Faculty of Resource Science and Technology

DETERMINATION OF HEAVY METALS IN WATER AND SEDIMENTS OF BATANG AI RESERVOIR AND BATANG AI RIVER

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Determination of Heavy Metals in Water and Sediments of Batang Ai Reservoir and Batang Ai River

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Declaration

I, Liew Siaw Ying, final year student of Chemistry Resource hereby declare that this dissertation is my own work and effort with the guidance of my supervisor, Dr. Sim Siong Fong. No part of the dissertation has previously been submitted for any other degree, university or institution of higher learning.

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(Liew Siaw Ying)

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List of Abbreviations

AAS	Atomic absorption spectrometer
Al	Aluminium
ANOVA	Analysis of variance
APHA	American Public Health Association
As	Arsenic
ATSDR	Agency for Toxic Substances and Disease Registry
CCA	Chromated-copper-arsenate
CCME	Canadian Council of Ministers of the Environment
Cd	Cadmium
CF	Contamination factor
Cr	Chromium
CRM	Certified Reference Material
Cu	Copper
DOE	Department of Environment
EPA	Environmental Protection Authority
ERL	Effects Range-Low
Fe	Iron
FIMS	Flow Injection Mercury System
GPS	Global Positioning System
HCl	Hydrochloric acid
Hg	Mercury
HNO_3	Nitric acid
I_{geo}	Geoaccumulation Index
IZA	International Zinc Association
LOD	Limit of detection
Mn	Manganese
MOH	Ministry of Health
ND	Not Detected

Ni	Nickel
NWQS	National Water Quality Standard
Pb	Lead
PLI	Pollution load index
ppm	Part per million (mg/L)
SALCRA	Sarawak Land Consolidation and Rehabilitation Authority
SD	Standard deviation
Se	Selenium
SIWRM	Sarawak Integrated Water Resource Management
SPSS	Statistical Package for Social Sciences
TEL	Threshold Effects Levels
UNEP	United Nations Environment Programme
USEPA	United States Environmental Protection Agency
Zn	Zinc

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Determination of Heavy Metals in Water and Sediments of Batang Ai Reservoir and Batang Ai River

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ABSTRACT

Heavy metals are one of the vital contaminant in the environment. The objective of the study is to determine the distribution and concentration of zinc (Zn), copper (Cu), iron (Fe), nickel (Ni), lead (Pb), selenium (Se), cadmium (Cd), arsenic (As), manganese (Mn), and mercury (Hg) in water and sediments of Batang Ai Reservoir and Batang Ai River. The water and sediment samples were collected from a total of 16 stations from upstream, downstream and reservoir of Batang Ai River. 4 other stations were located at the fish ponds of SALCRA Batang Ai Inland Fisheries Station. Triplicates of the water and sediment samples were collected from each sampling station. The water at the sub-surface (0-15cm) and the surface sediment were collected, preserved and stored at 4 ℃ before being transported to laboratory for further analysis. In the laboratory, the samples were acid digested, filtered and diluted for analysis with atomic absorption spectrometer (AAS) and mercury analyser. The average concentration of heavy metals in the water decreased in the order of Fe > Zn > Mn > Hg. The average concentration of heavy metals in the sediment decreased in the order of Fe > Mn > Cu > Zn > Ni > Pb > Hg. The quality of sediments was determined through the assessment of contamination factor (CF), geoaccumulation index (I_{geo}) and pollution load index (PLI). The risk of contamination in Batang Ai Reservoir and Batang Ai River is determined as low.

Keywords: Heavy metals; water; sediment; reservoir; river

ABSTRAK

Logam berat merupakan salah satu daripada bahan pencemaran yang penting dalam alam sekitar. Objektif kajian ini adalah untuk mengkaji pengedaran dan konsentrasi zink (Zn), kuprum (Cu), besi (Fe), nikel (Ni), plumbum (Pb), selenium (Se), kadmium (Cd), arsenik (As), mangan (Mn), dan merkuri (Hg) dalam air dan sedimen dari Takungan Batang Ai dan Sungai Batang Ai. Sampel air dan sedimen telah dikumpul daripada 16 stesen dari hulu, hillir, dan takungan Sungai Batang Ai, manakala 4 stesen yang lain telah dikumpul daripada kolam ikan di Stesen Peikanan Darat SALCRA, Batang Ai. 3 sampel untuk air dan sedimen telah dikumpul daripada setiap stesen. Air di bawah permukaan (0-15 cm) dan sedimen dari permukaaan dikumpul dan disimpan pada suhu 4 ℃, sebelum dihantar ke makmal untuk analisis yang seterusnya. Di dalam makmal, sampel-sampel yang diproses telah dianalisis dengan menggunakan Spektrofotometer Serapan Atom (SSA) dan mesin analisis merkuri. Purata konsentrasi logam berat dalam air menurun dengan susunan Fe > Zn > Mn > Hg manakala purata konsentrasi logam dalam sedimen menurun dengan susunan Fe > Mn > Cu > Zn > Ni > Pb > Hg. Kualiti sedimen ditentukan berdasarkan penilaian faktor pencemaran, indeks geoakumulasi, dan Indeks Beban Pencemaran. Tahap pencemaran di Takungan Batang Ai dan Sungai Batang Ai dinilaikan sebagai rendah.

Kata kunci: Logam berat; Air; Sedimen; Takungan; Sungai

1.0 Introduction

1.1 General

According to Hashmi (1997), the Batang Ai River was dammed and Batang Ai Reservoir was constructed for hydroelectric power in 1985. Besides power generation, other activities such as aquaculture and tourism are also found along the Batang Ai River and Reservoir (Sovacool and Bulan, 2011).

Heavy metals are one of the most vital contaminants in the aquatic environment. Hawkes (1997) defines heavy metals as elements in the period 4 or more, from Group 3 to 16 of the periodic table, for instance, mercury (Hg), cadmium (Cd), manganese (Mn), lead (Pb), arsenic (As), and nickel (Ni). As asserted by Adu (2010), heavy metals occur in the aquatic environment naturally or through anthropogenic activities such as industrial wastewater and domestic sewage. Sim *et al.* (2014) explains that heavy metals often exist in the aquatic environment due to their ability to bind with sediment particles or their solubility in water. According to Patil *et al.* (2014), heavy metals are toxic, persistent in the environment, having the propensity to accumulate in the living organisms and thus amplify through the food chain.

The accumulation of the heavy metals in water and sediments will eventually reach human and other living organisms through the aquatic ecosystem, since heavy metals can be transferred through the food chain. Singh *et al.* (2011) asserted that the accumulation of heavy metals in human's body will cause adverse health problems. For instance, they inhibit the biological function of the essential nutritional minerals by replacing their position. In plants, the accumulation of heavy metals could damage the cellular components (Singh *et al.*, 2011). Therefore, the study of heavy metals in water and sediments of rivers and reservoirs had been conducted continuously in order to investigate their status in the environment.

1.2 Problem Statement

A number of studies had been done in Batang Ai Reservoir and River. The studies on the concentration of sulphide, dissolved oxygen and phosphorus as well as the impacts of cage aquaculture activity on the water quality had been conducted by Paka *et al.* (n.d.), Ling *et al.* (2013), and Paka *et al.* (2009), respectively. Furthermore, the distribution of trace and major elements in the hydroelectric reservoir was studied by Sim *et al.* (2014). To date, the study on the distribution of heavy metals in water and sediments of Batang Ai Reservoir and River is yet scarce to determine the stability of heavy metals in the environment.

1.3 Objective

Due to the threat of heavy metals to living organisms, the objective of the study is to determine the distribution and concentration of heavy metals in water and sediments of Batang Ai Reservoir and Batang Ai River.

2.0 Literature Review

2.1 Study Area

Batang Ai River was dammed in 1985 (Hashmi, 1997). Batang Ai Dam has a maximum height above foundation of 110 m and approximately 40 to 45 m^3 s^{-1} of allowable volume rate of flow of water (Al-Zubaidy and Rigit, 1997). According to Ling *et al.* (2013), Batang Ai Reservoir is located at Batang Ai National Park, Lubok Antu, Sarawak, Malaysia. It is approximately 250 km in the direction of south east from Kuching, Sarawak. Batang Ai Reservoir is a tropical reservoir (Ling *et al.*, 2013). Its catchment area is 1200 km^2, surface area is 85 km^2, gross storage volume is 2.38 km^3, active volume is 0.73 km^3, and surface elevation is 108 m above the mean sea level (Sarawak Integrated Water Resource Management (SIWRM), 2008). The activities conducted along the Batang Ai Reservoir and River are hydropower generation, freshwater aquaculture, and tourism (Sovacool and Bulan, 2011).

2.2 Heavy Metals

Heavy metals are one of the important groups of pollutant in the environment. According to Tamás *et al.* (2014), heavy metals are a group of elements that are loosely defined, including the transition metals and some metalloids. Duffus (2002) on the other hand stated that heavy metals are metals with density of 3.5 to 7 g cm^{-3}. Adu (2010) mentioned that heavy metals could remain and accumulate in the environment without breaking down, especially in sediments and lakes. They can be transferred from one environment compartment to another (Adu, 2010). As explained by Sim *et al.* (2014), heavy metals are water-soluble and they can also bind to the sediment particles. Therefore, they often exist in the aquatic environment after being released from the natural or anthropogenic sources (Sim *et al.*, 2014). According to Patil *et al.* (2014), heavy metals are toxic, persistent in the environment, having the propensity to accumulate in the living organisms and thus amplify through the food chain. Adu (2010) mentioned that the concentration of heavy metals in terrestrial and aquatic organisms is controlled by the size of the source and adsorption in soils and sediments. Often, the free ion of the heavy metals is the most bioavailable form and its concentration can be used to indicate toxicity (Adu, 2010). Duruibe *et al.* (2007) stated that some of the elements such as Fe and Zn are essential for human, nonetheless they possess toxic effect when exceeding the tolerance limit. Most of the heavy metals are highly toxic even at a low concentration (Duruibe *et al*, 2007).

In the aquatic systems, there are many factors which affect the partitioning of heavy metals in the surface water and sediment. These factors include pH, redox environment for the water-sediment interface, organic matters, temperature, dissolved oxygen, activities of microorganisms, and river flow (Elder, 1988; Li *et al.*, 2013; Salomons, 1995). Salomons (1995) highlighted that pH is the most vital factor that controls metal speciation, solubility from surfaces of mineral, transport and also the bioavailability of metals in the water. In addition, the solubility of metal hydroxide and the adsorption-desorption process are also influenced by pH. Different adsorption behaviour of metals differs according to pH. Under the pH of natural water, most of the metal hydroxides have very low solubility. At lower pH, the solubility of metal hydroxides increase due to the direct relationship of hydroxide ion activity with pH. As this takes place, more dissolved metals are formed, increasing the bioavailability of the element for the biological processes. (Salomons, 1995).

2.3 Sources of Heavy Metals

According to Adu (2010), heavy metals are naturally occurring elements in the Earth's crust. Heavy metals exist naturally with a range of natural background concentrations in soils, waters, sediments and organisms. The weathering of soils and rocks may naturally release heavy metals into the aquatic environment, such as rivers and lakes (Indian and Northern Affairs Canada, 2004). Greaney (2005) stipulated that the heavy metals enter the aquatic systems through the interaction between water, sediment, and atmosphere in nature. Its concentration changes due to natural hydrodynamic chemical and biological forces. However, the anthropogenic activities have changed the natural interaction until the water and aquatic life have been endangered. The most important factor that determines the activity and effect of heavy metals in the aquatic system is their ability to interact with the organic compounds in water and sediments. The organic compounds may come from nature, such as vegetative decay, or as a consequence of pollution like the organic discharge from municipal and industrial sources (Greaney, 2005). Some of the anthropogenic sources of heavy metals are listed in the section below.

2.3.1 Metalliferous Mining and Smelting of Mineral Ores

According to Jang *et al* (2005), the mine tailings may release As, Pb, Cu and Cd into the ground and surface water system. Scoullos *et al.* (2001) stated that Hg is released from the mining of gold. Pacyna (1987) highlighted that the major volatile pollutants that are released from the smelting of minerals ores are As, Cd, Pb and Hg.

2.3.2 Agriculture

As stipulated by Keskin (2010), the excessive use of fertilisers and pesticides in agriculture would pollute the surface water and groundwater with nitrate and heavy metals. Typical heavy metals that come from agricultural activities are Pb and Hg (Keskin, 2010). Puttaiah and Kiran (2008) stated that the use of phosphate fertiliser also increased the leaching of Cd into the lake. Besides that, Jones and Jarvis (1981) mentioned that some common pesticides that are used in agriculture contain considerable amount of metals. About 10% of the chemical used for pesticides in UK contain Hg, Zn, Cu, Pb, and Mn. One of the examples is the Bordeaux mixture (copper sulphate) (Jones and Jarvis, 1981). McLaughlin *et al.* (2000) highlighted that lead arsenate was utilised as insect pesticide in the fruit orchard. Arsenic-containing compounds were widely used to control pest in banana in Australia and New Zealand (McLaughlin *et al.*, 2000).

2.3.3 Domestic and Industrial Wastewater

Scheinberg (1991) asserted that Pb, Cu, Zn and Cd are generally found in domestic sewage. As stipulated by Houhou *et al.* (2009), the major source of Zn and Pb is the household activities. Barakat (2011) highlighted that the untreated industrial effluent contain a high concentrations of heavy metals including Cd, Cu, As, Cr, Pb, Ni, and Zn.

2.3.4 Animal Manures

Nicholson *et al*. (1999) asserted that animal feed contains heavy metals such as Cd, As, Pb, Cr, Cu, Ni, and Zn. They can be released to the land through manures of poultry, cattle and swine (Nicholson *et al*., 1999) and further leached into the aquatic ecosystem during raining.

2.3.5 Aquaculture

The antibiotic and agrochemicals formulated feed causes heavy metals pollutants in aquaculture (Ajiboye *et al.,* 2011). Seim *et al.* (1997) mentioned that the algaecide ($CuSO_3$), erosion from pipes made of Cu and net preservatives used in aquaculture may release Cu into water. Besides that, Se is also being introduced into the animal feeds, including fish feed (Ikem and Egilla, 2008; Khanal and Knight, 2010).

2.3.6 Construction

As mentioned by Moghaddam and Mulligan (2008), the use of chromated-copper-arsenate (CCA) treated wood in the construction results in the release of Cr, Cu, and As into water. Furthermore, Pb and Zn are released to the environment due to the use of lead-based and zinc-based paint in construction (United States Environmental Protection Agency (USEPA), 1996).

2.4 Biological Roles and Effects of Selected Heavy Metals

2.4.1 Lead (Pb)

According to Rastogi (2008), Pb is a non-essential metal with no biological function to human, animals and plants, although it is important in the industry. Duruibe *et al.* (2007) stated that an important effect of lead toxicity to human is abnormalities of physiological development. Besides that, lead toxicity also causes damage to the urinary tract, gastrointestinal tract, nervous system and brain (Duruibe *et al.*, 2007). In addition, Ogwuegbu and Muhanga (2005) stipulated that lead toxicity could cause damage to the kidneys, nervous system, reproductive system, and synthesis of erythrocytes. In plants, lead may harm the biochemical, physiological, and morphological functions, for example, hindering the synthesis of ATP energy and growth of plants (Pourrut *et al.*, 2011).

2.4.2 Cadmium (Cd)

Eisler (2000) mentioned that cadmium is highly toxic and it has no biological function to human, plants and animals. According to Duruibe *et al.* (2007), cadmium is toxic to human even at a very low concentration. Chronic exposure to chromium causes kidney and myocardial impairment, cadmium pneumonitis, increment in blood pressure and bone defects (Duruibe *et al.*, 2007). Besides, Kumar and Singh (2010) explained that cadmium presents at high concentration may cause changes in the functions and structures of the organs in fish, such as the kidneys and gills. In addition, cadmium may weaken the immune system of fish and mammals (Giari *et al*., 2007; Kim *et al*., 2000; Zelikoff *et al*., 1995).

2.4.3 Mercury (Hg)

As stipulated by Duruibe *et al.* (2007), mercury is toxic and its function in human is not yet discovered. Inorganic mercury affects the foetal development (Duruibe *et al.,* 2007). Järup (2003) mentioned that inorganic mercury may cause lung damage, psychological and neurological problems, and kidney damage. The psychological and neurological problems are reversible after the termination of exposure. The organic form of mercury, such as dimethylmercury causes nervous system and vision damage and also death (Järup, 2003). In plants, mercury may influence photosynthesis and decrease the uptake of water (Sas-Nowosielska *et al.,* 2008).

2.4.4 Arsenic (As)

Järup (2003) asserted that inorganic arsenic is very toxic and large amount will cause gastrointestinal problems, cardiovascular and central nervous systems problems and even death. Populations that are exposed to arsenic through drinking water have higher risk of mortality from lungs, bladder and kidney cancer. The studies show that the higher the exposure, the higher risk of getting these cancers. In addition, the risk of skin cancer and other skin lesion such as pigmentation changes also increases due to the exposure to arsenic (Järup, 2003).

2.4.5 Zinc (Zn)

International Zinc Association (IZA) (2011) mentioned that zinc is a vital trace element to organisms. It has many biological functions, such as maintaining the fertility, stimulating growth, and improving the immune system. However, the maximum amount of zinc that an adult can tolerate is 40 mg per day. When exceeding this tolerance limit, it may cause gastrointestinal problems, headache, and affect the function of other elements, mainly copper and iron in the body (IZA, 2011).

Frassinetti *et al.* (2006) claimed that zinc is important for the normal reproduction and growth of higher animals and plants. In plants, severe zinc toxicity may cause the death of leaves and retardation of the root growth (Harmens *et al*., 1993; Ren *et al*., 1993). In animals, severe zinc toxicity may cause difficulty in swallowing and reducing the growth rate (Medeiros and Wildman, 2013).

2.4.6 Selenium (Se)

As stated by Agency for Toxic Substances and Disease Registry (ATSDR) (2003), selenium can be useful and harmful to human. Se is a trace element which is required by human body to maintain good health. However, the daily intake of Se more than 5 mg/kg body weight may lead to undesirable health effects. The symptoms of short term oral exposure to high concentration of Se are diarrhoea, nausea, and vomiting. Chronic oral exposure to compounds containing high Se content can result in selenosis, which has the symptoms such as neurological abnormalities and nail brittleness. Brief exposure to high amount of elemental Se or SeO_2 can cause bronchitis, breathing difficulties, and respiratory tract irritation whereas long term exposure may leads to respiratory irritation, coughing and bronchial spasms (ATSDR, 2003).

In animals, high concentration of selenium may affect the reproductive cycle in females, besides reducing the production of sperm in males (ATSDR, 2003).

2.5 Selected Experiences of Heavy Metals in the Aquatic Environment

2.5.1 Avsar Dam Lake, Turkey

Öztürk *et al.* (2009) highlighted the concentration of heavy metals in the Avsar Dam Lake: 0.0001 - 0.0012 mg/L for Cd, 0.001 - 0.012 mg/L for Cr, 0.01 - 0.02 mg/L for Cu, 0.28 - 2.39 mg/L for Fe, 0.0004 - 0.012 mg/L for Ni, and 0.0003 - 0.019 mg/L for Pb. The concentrations of heavy metals in the lake were increased in the order of Cd < Ni < Cr < Pb < Cu < Fe. The concentrations found in sediments samples were 0.34 - 1.23 mg/L for Cd, 9.41 - 19.9 mg/L for Cr, 18.2 - 38.4 mg/L for Cu, 19680 - 28560 mg/L for Fe, 19.8 - 39.4 mg/L for Ni, and 0.64 - 6.35 mg/L for Pb. The concentrations of heavy metals in the sediment increased in the order of Cd < Pb < Cr < Cu < Ni < Fe (Öztürk *et al.*, 2009).

2.6.2 Manwan Reservoir, Lancang River

Wong *et al.* (2012) reported the mean concentrations of heavy metals in sediments in a descending order: Al (46,348.22 µg/g) > Fe (32,444.82 µg/g) > Mn (564.11 µg/g) > Zn (156.69 µg/g) > Cr (54.70 µg/g) > Pb (47.14 µg/g) > As (40.64 µg/g) > Cu (38.94 µg/g) > Cd (1.41 µg/g). The quality of sediment assessed by sediment quality guideline showed that As and Cd were found at almost all sites and Cu, Cr, Zn, and Pb at a few sites with concentrations more than the Threshold Effects Levels (TEL) or Effects Range-Low (ERL) value. This meant the concentrations of heavy metals were high and may cause dreadful effect (Wong *et al.*, 2012).

3.0 Materials and Methods

3.1 Sampling

Water and sediment samples were collected from a total of 16 stations from upstream, downstream and reservoir of the Batang Ai River. Four other stations were located at the fish ponds of SALCRA Batang Ai Inland Fisheries Station. The sampling locations are shown in Figure 1 with S indicating stations at the river (S1 – S16) whilst P referring those at the fish ponds (P1 – P4). Table 1 summarises the GPS coordinates of the sampling locations.

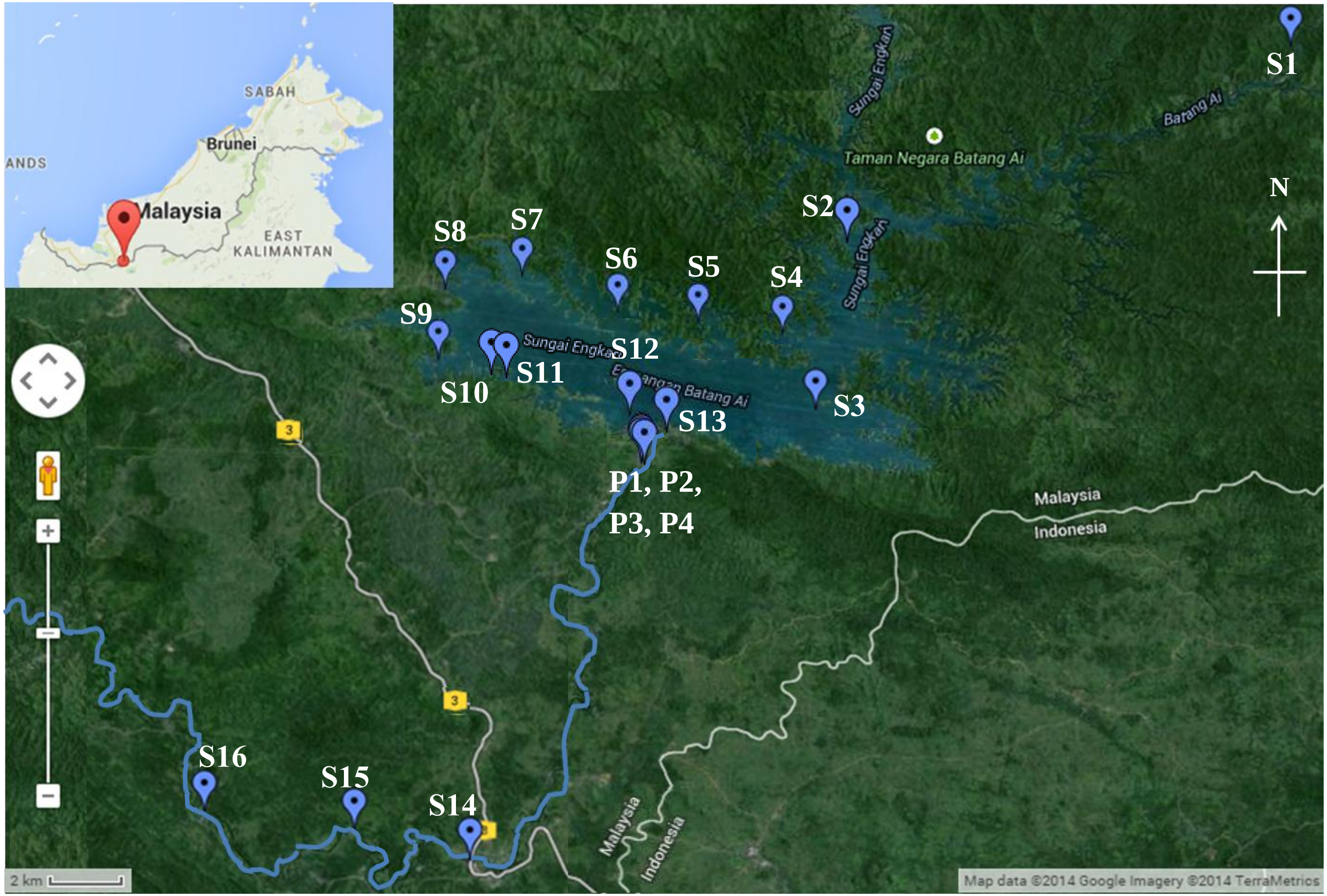

Figure 1: Sampling locations in Batang Ai Reservoir, Batang Ai River, and fish ponds in Batang Ai Inland.

Table 1: GPS coordinates and types of sample collected from each sampling station.
* Samples collected from fish ponds at SALCRA Batang Ai Inland Fisheries Station.

Station	Description	GPS Coordinates	Water	Sediment
S1	Batang Ai- Upstream, flowing	N01° 14’ 47.4” E112° 02’ 14.6”	✓	✓
S2	Confluence of Batang Ai & Engkari	N01° 11’ 40.8” E111° 55’ 32.6”	✓	✓
S3	No aquaculture	N01° 09’ 08.9” E111° 55’ 03.7”	✓	✓
S4	Future SALCRA- Teluk Santu	N01°10’23.76’’ E111°54’42.85’’	✓	✓
S5	Future SALCRA-Teluk Pudai	N01°10’36.28’’ E111°53’5.54’’	✓	✓
S6	Cage aquaculture site- Able aquaculture (Teluk Telaus)	N01°10’42.9’’ E111°52’00.5’’	✓	✓
S7	Lepong Mawang (Nanga Tiga)	N01°11’09.3’’ E111°50’37.8’’	✓	✓
S8	Sebangki Dam	N01°11’01.7’’ E111°49’23.7’’	✓	✓
S9	Teluk Kaong	N01°09’52.6’’ E111°49’25.0’’	✓	✓
S10	Aquaculture	N01° 09’ 42.0” E111° 50’ 10.4”	✓	✓
S11	Abandoned cage culture site- Tiang Laju	N01°09’40.0’’ E111°50’23.5’’	✓	✓
S12	Near outflow of reservoir	N01º 09’ 02.5” E111º 52’ 13.8”	✓	✓
S13	Outflow of reservoir	N01° 08’ 48.9” E111° 52’ 44.1”	✓	✓
S14	Bridge to Lubok Antu town	N01° 02’ 37.7” E111° 49’ 53.6”	✓	✓
S15	Batu Bebini	N01° 02’47.7” E111° 48’ 13.2”	✓	✓
S16	Near Longhouses at Langan Empit	N01° 03’ 19.1” E111° 46’ 04.2”	✓	✓
P1*	Fish pond at Batang Ai Inland Fisheries Station	N01° 08’ 26.8’’ E111° 52’ 22.9’’	✓	✓
P2*	Fish pond at Batang Ai Inland Fisheries Station	N01° 08’ 25.6’’ E111° 52’ 23.6’’	✓	-
P3*	Fish pond at Batang Ai Inland Fisheries Station	N01° 08’ 21.3’’ E111° 52’ 25.4’’	✓	-
P4*	Fish pond at Batang Ai Inland Fisheries Station	N01° 08’ 22.6’’ E111° 52’ 23.7’’	✓	✓