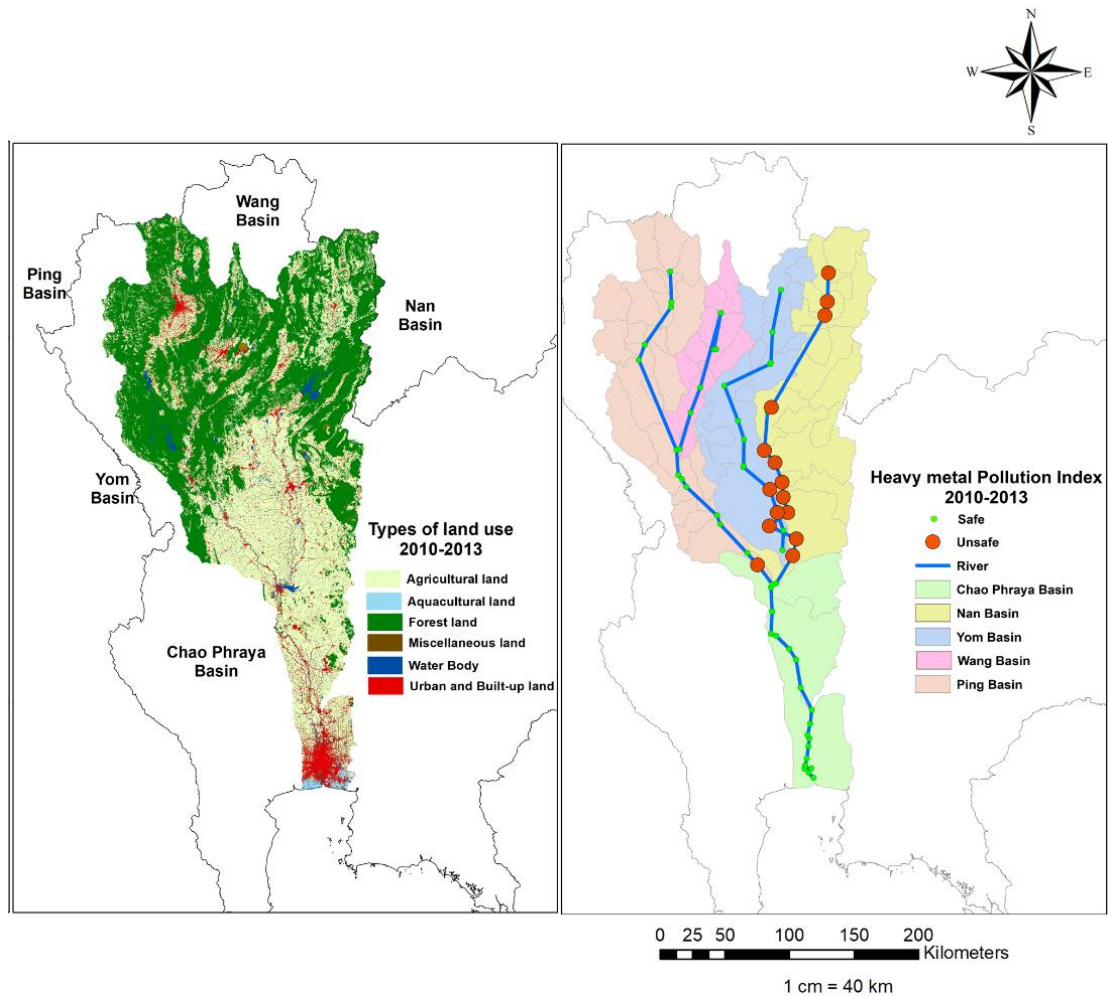


Figure 13 Relationship of HPI and change in land use in 2010-2013

HPI values of all stations were in range from 21.195 to 270.298, with an average value of 81.095. HPI value alongside Nan River was high, and most stations are unsafe to use as consume water.

The Yom Basin is a significant share of Thailand's rice production and export. Increasing the use of fertilizers in agriculture, the Yom Basin becomes the source of increase in total phosphate and other heavy metals. By having no significant structural measures for flow regulation, heavy rainfall received and the inadequate drainage capacity in the basin can cause flooding (Padiyedath Gopalan et al., 2022).

Due to Thailand's catastrophic flooding in 2011, some industrial estates were affected, which might lead to toxic chemicals in the wastewater treatment systems of industrial estates (Mingkhwan & Worakhunpiset, 2018). Also, deforestation and expansion of agricultural land during this period can cause high HPI value.

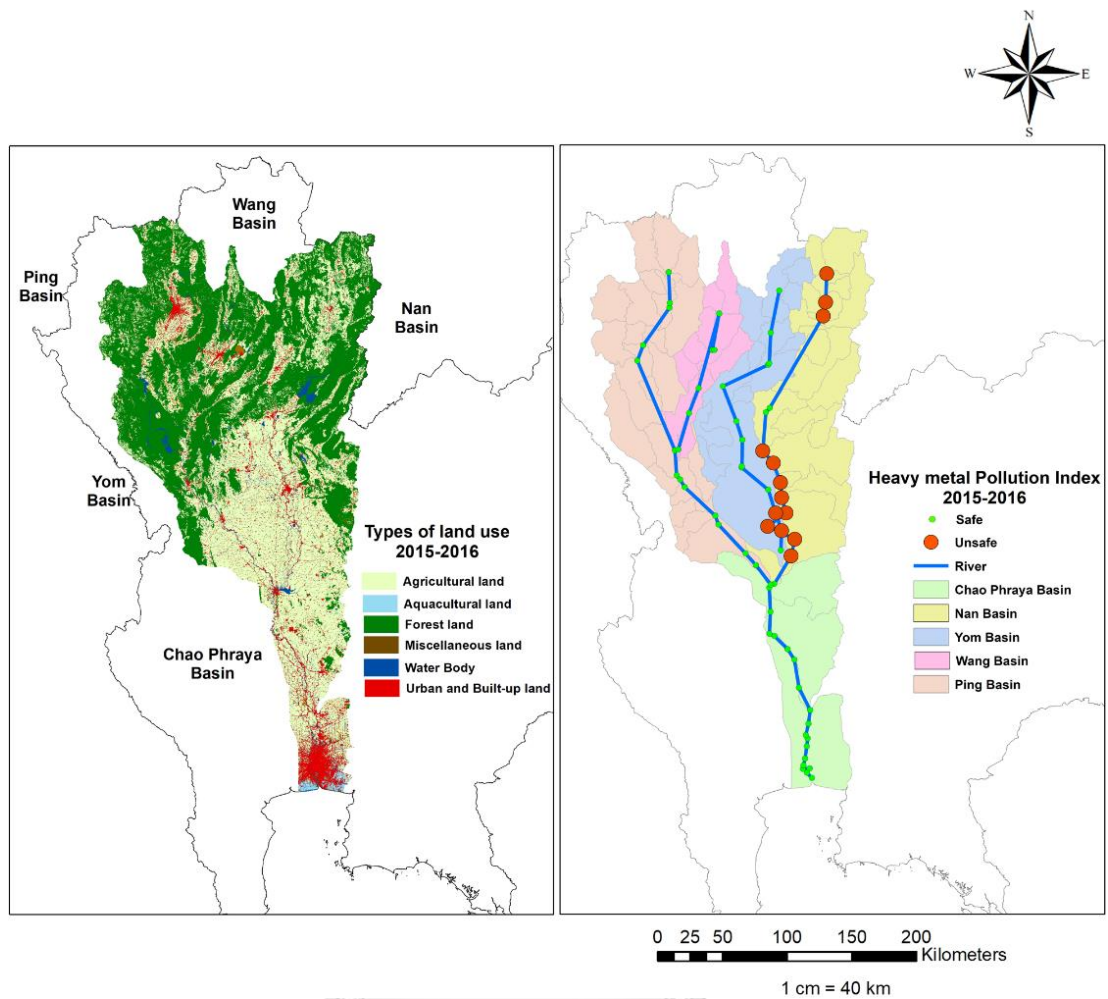


Figure 14 Relationship of HPI and change in land use in 2015-2016

HPI values of all stations were 22.821 to 469.626, with an average value of 72.59279. HPI value alongside Nan River is still high, and most stations are unsafe to use as consumed water.

In 2016, the RAI storm caused flooding and landslide in various areas in the northern and eastern parts of Thailand. Due to a lack of drainage capacity, the Yom Basin and Nan Basin accumulate water from the above area and spread the water out, which can cause a heavy metal leak from agricultural land (Thaiwater, 2016).

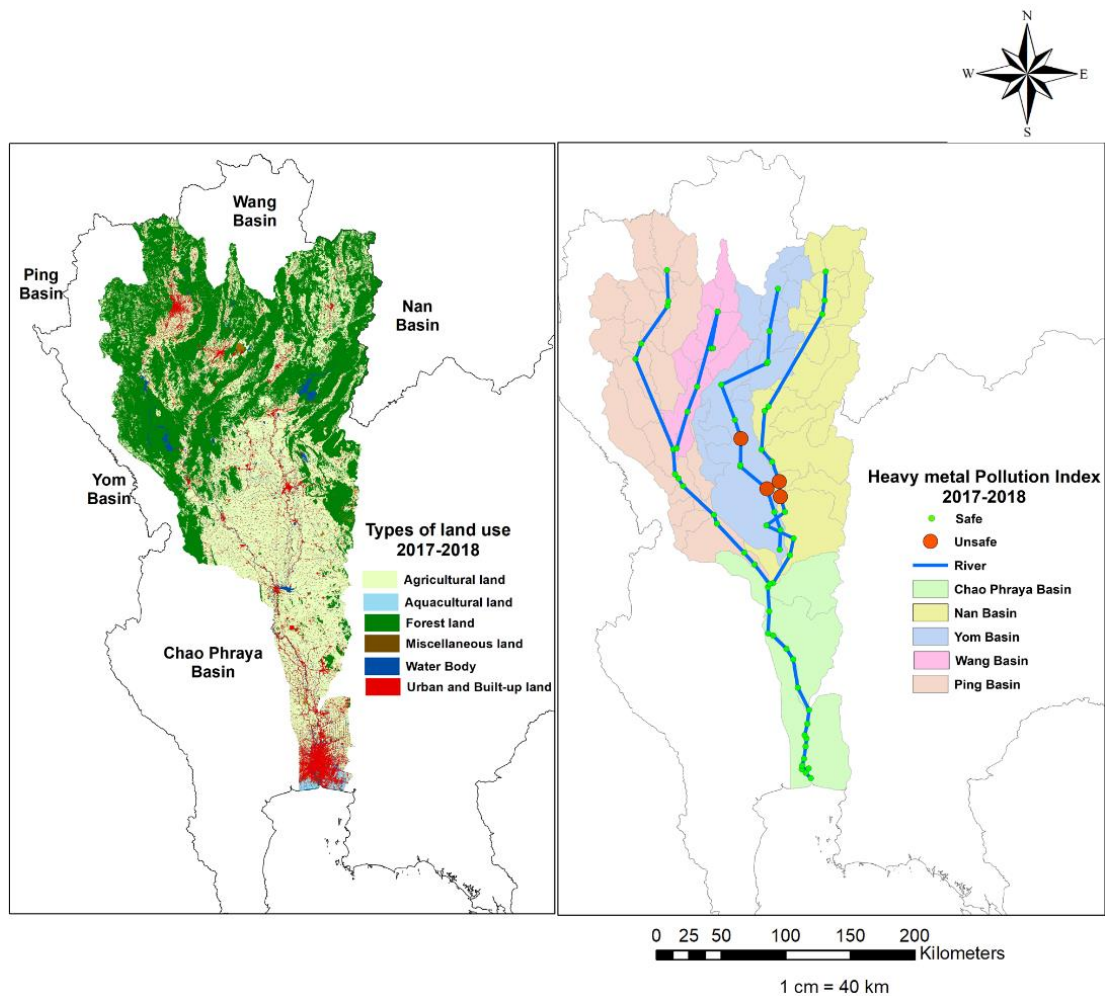


Figure 15 Relationship of HPI and change in land use in 2017-2018

The land use in Nan province was rapidly changed due to deforestation, and plowing mountainous areas for cultivation could increase overload runoff and enhance the hazard of heavy floods during monsoon season. These reasons might cause degraded water quality (Wongtui, 2016).

Although in 2017 and 2018, some stations had better changes in water quality, Pitsanulok province still has an unsafe station. However, the broad areas not in the mentioned area (about 85% of the basin areas) were safe to use as consumed water.

By the way, using surface water as consumed water still needs to consider other water quality parameters, not only heavy metals, and treat the water by a trusted organization.

From Hydro-Informatics Institute, the primary resources of water pollution were urban discharge, agricultural discharge and using chemical fertilizer, and industrial discharge in areas such as Pathumthani, Nonthaburi, and Samut Prakan province (Hydro-Informatics Institute, 2012).

Nateekhuncharoen and Ariyakanon (2021) had provide the relationship of water quality and each kind of heavy metal in Chao Phraya River Basin using multiple linear regression model and calculated Hazard Quotient (HQ) and Hazard Index (HI) of adult and children. Collaborate with this study, it should be indicated better understanding on the water quality and heavy metals in Chao Phraya River Basin.

As the Pollution Control Department (PCD) has been carried out regular water quality monitoring for Thailand's rivers for many years, they should consider this study results and manage plans with other ministries to reduce the risk from water consumption in the high risk areas not only control the agricultural discharge and fertilizer uses but also control other sources of heavy metals.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusions

The result that was found in this study can conclude the important finding as follow;

(1) From 2009 to 2011, some stations in Lampang, Chiang Mai, and Sukhothai province were at acceptable health risk. However, from 2012 to 2018, there was no station in acceptable health risk. Moreover, in 2015, 2017, and 2018, all stations were classified as very high risk.

(2) HPI value was unsafe around Nan River and Yom River area in Pichit, Pitsanulok, and Sukhothai province, and some stations were nearly raw water pumping stations. HPI value started to increase in Nan, Uttaradit, and Nonthaburi provinces in 2010 and decreased in 2017.

(3) From 2009 to 2013, Fe concentration positively correlated to turbidity (0.640), total phosphate (0.622), suspended solids (0.542), and Ni level (0.513) but negatively correlated to Zn level (-0.517) at $P < 0.01$. From 2014 to 2018, Fe concentration positively correlated to turbidity (0.900), suspended solids (0.671), Mn (0.607), and Ni level (0.512) at $P < 0.01$. Cd concentration is also positively correlated to Cr level (0.509).

(4) In 2009, the forest land, agricultural land, and urban and built-up land were 49.67%, 39.68%, and 5.74%, respectively. In 2018, the area of forest land decreased to 45.85%. On the other hand, agricultural land and urban and built-up land were increased to 43.14% and 6.51%, respectively. However, aquacultural land area decreased from 0.61% in 2009 to 0.56% in 2015 and slightly increased to 0.57% in 2018.

(5) HPI value in this study is unsafe in the agricultural land, especially in the Yom River, due to expanded agricultural land and fertilizer use for commercial and urban growth.

5.2 Recommendations for future work

According to the result of this study, the recommendations for further study to improve and more understanding are followed.

- (1) Field data collection can provide more precision and accuracy of the data.
- (2) Background and sediment data should be collected as some heavy metals can be accumulated in the surface sediment.
- (3) The water flow data should be collected as a high or low flow rate can provide more concentration of some heavy metals.
- (4) More models can provide more understanding of the relationship between heavy metal and heavy metal and land use.



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Appendix

Table 7 Each station HPI Value

Station	Year									
	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
CH01	60.531	44.511	44.940	29.961	52.301	41.951	25.234	21.185	20.639	44.943
CH03	51.194	61.349	38.011	36.303	44.647	30.983	32.050	30.441	33.546	16.501
CH06	51.194	61.349	38.011	36.303	44.647	30.983	32.050	30.441	33.546	15.462
CH08	37.238	55.313	30.700	57.001	39.020	30.983	32.050	30.441	33.546	36.428
CH10	55.304	71.861	31.145	28.704	32.224	29.042	17.665	21.756	20.593	51.278
CH12	51.194	61.349	38.011	36.303	44.647	26.435	32.050	30.441	33.546	36.428
CH15	47.300	142.428	32.350	24.968	67.048	28.239	24.163	20.726	74.382	31.890
CH16.1	41.727	81.864	29.681	29.288	35.142	22.496	26.745	31.322	21.865	33.050
CH17	45.861	92.423	28.776	46.479	49.335	30.983	32.050	30.441	33.546	35.489
CH18	57.704	68.171	37.834	33.421	43.602	27.952	33.310	41.600	77.768	35.745
CH20	50.676	43.016	31.982	26.601	53.477	33.689	32.050	30.441	33.546	36.428
CH21	60.313	90.655	41.369	67.876	53.481	23.252	79.540	18.271	51.744	52.517
CH24	73.375	54.972	38.768	40.081	37.462	38.873	65.685	52.539	23.007	54.728
CH25	51.194	61.349	38.011	36.303	44.647	30.983	32.050	30.441	33.546	36.428
CH27	51.194	61.349	38.337	36.303	44.647	30.983	32.050	30.441	33.546	36.428
CH28	49.038	42.208	34.949	30.367	52.984	25.913	24.718	71.529	28.243	46.782
CH30	49.517	54.551	99.137	49.745	40.318	40.856	40.745	40.817	42.019	47.582
CH32	54.001	60.425	60.377	48.547	39.854	40.697	41.113	42.937	40.088	47.870
NA01	72.697	96.524	117.673	66.289	71.601	55.629	74.863	74.992	49.622	41.458
NA02	66.152	101.456	95.712	74.175	255.650	115.638	102.370	95.511	56.448	41.488
NA03	86.244	166.877	167.043	101.533	217.460	160.111	157.667	111.778	65.114	41.665
NA04	75.964	91.832	126.989	112.164	285.337	166.165	112.887	88.734	59.823	41.521
NA05	356.299	79.284	219.163	244.375	105.656	116.041	149.632	129.405	52.055	41.638
NA06	88.517	107.607	170.094	248.865	124.497	98.455	96.661	170.271	233.692	68.404
NA07	136.458	87.375	204.504	257.288	113.723	71.094	150.401	235.453	234.160	61.364
NA08	49.539	67.238	253.314	95.512	102.561	132.369	139.298	109.735	77.255	52.346
NA09	74.339	68.859	188.336	86.534	106.520	85.230	316.291	133.614	79.270	58.906
NA10	33.443	41.967	129.907	90.735	95.014	86.153	81.539	72.925	84.648	48.228
NA11	92.026	79.041	245.561	102.266	107.537	59.164	91.518	46.217	78.738	48.727
NA12	73.521	141.640	235.090	232.268	310.914	434.150	132.530	146.441	78.214	69.129
NA13	69.931	195.625	339.637	150.851	219.513	164.179	163.768	500.436	78.997	38.339
NA14	56.516	201.847	246.936	134.786	497.622	1084.83	204.951	119.095	62.981	37.767
PI01	46.628	41.028	53.707	41.264	41.150	46.238	41.031	44.987	49.255	37.138
PI02	46.802	490.004	66.520	46.214	45.991	40.052	40.878	41.454	44.485	37.318
PI03	55.089	44.582	77.722	43.404	52.329	43.452	40.998	41.902	46.860	37.231

Station	Year									
	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
PI04	79.631	63.970	67.755	46.989	40.241	43.593	41.049	40.732	49.376	37.201
PI05	47.786	65.924	61.110	40.741	40.443	43.466	40.755	40.559	42.830	37.403
PI06	42.342	99.372	59.608	58.871	39.715	36.012	40.875	42.612	38.250	45.151
PI07	43.165	70.528	66.450	49.506	40.791	37.760	40.974	43.722	43.020	44.464
PI08	50.388	72.287	65.196	40.741	40.226	38.859	40.922	40.736	30.867	43.926
PI09	61.487	65.655	70.923	40.741	40.791	33.475	41.079	40.972	37.471	47.143
PI10	44.947	61.087	50.840	40.036	35.122	39.678	33.657	36.637	41.957	32.325
PI11	29.233	29.998	26.641	25.181	22.004	28.947	20.223	23.165	31.055	21.291
PI12	44.947	61.087	50.840	40.036	35.122	40.655	27.574	36.637	41.957	32.325
PI13	44.947	61.087	50.840	40.036	35.122	39.763	39.866	36.637	41.957	34.373
PI14	31.856	23.864	23.204	28.490	21.944	43.155	20.019	23.328	43.656	40.514
YO01	51.931	76.002	92.657	97.603	106.564	72.331	154.081	71.323	56.028	49.991
YO02	81.037	70.731	92.664	112.822	116.048	104.255	176.813	77.870	69.072	49.987
YO03	65.608	36.249	62.158	444.759	112.606	111.200	126.725	79.185	52.789	49.589
YO04	154.836	119.052	152.213	182.329	128.924	67.220	77.275	103.764	55.201	258.892
YO05	77.976	63.942	77.693	100.023	86.884	68.891	94.774	53.295	50.836	79.855
YO06	77.976	63.942	77.693	100.274	86.884	68.891	94.774	53.295	41.049	78.336
YO07	78.800	59.130	58.931	66.329	67.201	36.944	74.361	68.063	41.947	233.970
YO08	77.976	63.942	77.693	100.274	86.884	68.891	94.774	43.339	60.465	80.826
YO09	77.976	63.942	77.693	100.274	86.884	68.891	94.774	53.295	45.124	65.194
YO10	77.976	63.942	77.693	100.274	86.884	68.891	94.774	53.295	45.124	65.194
YO11	77.976	63.942	77.693	100.274	86.884	68.891	94.774	53.295	45.124	65.194
YO12	77.052	94.287	66.762	68.575	70.535	72.340	78.308	55.355	48.573	41.673
YO13	78.800	63.942	77.693	100.274	86.884	68.891	94.774	53.295	45.124	65.194
WA01	51.340	69.365	57.899	42.874	40.794	51.178	40.662	41.672	45.966	75.075
WA02	35.664	30.516	30.638	30.621	19.115	66.975	23.402	36.876	43.164	61.882
WA03	27.246	21.930	21.730	23.644	17.476	101.265	18.587	32.929	40.845	62.058
WA06	35.664	30.516	30.638	30.621	19.115	66.975	23.402	36.876	43.164	61.882
WA4.1	35.664	30.516	30.638	30.621	19.115	66.975	23.402	36.876	43.164	61.882
WA5.1	35.664	30.516	30.638	30.621	19.115	66.975	23.402	36.876	43.164	61.882

Table 8 Each station Health risk value ($\times 10^{-4}$)

Station	Year									
	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
CH01	2.48	1.76	1.76	1.46	3.12	3.92	2.64	3.52	4.48	9.49
CH03	1.44	0.08	1.14	1.31	2.19	2.34	1.61	2.08	2.03	2.81
CH06	1.44	0.08	1.14	1.31	2.19	2.34	1.61	2.08	2.03	3.32
CH08	1.35	0.95	1.10	0.93	2.06	2.34	1.61	2.08	2.03	3.32
CH10	1.41	0.82	2.02	1.19	2.24	2.10	2.75	1.91	2.72	4.92
CH12	1.51	0.13	1.20	1.40	2.26	3.15	1.66	2.13	2.09	3.38
CH15	1.42	3.43	0.91	1.32	2.07	2.23	2.14	2.17	2.07	5.01
CH16.1	1.34	0.72	0.91	1.04	1.66	3.72	1.49	2.78	2.20	5.00
CH17	1.28	0.98	1.05	1.20	2.49	2.40	1.66	2.13	2.09	3.38
CH18	1.36	0.86	1.37	1.33	2.43	2.88	1.58	1.97	2.03	5.20
CH20	1.97	0.90	1.20	1.45	2.45	1.23	1.66	2.13	2.09	3.38
CH21	1.31	0.87	1.20	1.60	2.34	4.33	1.58	2.22	2.13	5.14
CH24	1.51	0.89	1.11	1.38	2.55	1.91	1.72	2.58	2.01	5.17
CH25	1.51	0.13	1.20	1.40	2.26	2.40	1.66	2.13	2.09	3.38
CH27	1.51	0.13	1.19	1.40	2.26	2.40	1.66	2.13	2.09	3.38
CH28	1.52	0.12	1.21	1.40	2.25	2.48	1.81	2.24	2.18	5.15
CH30	1.34	1.10	1.20	2.46	2.60	1.51	1.41	1.72	1.54	1.33
CH32	1.26	1.11	1.15	2.37	2.02	1.66	1.43	1.49	1.63	1.45
NA01	2.12	1.74	1.33	2.45	1.64	1.34	1.60	1.32	1.71	1.48
NA02	1.38	1.60	1.34	1.63	2.11	1.46	2.28	2.66	6.67	1.38
NA03	1.29	1.61	2.14	1.35	1.81	1.44	2.61	2.94	7.98	1.39
NA04	1.40	1.39	2.16	1.66	1.53	1.81	2.43	2.75	7.94	1.27
NA05	1.37	1.49	2.82	1.57	1.32	2.27	2.81	2.96	7.98	1.43
NA06	1.47	2.12	1.78	1.05	3.01	1.60	2.24	2.49	11.10	11.10
NA07	1.63	1.56	1.35	1.11	1.16	1.46	2.62	2.82	11.10	10.98
NA08	1.06	1.09	4.07	1.18	1.29	1.80	2.42	2.85	11.10	10.98
NA09	1.26	0.80	2.13	1.15	1.76	2.27	2.57	2.69	11.10	10.98
NA10	1.12	0.87	4.15	1.05	1.63	1.31	2.47	2.58	11.10	10.98
NA11	1.50	0.92	1.87	0.90	1.06	1.00	2.57	3.39	11.10	10.98
NA12	1.24	1.98	3.10	2.79	4.10	5.20	2.68	3.80	8.97	3.74
NA13	1.39	5.16	4.01	5.03	2.81	2.28	2.28	4.65	11.10	3.58
NA14	1.12	3.62	1.55	2.37	3.88	14.32	3.79	3.17	6.03	3.18
PI01	1.34	1.86	1.36	2.03	1.98	1.44	1.38	1.43	1.55	1.45

Station	Year									
	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018
PI02	1.21	2.20	1.22	2.32	1.70	1.36	1.51	1.52	1.79	1.33
PI03	1.35	1.79	1.46	1.88	2.20	1.65	1.54	1.64	1.70	1.48
PI04	1.26	1.84	1.03	1.90	2.06	1.62	1.25	1.50	1.58	1.64
PI05	1.39	2.61	1.17	1.86	1.88	1.77	1.57	1.47	1.63	1.68
PI06	1.57	2.00	1.21	1.88	1.71	1.25	1.56	1.45	4.15	11.01
PI07	1.47	2.54	1.52	1.89	1.73	1.73	1.49	1.54	3.89	11.01
PI08	1.21	2.48	1.32	1.86	1.95	1.40	1.36	1.46	3.96	11.01
PI09	9.39	3.03	1.49	1.86	1.74	1.55	1.40	1.57	3.84	11.01
PI10	1.44	1.89	1.01	1.74	1.56	1.16	1.34	1.45	2.08	2.33
PI11	1.03	1.24	0.41	1.01	0.82	2.57	1.11	1.40	2.02	1.51
PI12	1.44	1.89	1.01	1.74	1.56	0.59	1.45	1.45	2.08	2.33
PI13	1.45	1.89	1.01	1.74	1.56	0.96	1.22	1.45	2.08	1.29
PI14	0.96	1.23	0.43	1.35	0.93	0.84	1.11	1.21	1.26	1.15
YO01	1.69	1.90	0.80	1.31	2.07	1.23	3.19	2.80	6.29	1.57
YO02	1.97	2.14	0.76	1.53	1.94	1.58	4.33	3.25	8.50	1.42
YO03	1.72	0.61	0.84	1.71	1.60	1.73	4.08	3.16	7.88	1.80
YO04	2.47	0.75	0.76	2.53	2.39	1.37	2.37	3.23	11.01	11.05
YO05	1.04	0.83	0.60	2.21	1.39	1.05	2.13	1.20	11.01	11.05
YO06	1.04	0.83	0.60	1.61	1.39	1.05	2.13	1.20	6.20	11.05
YO07	0.67	1.44	0.38	1.86	0.86	0.71	1.07	1.00	4.11	11.05
YO08	1.04	0.83	0.60	1.61	1.39	1.05	2.13	0.59	11.01	11.05
YO09	1.04	0.83	0.60	1.61	1.39	1.05	2.13	1.20	6.13	4.13
YO10	1.04	0.83	0.60	1.61	1.39	1.05	2.13	1.20	6.13	4.13
YO11	1.04	0.83	0.60	1.61	1.39	1.05	2.13	1.20	6.13	4.13
YO12	0.74	1.41	0.53	1.54	0.86	0.90	2.44	2.19	1.62	1.66
YO13	0.65	0.83	0.60	1.61	1.39	1.05	2.13	1.20	6.13	4.13
WA01	1.10	6.72	1.58	1.98	1.73	1.42	1.37	1.48	3.83	11.81
WA02	0.47	3.33	0.81	1.77	1.31	1.06	1.17	1.34	3.46	6.10
WA03	0.29	1.75	0.48	1.72	1.05	0.86	1.12	1.34	3.19	3.58
WA06	0.47	3.33	0.81	1.77	1.31	1.06	1.17	1.34	3.46	6.10
WA4.1	0.47	3.33	0.81	1.77	1.31	1.06	1.17	1.34	3.46	6.10
WA5.1	0.47	3.33	0.81	1.77	1.31	1.06	1.17	1.34	3.46	6.10

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