ABUNDANCE AND DISTRIBUTION OF MICROPLASTICS AND PERSISTENT ORGANIC POLLUTANTS IN SELECTED RIVERS OF PENINSULAR MALAYSIA

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FACULTY OF SCIENCE UNIVERSITI MALAYA KUALA LUMPUR

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ABUNDANCE AND DISTRIBUTION OF MICROPLASTICS AND PERSISTENT ORGANIC POLLUTANTS IN SELECTED RIVERS OF PENINSULAR MALAYSIA

ABSTRACT

Nearly 70 to 80 percent of microplastics are believed to be transported to the sea through rivers. Yet, less attention has been given to the accumulation of microplastics in the freshwater environment. They are emerging anthropogenic contaminants on account of their potential to adsorb and/or release POPs from and/or to the surrounding environment, and become the vector transferring these pollutants from water to the food web. This study is aimed to explore the abundance and distribution of microplastics and POPs in six rivers of Peninsular Malaysia, which comprised of three rivers from the West Coast (Sepetang River, Serkam River, and Ayer Masin River) and three rivers from the East Coast (Sedili Besar River, Cherating River, and Semerak River). The objectives of this research are to identify the anthropogenic activities that may contribute to the generation of microplastics and POPs, to investigate the abundance and distribution of microplastics, to determine the concentrations of POPs, and, to analyze the relationship between the microplastics abundance, POPs concentration and anthropogenic activities along these rivers. Eight sites were established along each river to ensure maximum coverage of the rivers studied, while anthropogenic activities were investigated through observation of 5 km radius along each river. River sediment samples were collected using shovel, while plankton nets were utilized to capture microplastics in river water. Density separation was conducted to extract microplastics in the sediment, using concentrated NaCl solution followed by wet sieving through a set of Tyler Sieves. Identification of microplastics was conducted based on morphological characteristics (i.e. type, size and colour). As for POPs identification, Liquid-liquid Extraction and Soxhlet Extraction were performed, followed

by GC-MS/MS analysis. Sepetang River, Cherating River, and Ayer Masin River substantially demonstrated greater number of anthropogenic activities, characterized as the hotspots of anthropogenic activities, followed by Sedili Besar River, Serkam River and Semerak River. Correspondingly, the highest prevalence of microplastics abundance in river sediment was discovered in Sepetang River with an average abundance of 101.39 \pm 54.69 particles/kg, while in river water, Ayer Masin River had the greatest number of microplastics with average abundance of 0.0101 ± 0.0052 particles/m³. Majority of the microplastics were films and white in colour, with a dominant size fractions of 1.0 to 5.0 mm in river sediment, and 0.1 to 0.5 mm in river water. 1,2-Benzenedicarboxylic acid, diisooctyl ester (DIOP), which are generally grouped in Phthalate esters (PAEs), was the dominant POPs identified. The highest concentration of POPs in river sediment was observed in Sedili Besar River (677.49 ppm), while the highest concentration in river water was recorded in Cherating River (153.41 ppm). Generally, it can be said that the problem of microplastics and POPs pollution rooted in the prevailing production and consumption pattern of plastic materials, on grounds of great contributions from various anthropogenic activities. Correlation study revealed no relationship between the abundance of microplastics and POPs concentrations in rivers of Peninsular Malaysia. Overall, the results of this study could provide valuable background information for microplastics and POPs pollution in selected rivers of Peninsular Malaysia.

Keywords: anthropogenic, microplastic, persistent organic pollutant, river

KEKERAPAN DAN TABURAN MIKROPLASTIK DAN PENCEMAR ORGANIK TEGAR DI SUNGAI-SUNGAI TERPILIH DI SEMENANJUNG MALAYSIA ABSTRAK

Hampir 70 hingga 80 peratus mikroplastik dipercayai dihanyutkan ke laut melalui sungai. Namun, pengumpulan mikroplastik di persekitaran air tawar kurang diberikan perhatian. Mikroplastik merupakan pencemar antropogenik kerana berpotensi untuk menyerap dan/atau melepaskan pencemar organik tegar (POPs) dari/ke persekitaran di sekelilingnya, dan menjadi vektor yang memindahkan bahan pencemar ini dari air ke jaringan makanan. Kajian ini bertujuan untuk meneroka kekerapan dan taburan mikroplastik dan POPs di enam sungai terpilih di Semenanjung Malaysia. Tiga sungai dipilih dari Pantai Barat (Sungai Sepetang, Sungai Serkam, dan Sungai Ayer Masin) dan tiga sungai dipilih dari Pantai Timur (Sungai Sedili Besar, Sungai Cherating, dan Sungai Semerak) Semenanjung Malaysia. Objektif kajian adalah untuk mengenal pasti aktivitiaktiviti antropogenik yang boleh menyumbang kepada penjanaan mikroplastik dan POPs, untuk mengesan kekerapan dan taburan mikroplastik, untuk menentukan kepekatan POPs, serta untuk menganalisis hubungan antara taburan mikroplastik, kepekatan POPs dan aktiviti antropogenik di sepanjang sungai-sungai tersebut. Lapan lokasi telah dipilih di sepanjang sungai untuk memastikan liputan maksima kawasan kajian, sementara aktiviti antropogenik disiasat melalui pemerhatian di sekitar 5 km radius di sepanjang sungai. Sampel sedimen sungai dikumpulkan menggunakan sekop, sementara jaring plankton digunakan untuk mengumpul mikroplastik di dalam air sungai. Pemisahan ketumpatan dilakukan untuk mengekstrak mikroplastik di dalam sedimen, dengan menggunakan larutan NaCl pekat diikuti dengan penapisan basah menggunakan penapis Tyler. Identifikasi mikroplastik dilakukan berdasarkan ciri-ciri morfologi (iaitu jenis, saiz dan warna). Bagi identifikasi POPs, pengekstrakan Cecair-cecair dan Pengekstrakan Soxhlet telah dilakukan, diikuti dengan analisis GC-MS/MS. Sungai Sepetang, Sungai Cherating, dan Sungai Ayer Masin mempunyai aktiviti antropogenik yang lebih banyak, yang disifatkan sebagai titik panas aktiviti antropogenik, diikuti oleh Sungai Sedili Besar, Sungai Serkam dan Sungai Semerak. Sejajar dengan itu, taburan tertinggi mikroplastik di sedimen sungai ditemui di Sungai Sepetang dengan jumlah purata 101.39 ± 54.69 bilangan/kg, manakala Sungai Ayer Masin mempunyai taburan mikroplastik terbesar di dalam air sungai, dengan taburan purata 0.0101 ± 0.0052 bilangan/m³. Sebahagian besar mikroplastik adalah filem dan berwarna putih, dengan pecahan saiz dominan 1.0 hingga 5.0 mm untuk sedimen sungai, dan 0.1 hingga 0.5 mm untuk air sungai. 1,2-Benzenedicarboxylic acid, diisooctyl ester (DIOP), yang umumnya dalam kumpulan Phthalate ester (PAE), adalah POPs yang dominan yang dikenal pasti, dengan kepekatan tertinggi di dalam sedimen sungai yang diperolehi dari Sungai Sedili Besar (677.49 ppm), manakala kepekatan tertinggi di dalam air sungai yang direkodkan adalah dari Sungai Cherating (153.41 ppm). Pada amnya, boleh dikatakan bahawa masalah pencemaran mikroplastik dan POPs adalah akibat daripada pengeluaran dan penggunaan bahan plastik yang tinggi, dengan sumbangan besar daripada pelbagai aktiviti antropogenik. Kajian korelasi menunjukkan tiada hubungan selari di antara taburan mikroplastik dan kepekatan POPs di sungai-sungai di Semenanjung Malaysia. Secara keseluruhan, hasil kajian ini dapat memberikan maklumat latar belakang yang bernilai untuk pencemaran mikroplastik dan POPs di sungai-sungai terpilih di Semenanjung Malaysia.

Kata kunci: antropogenik, mikroplastik, pencemar organik tegar, sungai

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LIST OF SYMBOLS AND ABBREVIATIONS

<	:	less than
dw	:	dry weight
WW	:	wet weight
sal	:	salinity
Т	:	Temperature
kg/person/day	:	kilogram per person per day
mg/L	:	milligram per liter
ng/L	:	nanogram per liter
particles/m ³	:	particles per cubic meter
ppm	:	parts per million
ppt	:	parts per trillion
n.d.	:	not determined
tonnes/day	:	tonnes per day
ABS	:	Acrylonitrile Butadiene Styrene
BDP	:	1,2-Benzenedicarboxylic acid, butyl decyl ester
СР	:	Cellophane
DCM	:	Dichloromethane
DDT	:	Dichlorodiphenyltrichloroethane
DEHS	:	Decanedioic acid, bis(2-ethylhexyl) ester
DIOP	:	1,2-Benzenedicarboxylic acid, diisooctyl ester
DOE	:	Department of Environment
EPS	:	Expanded Polystyrene
EU	:	European Union
FAO	:	Food and Agriculture Organization of the United Nations

GCMS/MS	:	Gas Chromatography Mass Spectrometry
GDP	:	Gross Domestic Product
GESAMP	:	United Nations Group of Experts on the Scientific Aspects of Marine Pollution
GPML	:	Global Partnership on Marine Litter
HCl	:	Hydrochloric acid
HDPE	:	High-density polyethylene
H_2O_2	:	Hydrogen peroxide
HNO ₃	:	Nitric acid
IPEP	:	International POPs Elimination Project
IUCN	:	International Union for the Conservation of Nature and Natural Resources
КОН	:	Potassium hydroxide
LDPE	:	Low density polyethylene
LLDPE	:	Linear low-density polyethylene
MEHP	:	1,2-Benzenedicarboxylic acid, mono(2-ethylhexyl) ester
MSW	:	Municipal Solid Waste
NaCl	:	Sodium chloride
NAOH	:	Sodium hydroxide
NGOs	:	Non-government organizations
NOAA	:	National Oceanic and Atmospheric Administration
OCDD	:	Octachlorodibenzodioxin
PAEs	:	Phthalate esters
PAHs	:	Polycyclic aromatic hydrocarbons
PAM	:	Polyacrylamide
PAN	:	Polyacrylonitrile
PBDE	:	Polybrominated diphenyl esters

PCBs	:	Polychlorinated biphenyls
PCDD	:	Polychlorinated dibenzodioxins
PFOs	:	Perfluorooctanesulfonic acid
PE	:	Polyethylene
PETE	:	Polyethylene terephthalate
PMMA	:	Polymethyl methacrylate
POPs	:	Persistent Organic Pollutants
PP	:	Polypropylene
PS	:	Polystyrene
PSS	:	Poly(Styrenesulfonate)
PTFE	:	Poly tetra fluoroethylene
PVC	:	Polyvinyl chloride
TAIEF	:	Environmental Technical Assistance and Information Exchange Facility
TDS	:	Total Dissolved Solids
UNCLOS	:	United Nations Convention on the Law of the Sea
UNDP	:	United Nations Development Programme
UNEP	÷	United Nation Environmental Programme
UNICEF	:	The United Nations Children's Fund
USDA	:	United States Department of Agriculture
USEPA	:	United States Environmental Protection Agency
WHO	:	World Health Organization
WWAP	:	United Nations World Water Assessment Programme
WWF	:	World Wide Fund for Nature

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CHAPTER 1: INTRODUCTION

1.1 River Pollution

Rivers perform a suite of ecological functions (e.g. providing habitat, medium of transportation, aquaculture, and shielding effects) and are deemed to be essential for providing a prerequisite of decent public health and aquatic life (Liao *et al.*, 2017). In recent years, anthropogenic activities which release pollutants, accompanied by natural processes (i.e. precipitation inputs, erosion, and seasonal effects) have caused river ecosystems to suffer from immense levels of land-based pollutant loads (Tian *et al.*, 2019).

On top of that, improper waste management, uncontrolled urbanization, incomplete wastewater treatment, as well as, insufficient adherence to laws and regulations, are considered to be among the contributing factors to river pollution worldwide (Mishra *et al.*, 2017). In Malaysia, 43% of 477 rivers overseen by the Department of Environment Malaysia (DOE) in 2017, were found to be slightly polluted with 11% classified as polluted (Figure 1.1) (EQR, 2017).



Figure 1.1: River Water Quality Trend in Malaysia (2008-2017) (EQR, 2017).

River pollution have led to the deterioration of river water quality, biodiversity and river functionality, leaving river ecosystem health under huge threat (Wu *et al.*, 2018; Pan *et al.*, 2019; Zhao *et al.*, 2019).

1.2 Plastics

Plastic has undoubtedly extraordinary properties: is easy to process, low production costs, durable, lightweight and versatile, evidenced by continuing increase and tremendous consumption of polymers worldwide. They are fundamental in the advancement of technologies with extensive applications in industry, construction, medicine and food protection (Cole *et al.*, 2011). Cumulatively, from 1.4 million tonnes of plastics produced in 1950s, there is an increment of nearly 200-fold, reaching 7.8 billion tonnes in 2015, and is expected to reach 30 billion tonnes by 2050 (Plastics Europe, 2018).

Side effect of the mass plastic production is an enormous plastic waste that ends up in the environment, causing deleterious environmental, health and economic impacts. Globally, in 2015, around 302 million tonnes of plastics ended up as waste from 407 million tonnes of primary plastics produced (Hannah & Max, 2019). In developed countries particularly the United States, plastic waste has reached 35.7 million tonnes in 2014, indicating a 13% increase over 2011 (Themelis & Mussche, 2014) and in the United Kingdom, there is over 60% increment of plastic waste from 2005 to 2015 (Anuar *et al.*, 2017). The same trend was also observed in developing countries such as Malaysia which experienced a growth in plastic waste by 18% in five years (Abnisa *et al.*, 2013).

In addition, 2 to 5% of global plastic waste generated are mismanaged and ultimately at risk of entering the oceans and other environments (Cho *et al.*, 2019). The biggest contributor being China of 28%, followed by Indonesia (10%), while the Philippines and Vietnam both contribute 6% of the mismanaged plastic waste (Jambeck *et al.*, 2015). Across many low-to-middle-income countries in Sub-Saharan Africa, between 80 and 90% of plastic waste is inadequately disposed of, and thus plastic pollution has been highlighted as a contaminant of global environmental and economic concern (Botterell *et al.*, 2019).

At global level, plastic waste constitutes 83 to 87% of all marine debris documented, where an estimated 5 to 12 million tonnes of plastics (i.e. between 1.5 and 4% of plastics produced) is released into the marine environment annually (Troost *et al.*, 2018). Recent estimate illustrates that more than 5.25 trillion marine plastic debris (i.e. 243,978 tonnes in weight) are floating in sea around the world (Eriksen *et al.*, 2014).

Along with direct inputs from seas, 70 to 80% of marine debris were reported to be transported through river systems (Bowmer & Kershaw, 2010), of which plastics are transported at an astounding amount of 1.15 to 2.41 million tonnes per year (Lebreton *et al.*, 2017). Astonishingly, 86% are from Asian rivers, with 7.8% from Africa and South America at 4.8% (Lebreton *et al.*, 2017).

The notorious plastic debris exists in a wide variety of sizes, ranging from metres to micrometres, with 92% are reported to be of microplastics (Van Sebille *et al.*, 2015; Wesch *et al.*, 2016). Consecutive sections discuss microplastics and their associated impacts.

1.3 Microplastics

By definition, microplastics are small plastic particles of less than 5 mm in diameter, that may either be manufactured for particular industrial or domestic applications (primary microplastics) or result from the fragmentation of larger plastics (secondary microplastics) caused by waves, UV induced photolysis and microbial decomposition of discarded plastics (Saliu *et al.*, 2018). Examples of primary microplastics include microbeads used in personal care products, pre-production pellets used as precursors to manufacture plastic products, and fibers derived from fabrics made with synthetic materials, such as acrylic and polyester (Browne *et al.*, 2011; Eriksen *et al.*, 2013).

Approximately 5,000 to 94,500 microbeads could be discharged from an exfoliant in a single use and a single garment alone could release up to one million fibers per washing (Napper *et al.*, 2015). In Malaysia, an estimated 0.199 trillion microbeads are released into the marine environment annually (Praveena *et al.*, 2018). Various literatures have demonstrated the differences of microplastic concentrations spatially (McCormick *et al.*, 2014). For instance, watersheds that are closer to urban areas have been observed to contain higher concentrations of microplastics (Mani *et al.*, 2015; Baldwin *et al.*, 2016; Watkins et al., 2019).

Microplastics persist in the environment for a very long time due to their relatively stable chemical properties coupled with extremely low degradation rate (Roy *et al.*, 2011). Additionally, the bioaccumulation potential of microplastics increases as the size decreases (Wagner *et al.*, 2014). Owing to their small size, microplastics may be ingested by an array of aquatic biota ranging from plankton and fish to birds and even mammals (Wright *et al.*, 2013). Microplastics ingestion has been identified in more than 220 species globally, in which if current rate of microplastics accumulation persists, 99% of seabird species are in high probability to ingest microplastics, by 2050 (Li *et al.*, 2019).

On top of that, microplastics are being recognized as an emerging anthropogenic contaminant because of their potential to adsorb organic contaminants such as Persistent Organic Pollutants (POPs) and priority metals from the surrounding environment (Brennecke *et al.*, 2016; Liu *et al.*, 2019). Microplastics particles contain a multitude of chemical additives and the hydrophobicity along with high surface area to volume ratio leads to sorption of contaminants (Bakir *et al.*, 2014). Section 1.3 discuss POPs and their occurrences in the environment.

Via biomagnification and bioaccumulation, microplastics will become a transport vector of hazardous chemicals from aquatic environment to the food web, exposing risks to biota, which include humans and environment (Fauziah *et al.*, 2018). Recently,

substantial amounts of microplastics were detected in bottled waters (i.e. concentrations up to 6, 292 ± 10 , 521 particles/L) which may jeopardize food security and human health as they can be directly ingested by consumers (Oßmann *et al.*, 2018).

Furthermore, microplastics can transport alien species, as well as, becoming the reservoirs in the transmission of pathogen. This is due to the fact that microplastics are subjected to biofouling that leads to colonization by microorganisms including invertebrates, which simultaneously widen the threats of microplastics (Andrady, 2011).

1.4 Persistent Organic Pollutants (POPs)

The prevalent use of POPs has caused deleterious impacts to human and the environment. Sources of POPs include being specifically produced by industries for a wide variety of applications such as pesticides and polymers; as well as, unintentionally generated as by-products of industrial activities or combustion processes (Agamuthu & Narayanan, 2013). Toxicological phenomenon caused by POPs is highly damaging as these compounds remain intact in the environment exceptionally for a long period of time as they are non-biodegradable in nature (i.e. resist to photolytic, chemical and biological degradation) (Gaur *et al.*, 2018). The high octanol-water partition coefficients of media with POPs exacerbates the accumulation of such contaminants in the environment, making them natural POPs sinks (Espinosa-Reyes *et al.*, 2019).

Phthalate esters (PAEs) such as di-isobutyl phthalate, di-n-butyl phthalate and di-(2ethylhexyl) phthalate, most commonly used as plasticizers in the polymer industry have been classified as a major group of POPs by the US EPA and the European Union, considering their harmful nature (i.e. teratogenic, mutagenic, and carcinogenic) (Agamuthu *et al.*, 2016; Li *et al.*, 2018; Zhang *et al.*, 2018). The global production of PAEs escalated from 1.6 million tonnes in 1975 to more than 7 million tonnes in 2011, and they represent 70% of the world consumption of plasticizers (Rabodonirina *et al.*, 2015). These compounds are of great concern due to them not chemically bound to the host polymers which consequently being released gradually into the environment (Xu *et al.*, 2005). Recently, PAEs were observed to contaminate the water and sediment of rivers, such as the Pearl River, China (5,340 ng/L)(Liu *et al.*, 2014) and Moscow River, Russia (85 ng/L)(Eremina *et al.*, 2016), as well as, lakes, like the Epe and Lagos Lagoons in Nigeria (180 ng/L) (Adeogun *et al.*, 2015).

1.5 Problem Statement

Urbanization and industrialization pose significant threat to the wellbeing of river ecosystem. Because of their pivotal roles in ecological, human health, and in the economic development, it is essential to prevent and control declining river water quality, especially from the influx of microplastics of both primary and secondary sources.

It is indisputable that microplastics are notorious and that rivers transported significant amounts of these contaminants. Nevertheless, environmental data linked to the abundance of microplastics in freshwater environment have yet to be adequately addressed, as compared to that in the marine environment.

Microplastics studies in Malaysia have only been focusing on their concentration in marine environment that include beaches (Fauziah *et al.*, 2015; Noik & Tuah, 2015), mangrove areas (Jayanthi *et al.*, 2014), intertidal zone (Ismail *et al.*, 2009), core sediment (Matsuguma *et al.*, 2017), water samples from Kuala Nerus and Kuantan port (Khalik *et al.*, 2018), including microplastics ingestion in marine biota (Ibrahim *et al.*, 2016; Karami *et al.*, 2017).

The great concerns about microplastics are their association with toxic chemicals such as POPs and subsequent exposure of these chemicals to multiple kinds of organisms that ingest the microplastics (Rochman *et al.*, 2014). Furthermore, the leaching of POPs into the natural environment is also of rising concern as these contaminants readily bind to the particle fraction in waters and sediments that can later be taken up by organisms, consequently biomagnified. Additionally, there is an urgent need to look into POPs concentrations in freshwater environment as their concentrations are deemed to be higher than in marine environment, due to proximity to the use of these chemicals (Dris *et al.*, 2015). Thus, the study is hoped to benefit the regulatory agencies and policy makers with baseline information to bring in appropriate and effective managerial actions.

1.6 Objectives of Research

In order to develop appropriate policy and management tools to address the emerging issue of microplastics and POPs, comprehensive data on their abundance and distribution are crucially needed. Therefore, this study aimed to explore the abundance and distribution of microplastics and POPs in six selected rivers in Peninsular Malaysia. The objectives of this research are:

- To identify anthropogenic activities that may contribute to the presence of microplastics and POPs along selected rivers.
- 2. To characterize and determine the abundance and distribution of microplastics in river sediments and river water of Peninsular Malaysia.
- To determine the concentrations of POPs in river sediments and river water of Peninsular Malaysia.
- To analyze the relationship between microplastics abundance, concentrations of POPs and anthropogenic activities in selected rivers.

CHAPTER 2: LITERATURE REVIEW

2.1 Municipal Solid Waste (MSW)

Waste, either in solid, liquid or gaseous state, is well-defined as any unusable or unwanted substance or material (Golomeova *et al.*, 2013). MSW is defined as "the aggregate of the unwanted materials, which are generated from the daily activities of man as they interact with their environment, mostly waste from residences, commercial centres and institutions" (Ibikunle *et al.*, 2019). The consecutive sections entail the MSW generation, composition, treatment technologies, and the impacts associated from the illegal disposal of MSW.

2.1.1 MSW Generation

Solid waste has become a new threat to global sustainability in the last decades, due to the population explosion, along with the advancement of technological innovations, and profound changes in habits and lifestyle patterns (Lino & Ismail, 2018; Omari *et al.*, 2018). In 2016, the global solid waste reached 2.01 billion tonnes and it is anticipated that by 2050, 3.40 billion tonnes of solid waste will be generated (Indrawan *et al.*, 2018; Kaza *et al.*, 2018; Dalmo *et al.*, 2019). Globally, East Asia and the Pacific region are identified to generate the most, at 23% of 468 million tonnes annually (Figure 2.1) (The Economist, 2018).

Asia alone generates more than one million tonnes of MSW per day, and it is projected that the figure will surpass 1.6 million tonnes daily by 2025, making it the largest waste-producing continent on Earth (Hoornweg & Bhada-Tata, 2012).



Figure 2.1: Regional and per capita solid waste generation (The Economist, 2018).

On top of that, there has been a general trend regarding average MSW generation increase with nominal Gross Domestic Product (GDP) and the human population of a region or a country (Lee *et al.*, 2016; Cohen, 2017). The waste management trend increases at a slightly lower rate than GDP, but greater than that of population growth (Figure 2.2) (Simões & Marques, 2012).



Figure 2.2: MSW generation, GDP and population in OECD countries, 1980-2020 (Simões & Marques, 2012).

With a projected population of over 31.6 million in 2017, Malaysian generates more than 33,130 tonnes of MSW per day (Zainu & Songip, 2017), which surpassed the Government's waste generation projection of 30,000 tonnes/day by 2020 (Global Environment Centre, 2019). The average per capita MSW generation is approximately 0.85 kg/person/day, with roughly 1.5 kg/person/day in major cities such as Kuala Lumpur (Budhiarta *et al.*, 2012). Furthermore, MSW generation increased approximately 3% annually in the urban areas of Malaysia (Agamuthu & Tanaka, 2014).

In general, if solid waste is not effectively taken care of, it will result in grave environmental degradation (Johari *et al.*, 2012). It is noteworthy that an effective waste management depends on a thorough consideration of the waste composition (Taiwo, 2011). The subsequent section elaborates on the composition of MSW.

2.1.2 MSW Composition

Waste composition is defined as "the individual material fractions of the waste stream as a percentage of the total mass generated" (Dahlén & Lagerkvist, 2008). This information not only represents the basis of any waste management system planning and development, but is also crucial to establish baselines and evaluate the effectiveness of environmental policies (Edjabou *et al.*, 2017). Table 2.1 summarizes the waste types and their sources (Hoornweg & Bhada-Tata, 2012).

Ta	ble	2.1	1:	T	ypes	and	sources	of	waste	(H	loornweg a	&	Bhada	a-Tata	, 2	20	12).
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Туре	Sources
Organic	Food scraps, wood, yard (leaves, grass, brush) waste, process residues
Paper	Paper scraps, newspapers, cardboard, magazines, boxes, bags,
	wrapping paper, shredded paper, paper beverage cups, telephone books
Plastic	Bottles, containers, lids, packaging, cups, bags
Glass	Bottles, broken glassware, colored glass, light bulbs
Metal	Cans, tins, foil, non-hazardous aerosol cans, appliances (white goods),
	railings
Other	Textiles, multi-laminates, e-waste, appliances, ash, other inert materials

The MSW composition varies depending on the life style, economic development, culture, climate, and waste management regulations (Thitame *et al.*, 2010; Abdel-Shafy & Mansour, 2018). Generally, greater proportions of inorganic material fractions are observed in high-income countries, while the middle- and low-income countries generate greater organic material fraction (Figure 2.3) (Chen, 2018; Kaza *et al.*, 2018).



Figure 2.3: Waste composition (in percentage) by income level (Kaza et al., 2018).
Similar trend is anticipated in the year 2025 according to the World Bank estimates,

as tabulated in Table 2.2 (Kaza et al., 2018).

Table 2.2: Global waste composition by income level of 2025 estimates (Kaza *et al.,* 2018).

Income level	MSW Composition (2025 estimates) (%)					
	Organic	Paper	Plastic	Glass	Metal	Other
High	28	30	11	7	6	18
Upper-middle	50	15	12	4	4	15
Lower-middle	55	10	13	4	3	15
Low	62	6	9	3	3	17

In Asia, organic material dominates by 75% from the total waste stream (Johari *et al.,* 2014). In Malaysia specifically, roughly 45% from the 25,000 tonnes per day of waste generation in Peninsular Malaysia is food waste (Figure 2.4) (Zainu & Songip, 2017).



Figure 2.4: Composition of MSW reported in Peninsular Malaysia (Zainu & Songip, 2017).

2.1.3 MSW Treatment Technologies

Rapid population growth coupled with rampant urbanization and industrialization represent a global challenge towards instigating environmentally sound MSW management, especially in rapidly growing cities (Samsudin & Don, 2013; Ramachandra

et al., 2018). The capability to reduce waste volumes, as well as, the potential to efficiently manage the waste, with minimal health and environmental impacts, are the key factors to be considered in choosing the right treatment technique (Liu *et al.*, 2017). On top of that, economic viability and social acceptability of the systems, are among the other criteria that need to be looked into, besides considering the improvement of energy and material recovery (Ohnishi *et al.*, 2018; Sebastian *et al.*, 2019).

MSW is generally managed in one of the three ways: landfilling, biological treatment, or thermal treatment (Hong *et al.*, 2017). Globally, 40% of waste is landfilled with 33% of waste is still openly dumped (Figure 2.5) (Kaza *et al.*, 2018). Moreover, approximately 11% of waste is treated through modern incineration with 19% of waste undergoes materials recovery through recycling and composting (Chen, 2018).





The most common treatment and disposal method practiced in a particular country is largely associated with the country's economic development. Figure 2.6 delineates the global MSW treatment and disposal methods, by income level (Statista, 2019). 93% of waste is openly dumped in low-income countries while landfilling is commonly utilized in upper-middle-income countries, at 54% (Agamuthu *et al.*, 2007). However, the rate of landfills declines to 39% in high-income countries, with 22% of waste being incinerated, and with 35% waste diversion to recycling and composting (Kaza *et al.*, 2018).



Figure 2.6: Global MSW treatment and disposal methods, by income level (Statista, 2019).

Landfills that are not adequately controlled and managed such as unsanitary open disposal sites, use up valuable land and poses significant risks to environment and human health (Chen, 2018). Yet, only a mere of 8% of waste is disposed in sanitary landfills (Vodyanitskii, 2016).

In Malaysia, out of 269 operating landfills in Malaysia, only 153 are sanitary (Table 2.3) (Ministry of Housing & Local Government, 2019). Additionally, 74 of the landfills nationwide or 49% of them are expected to reach the end of their lifespan by 2020 (The Star, 2019).

State	Operating Landfill		Non-operating
	Sanitary	Non-sanitary	- Landfill
Johor	12	9	25
Kedah	7	4	8
Kelantan	11	11	9
Malacca	1	0	7
Negeri Sembilan	5	3	14
Pahang	12	4	20
Perak	16	13	15
Perlis	1	0	2
Penang	2	0	1
Sabah	22	21	4
Sarawak	46	43	20
Selangor	8	2	15
Terengganu	10	6	11
Federal Territory	-	0	10
Total	153	116	161

Table 2.3: Current status of MSW disposal sites in Malaysia (Ministry of Housing &Local Government, 2019).

Biological treatment of MSW on the other hand is deemed to be more environmentalfriendly, but somewhat consumes more time to degrade than thermal treatment (Tozlu *et al.*, 2016). Thermal treatment such as incineration, gasification, and pyrolysis can significantly reduce waste mass and volume by about 70% to 80% and 80% to 90%, respectively, and are primarily employed in high-capacity and land-constrained countries (Abd Kadir *et al.*, 2013).

Incineration has been widely employed in recent times. However, it may not always be feasible as it largely depends on waste characteristics, which in turn, is influenced by the local demography, social status and cultural differences, including seasonal fluctuations and topography (Rajaeifar *et al.*, 2017; Sebastian *et al.*, 2019). In addition, incineration plants that lack adequate control strategy to keep the emissions of dioxins and furans below the allowable limits, can lead to adverse health impacts (Aniekan & Ikechukwu, 2016; Indrawan *et al.*, 2018).

As in Malaysia, there were five small scale incinerators (i.e. <100 tonnes capacity) namely in Pulau Langkawi of 91 tonnes/day, Cameron Highlands at 36 tonnes/day, Pulau Pangkor at 18 tonnes/day, Pulau Tioman at 9 tonnes/day, and Labuan at 54 tonnes/day (Aja & Al-Kayiem, 2014). Nevertheless, on account of faulty design, poor maintenance, improper operation, as well as, high diesel usage on grounds of high moisture content nature of MSW in Malaysia, these incinerators are no longer operating (Jereme *et al.,* 2013).

Waste issues can be resolved when they are reused and recycled and are channeled as raw materials for other production processes towards safeguarding the limited natural resources, otherwise referred to as zero waste (Ayeleru *et al.*, 2018). It also emphasizes the industries to redesign their products so that wastes can be eradicated in the production processes (Allen *et al.*, 2012). Countries such as Japan, Denmark, Germany, Netherlands, Sweden, Belgium and Switzerland disposed less than 3% of MSW in sanitary landfill, incinerated more than 35% and recycled more than 40% of their waste (Lino & Ismail, 2018).

It is noteworthy that every treatment technology has its own pros and cons that it is of vital importance for the decision-makers to make thorough consideration in managing the waste in the best possible ways. In the absence of that, it is impossible to monitor, control and improve waste management system that minimizes the peril it may pose (Palanivel & Sulaiman, 2014). The subsequent section deliberates the illegal disposal of MSW and its consequent impact to the environment.

2.1.4 Illegal disposal of MSW and its Impact to the Environment

Incessant increase in waste generation owing to the economic, demographic and technological advancement of the community has indisputably led to disposal problems in many areas of the world (Aniekan & Ikechukwu, 2016). This is partly due to the fact

that these advancements create larger population centres, making the collection of all waste and the securing of land for treatment and disposal more and more challenging (Kaza *et al.*, 2018). Figure 2.7 depicts the global proportion of MSW that is collected (The Sustainable Development Goals Report, 2018).



Figure 2.7: Proportion of collected MSW (%) (The Sustainable Development Goals Report, 2018).

In addition, report from WHO/UNICEF validates that many cities in the world have yet to achieve adequate solution for their solid waste (WHO *et al.*, 2015). According to Kaza *et al.* (2018) 93% of waste generated in low-income countries has been reported to be illegally dumped or burnt on roads, open land, or waterways, which are environmentally unsafe. Figure 2.8 illustrates the estimated quantity of waste burned by country, residentially and in dumps (Wiedinmyer *et al.*, 2014). Illegal disposal and littering are still very common in Malaysia, in which according to the Ministry of Housing and Local Government, 60% of the 32 million Malaysian citizens fail to dispose their waste into the provided trash bins (The Sun Daily, 2019).



Figure 2.8: Estimated quantity of waste burned by country, residentially (A) and in dumps (B) (Wiedinmyer *et al.*, 2014).

These indiscriminate disposals of MSW have led to a plethora of environmental problems such as infestation of pests, contamination of land, water and soil environment due to the leaching of nutrients, while contaminating the air through the emission of greenhouse gases (Lino & Ismail, 2018; Ibikunle *et al.*, 2019). Apart from that, the World Bank Group (2019) has expressed their apprehensions on the never-ending threats from illegal disposal of MSW, that goes;

"Illegal disposal from poorly managed waste is contaminating the world's oceans, clogging drains and causing flooding, transmitting diseases, increasing respiratory problems from burning, harming animals that consume waste unknowingly, and affecting economic development, such as through diminished tourism."

In recent years, the problem of river pollution is becoming more and more critical primarily due to the illegal disposal of MSW, on grounds of rapid growth in human population, industrial production, and commercial activities (Wang *et al.*, 2012). Rapid

urbanization has always been accompanied by river pollution, due to its incongruity with the developments of efficient MSW management (Maroušek *et al.*, 2019; Zhang *et al.*, 2019). The following sections further discuss the issue of river pollution.

2.2 River Pollution

Rivers have been a major part in human life for millennia, that perform a suite of ecological functions including habitat, water supply, shielding effects, as well as, transport routes, aquaculture, tourism and recreation (Kong *et al.*, 2016; Zhuang *et al.*, 2018). For instance, the Malaysian rivers and their tributaries support an immense diversity of aquatic biodiversity, including more than 600 freshwater fish species (UNDP, 2019).

Being strongly linked to human activities, as well as, through natural processes, river pollution is not something not unheard of. In fact, it is a worrying phenomenon that has become a worldwide concern (Zhao *et al.*, 2019b). The subsequent sections deliberate on the sources of river pollution and its impact to the environment.

2.2.1 Sources of River Pollution

UNEP documented that pollution started when humans began to farm the land and settle in villages and towns many thousands of years ago (Ara, 2003). With rapid urbanization, the area allocated for irrigation has doubled since the 1960s to more than 320 million hectares, the number of livestock has tripled since the 1970s to over 24.2 billion, and the aquaculture has grown more than twenty-fold since the 1980s (Evans *et al.*, 2019; FAO, 2019).Without proper management and operation, it is indubitable that rivers worldwide have been significantly disrupted (Deng *et al.*, 2016).

The water quality of rivers is characterized by high level of heterogeneity in time and space (Al-Badaii *et al.*, 2013). In general, river pollutants can be broadly classified into

organic, inorganic, radioactive and acid/base, of anthropogenic or natural origin (Quesada *et al.*, 2019). Additionally, their concentrations are typically subjected to seasonal variations, due to the seasonality of precipitation, surface runoff, interflow and baseflow (Xu *et al.*, 2018).

River pollution is attributed to either two root sources namely point, and non-point source of pollution (Maschal & Truye, 2018). Point source include specific sources such as drain pipes, oil wells, ditches or sewer outfalls, as well as, effluent from municipal sewage treatment plant, industries, refineries, and underground coal mines (Wu *et al.,* 2013). Another significant point source pollution is waste dumping into rivers or the river banks (Figure 2.9), which is a criminal offence that in the most severe cases in the United Kingdom, it can attract a maximum fine of £50,000 or a five-year jail term (UK Environmental Law Association, 2017).



Figure 2.9: Waste clogging up Marilao River in Manila, the Philippines from river dumping of waste (Photo credit: AFP Photo/Noel Celis).

Non-point or diffuse source on the other hand does not originate from a statutory point source, which enter the riverine system from soils or groundwater systems, and from the

atmosphere via rainwater (Lai *et al.*, 2011). Soils and groundwater may contain the residue of agricultural practices, such as uncontrolled spreading of slurries and manure, disposal of sheep dip, tillage, ploughing of land, use of pesticides and fertilizers, whereas, atmospheric pollutants could be derived through gaseous emissions from automobiles and factories (Hari, n.d.). In general, these spatially-dispersed loads are more difficult to be identified, isolated, and controlled, as compared to point source pollution (Ouyang *et al.*, 2009).

Approximately 2,200 tonnes of rubbish, equivalent to the weight of more than 300 adult African elephants, is dumped into Malaysian rivers, drains and waterways every month (The Straits Times, 2016), making them a very serious source of pollution to rivers. The major point source pollution affecting the Malaysian rivers are sewage disposal, and discharges from small- and medium-sized industries which are not equipped with proper effluent treatment facilities, while non-point sources mainly coming from land clearing and earthworks activities (Juahir *et al.*, 2011).

2.2.2 River Pollution and its Impact to the Environment

In recent decades, almost 60% of all river basins around the world have been impacted simultaneously by human activities (Jia & Chen, 2013; Kong *et al.*, 2016). This phenomenon has strongly affected their quantity, morphology and structure, which have been documented to decrease in intensity from the city to the suburbs (Deng *et al.*, 2016).

Furthermore, the emissions from point and non-point pollution sources are deteriorating river water quality and biological habitat fragility, as well as, exerting disturbance in the hydrological cycles, that have led to serious water resource problems (Pan *et al.*, 2015; Zhao *et al.*, 2019b). Once discharged, river pollutants may be disseminated in the water, sediments and biota of aquatic systems, consequently bioaccumulated and biomagnified via the food chain (Zhao *et al.*, 2013); Ali *et al.*, 2016).

Researchers have documented the connections between river water pollution and acute water-borne diseases which include cholera, dysentery, hepatitis, cryptosporidiosis, diarrhoea, giardiasis, and typhoid, including the risk of carcinogenic diseases (Roushdy *et al.*, 2012; Lu *et al.*, 2015). Astonishingly, about 2.3 billion people globally are suffering from water-borne diseases and among them, 2.2 billion people live in developing countries, with 1.8 million people, mostly children die due to waterborne diseases every year (Duflo *et al.*, 2015).

Serious issues in river environments have posed great challenge and have gradually hinder social and economic development (Bocaniov & Scavia, 2016; Luo *et al.*, 2019). Therefore, it is of vital importance for the land use activities to be carefully planned and controlled, on account of protecting the water resource and quality status (Al-Badaii *et al.*, 2013). One promising way to deal with such complex problem is to consider them integratedly, bringing together all engineers, participant planners, social and natural scientists, landscape architects, local officials, and general public for the common good (Su *et al.*, 2011a).

There is no doubt that river pollution today is primarily attributed by the incessant increase in global plastic waste generation, that results in the accumulation of plastic waste in rivers at large amounts (Castro-Jiménez *et al.*, 2019). Material composition consisting of up to 95% of plastics has been recorded in nearly every aquatic ecosystem, including many freshwater rivers and lakes (Eerkes-Medrano *et al.*, 2015; Auta *et al.*, 2017). Further discussion on plastic issue is deliberated in the following sections.

2.3 Plastics

The word "plastic" comes from "plasticus," which is a Latinization of the ancient Greek adjective "plastikos" (fit for moulding) (Macionis, 2018). They are the synthetic organic polymers, which are low-cost, lightweight, and durable, with high strength-toweight ratio (Wang *et al.*, 2019). Synthetic plastics have been invented in the early 20th century but has only been widely used after the World War II in 1950s (Geyer *et al.*, 2017).

Today, various economic activities as diverse as packaging, construction, transportation, healthcare and electronics depend heavily on plastics as raw materials, including industries like the airplane manufacturers that use up to 50% by weight of plastic content in their products (Andrady & Neal, 2009; Milios *et al.*, 2018). The subsequent sections discuss the plastic production and composition, followed by their management technologies, and impacts from the mismanagement of plastic wastes.

2.3.1 Plastic Production and its Composition

Since 1954, plastic production has increased by 20-fold, reaching 282 million tonnes in 2014, and is expected to quadruple by 2050, to be over one billion tonnes/year (Figure 2.10) (Ryan, 2015). The global shift from reusable to single-use products has put packaging sector to be the largest market of plastics (Dahlbo *et al.*, 2018). The most common single-use plastics found in the environment are cigarette butts, plastic drinking bottles, plastic bottle caps, food wrappers, plastic grocery bags, plastic lids, straws and stirrers, other types of plastic bags, and foam take-away containers (Hopewell *et al.*, 2009). These are the waste products of a throwaway culture that treats plastic as a disposable material rather than a valuable resource to be captured, re-used, and repurposed (IUCN Water Programme, 2015).



Figure 2.10: Global plastic production and future trends (Ryan, 2015).

Plastics cover an extensive range of synthetic polymeric materials, producing large spectrum of different final products, as tabulated in Table 2.4 (Hanvey *et al.*, 2017). Polyethylene, polypropylene and polystyrene are the main types of polymers detected in the environment (Leal *et al.*, 2019).

Plastic polymer	Applications	Density (g/cm ³)
PET/PETE	Food packaging, disposable beverage bottles, textiles (synthetic fibers), tape, thermal insulation	1.37–1.38
HDPE	Bottle caps, plastic lumber, fuel tanks, milk crates	0.93–0.97
PVC	Inflatable products, plumbing pipes, door and window frames, garden hoses, electrical cable insulation	1.10–1.47
LDPE	Plastic bags and, six-pack rings, flexible snap-on lids	0.91-0.92
PP (expanded or nonexpanded)	Bottle caps, rope, carpet	0.89–0.92
PS (expanded or nonexpanded)	Disposable cutlery, dinnerware, and take- away food packaging, building insulation, refrigerated bins (e.g., fish boxes)	0.28–1.04
Other resins, such as polycarbonate, nylon, and acrylic	Used for engineering purposes because of their thermal, electrical and chemical properties. e.g., electrical wire insulation	1.15–1.22

Table 2.4: The types of plastic and their common usage (Hanvey et al., 2017).

2.3.2 Plastic Waste and its Management Technologies

Plastic is one of the greatest environmental challenges facing the world. Globally, the rising of living standard highly attributed to rapid urbanization have led to a paramount increase in plastic waste generation, intensified by rapid population growth (Minghua *et al.*, 2009). In 2015 alone, more than 272 million tonnes of plastic waste were generated (Figure 2.11) comparable to the weight of 13 million adult blue whales (Geyer *et al.*, 2017). Among them, 94% are thermoplastics that can be recycled, and 6% are thermoset plastics that are non-recyclable (Aryan *et al.*, 2019).



Figure 2.11: Global primary plastic waste generation (1950 – 2015) (Geyer et al., 2017).

Figure 2.12 illustrates the global flow of plastic waste (World Economic Forum, 2018). Of the plastic waste generated, merely 14% was recycled while the remaining significant portion ends up in landfills or littered.

The recycling of plastics is urged by the need for closing material loops in order to minimize the pressure on utilization of natural resources, as well as, to reduce the negative impacts to the environment from littering of plastic waste (Dahlbo *et al.*, 2018). Plastic recycling can be primarily categorized into mechanical recycling and feedstock recycling.



Figure 2.12: Flow of plastic waste worldwide (World Economic Forum, 2018).

Mechanical recycling is applicable to all types of plastics which involves the recovery of products without significant alteration of its molecular structure or physical properties and to be reused for a similar purpose, whereas, feedstock recycling involves structural and molecular level changes to the plastic and conversion to its raw material or feed material to be used in a different application (Vanapalli *et al.*, 2019).

Overall, the areas that need further development across countries include better management of plastic waste, not to forget innovation in handling these wastes effectively. According to Erik Solheim, the Head of UN Environment:

"Plastic is not the problem. It is what we do with it. Changes in consumer behaviour will go a long way towards reducing plastic pollution"- Erik Solheim.

2.3.3 Mismanagement of Plastic Waste and its Impacts to the Environment

The advantages that plastics possess are causing them to accumulate at alarming levels, producing major global environmental concerns (Rodríguez *et al.*, 2018). Additionally, due to them requiring thousands of years to degrade, they become a serious source of pollution, posing detrimental impacts to human and the environment (Papong *et al.*, 2014). In 2010, 2 to 5% out of 249 million tonnes of plastic waste generated were mismanaged plastic waste (Jambeck *et al.*, 2015; Cho *et al.*, 2019). Globally, an

approximate 79% of plastic waste generated are accumulating in landfills or the natural environment around the world, mostly in Africa (57%), Asia (40%) and Latin America (32%) (Geyer *et al.*, 2017) (Jang *et al.*, 2018b).

Littering of plastic waste reduces the water permeability of soils, affecting soil fertility, and may also result in the blockage of drainage systems (Saikia & Brito 2012; Sharma & Bansal, 2016). Most importantly, unsound waste management practices which fail to effectively treat or contain plastic materials generally result in litter that ends up in oceans as marine plastic debris (Figure 2.13) (UNEP, 2014). Globally, the marine environment has become the end route to between 6 and 10% of plastics produced (Jambeck *et al.,* 2015; Troost *et al.,* 2018).



Figure 2.13: The accumulation of plastic waste at a port in Semporna, Sabah that will eventually ends up in the ocean (Photo credit: Rich Carey/Shutterstock).

Furthermore, it is worth noting that 70 to 80% of plastics entering marine environment were reported to be transported through rivers (Bowmer & Kershaw, 2010). As plastic is discarded into our waterways, rivers become conveyor belts of plastic debris, transporting

this dangerous and toxic cargo into the world's estuaries, deltas and oceans. Astoundingly, between 1.15 and 2.41 million tonnes of plastics are currently flowing from the global riverine system into the oceans every year, with top 20 polluting rivers mostly located in Asia (i.e. annual input of 1.21 million tonnes per year) (Table 2.5) (Lebreton *et al.*, 2017). A considerably high population density (i.e. 60% of the global population) combined with episodes of heavy rainfalls have resulted in Asia being the dominant contributor of plastic wastes in the oceans (Rochman *et al.*, 2016).

Country	River	Yearly average discharge (m ³ /s)
China	Yangtze	1.58 x 10 ⁴
China	Xi	5.53×10^3
China	Huangpu	$4.04 \ge 10^2$
China	Dong	8.54 x 10 ²
China	Zhujiang	1.33×10^2
China	Hanjiang	7.35×10^2
Indonesia	Brantas	8.18 x 10 ²
Indonesia	Serayu	3.70×10^2
Indonesia	Solo	$7.46 \ge 10^2$
Indonesia	Progo	2.79×10^2
India, Bangladesh	Ganges	2.08×10^4
Philippines	Pasig	$2.07 \text{ x } 10^2$
Myanmar	Irrawaddy	5.49×10^3
Thailand, Cambodia, Laos,	Mekong	6.01×10^3
China, Myanmar, Vietnam	_	
Taiwan	Tamsui	$1.08 \ge 10^2$
Nigeria, Cameroon	Cross	$2.40 \ge 10^2$
Nigeria	Imo	$2.79 \ge 10^2$
Nigeria	Kwa Ibo	1.92×10^2
Brazil, Peru, Columbia,	Amazon	$1.40 \ge 10^5$
Ecuador		
Colombia	Magdalena	5.93×10^3

Table 2.5: Top 20 polluting rivers as predicted by the global river plastic inputs model (Lebreton *et al.*, 2017).

Growing scientific literatures have clearly reported the threats posed to wildlife and the ecosystems, from the occurrence of plastic debris (Leal *et al.*, 2019). The impacts vary from entanglement and ingestion, to bio-accumulation and bio-magnification of toxics, either released from plastic items or adsorbed on the plastic particles, as well as, damages it caused to benthic habitats and communities (Richards & Beger, 2011; Gall & Thompson, 2015; Fossi *et al.*, 2018).

Plastic pollution can be subdivided into five basic categories; megaplastic (> 100 mm), macroplastic (> 20 mm), mesoplastic (20–5 mm), microplastic (< 5 mm) and nanoplastic (< 0.001 mm) (Barnes *et al.*, 2009). Of the five categories, microplastics account for 92% of the global plastic pollution (Eriksen *et al.*, 2014). Due to the ubiquitousness of microplastics, the following sections elaborate on their sources, abundance, impacts and regulations pertaining to microplastics.

2.3.4 Microplastics

The succeeding sections deliberate the origins of microplastics, their abundance and distribution in freshwater environment, fate and impacts, the analytical methods in monitoring microplastics pollution, and the global initiatives in tackling this issue.

2.3.4.1 Origins of Microplastics

Microplastics are released into the environment from either primary or secondary source (Andrady, 2011; Cole *et al.*, 2011). Primary source is the direct input of manufactured microsized plastic particles (primary microplastics), like personal care products in the form of microbeads (e.g., exfoliating facial cleansers, cosmetics) and commercial cleaning abrasives (Fendall & Sewell, 2009; Duis & Coors, 2016), plastic production pellets released through unintentional spills (Costa *et al.*, 2010) and the release of microfibers from synthetic textiles due to in-use wear and from fiber-containing laundry effluents (Browne *et al.*, 2011).

Approximately 1,900 synthetic fibers may be shed from one synthetic garment during each washing cycle and unfortunately most wastewater treatment plants fail to retain and eliminate microplastics of this form (Van Cauwenberghe *et al.*, 2015). A recent study

revealed that roughly 35% of primary microplastics that end up in the marine environment originated from fibers released from washing of synthetic clothes (Boucher & Friot, 2017; De Falco *et al.*, 2018).

Microplastics of secondary source are generated through the breakdown of larger plastic debris items into smaller pieces, so-called secondary microplastics (Rochman *et al.*, 2013), through environmental weathering processes from biological activities, UV radiations, mechanical abrasions, temperature fluctuations, wind and wave actions (Auta *et al.*, 2017; Ling *et al.*, 2017; Pan *et al.*, 2019).

On top of that, some scholars have summarized the main sources of microplastics which have been shown to correspond to certain morphological features and chemical compositions (Cole *et al.*, 2011; Hüffer *et al.*, 2017). A detailed level of classification based on both morphological and chemical composition of microplastics, may more precisely reflect their origins and usage, indirectly identifying their sources (Table 2.6) (Helm, 2017; Wang *et al.*, 2019).

Туре	Characte	Characteristics	
	Shape	Polymer composition	
Microbead	Spherical or	PE (primary),	Personal care consumer
	irregularly spherical	PMMA, PTFE,	products
		PP, nylon, PS, and	
		PET	
Pellet	Pellets (spherical,	PE, PP, PS, PVC,	Spilled or recycled raw
	ovoid or disk-shaped)	PC and so on	material
Film	Flexible film	PE, LLDPE,	Plastic bags and wrappers
		LDPE, HDPE, and	Agricultural film
		so on	Film for industrial or
			construction applications

Table 2.6: Source-specific classification system of microplastics in aquatic ecosystems (Helm, 2017; Wang *et al.*, 2019).

Туре	Characteristics		Source
	Shape	Polymer composition	
Fiber	Fiber/line	Polyester (PET), acrylic, PA, PVC,	Textile fibers from sewage or surface runoff
		PAN, PAM,	Ropes/line/net (mainly
		PE/LDPE, PP, PP-	used in fisheries for
		PE and so on	aquatic ecosystems)
			Other synthetic fiber
			Ropes/line/net (mainly
			used in fisheries for
			aquatic ecosystems)
			Fabric fibers from sewage
			Other synthetic fiber
Foam	Foam plastics (Styrofoam)	PS/EPS	Packing material (food containers)
			Foam floats or buoys used
			in fisheries
			Insulation board or thermal
			insulation products

Table 2.6, continued.

*PE: Polyethylene, PMMA: Polymethyl methacrylate, PTFE: Poly tetra fluoroethylene, PP: Polyethylene, PS: Polystyrene, PET: Polyethylene terephthalate, PA: Polyamide, PVC: Polyvinyl chloride, PAN: Polyacrylonitrile, PAM: Polyacrylamide, PC: Polycarbonate, LDPE: Low-density polyethylene, LLDPE: Linear low-density polyethylene, HDPE: High-density polyethylene.

Microplastics once in the environment can be transported via wind, commercial and domestic discharges to sewers, runoff into rivers, runoff into combined sewer systems and runoff directly into lakes and oceans (Figure 2.14) (Horton *et al.*, 2017).

Additionally, microplastics in poor-mobility environmental media, such as soil, could persistently exist for more than 100 years, resulting in great accumulation in the environment (Hu *et al.*, 2019a). Next sub-section deliberates the microplastics abundance in freshwater environment worldwide.



Figure 2.14: Possible environmental transport of microplastics (Horton et al., 2017).

2.3.4.2 Global Abundance and Distribution of Microplastics in Freshwater Environment

To date, microplastics have been found in freshwater environment worldwide, from developed urbanized areas (McCormick *et al.*, 2014) to remote mountain lakes such as the Lake Hovsgol, Mongolia (Free *et al.*, 2014), Qinghai-Tibet plateau, China (Xiong *et al.*, 2018) and subalpine lakes in Italy (Imhof *et al.*, 2013). Surprisingly, Lake Hovsgol has appeared to be more polluted with microplastics (i.e. 20,264 items/km²), than the more developed and densely populated Lake Huron (2,779 items/km²) and Lake Superior (5,391 items/km²) (Eriksen *et al.*, 2013).

Microplastics pollution varies geographically with locations (Fossi *et al.*, 2012, De Lucia *et al.*, 2014). Factors affecting the transportation and distribution pattern include large-scale forces such as currents driven by wind and geostrophic circulation (Law *et al.*, 2010), and turbulences (Turra *et al.*, 2014), as well as, the inherent properties of microplastics such as the density, shape and size (Eerkes-Medrano *et al.*, 2015). The aforementioned factors are more likely to play important role in larger freshwater environment like riverine systems. However, they become limited on smaller isolated

freshwater systems, where natural factors and long water residence time dominantly affect microplastics abundance (Free *et al.*, 2014).

Most of microplastics pollution studies in freshwater environment have been performed in Europe and North America (67%) with only a few studies reported for Asia (most of them in China; 16%), South America (Brazil, Argentina, Colombia and Chile; 11.8%), Africa (South Africa and Tanzania; 4%) and Australia (2%). Although many of plastic polluted rivers are in Asia, only 14% of the reviewed microplastics studies were carried out in this continent (Blettler *et al.*, 2018).

A recent review indicated that microplastics in freshwater environment were found to be greater than 1 million item/m³ (Li *et al.*, 2018). Quantitative data on microplastics abundance across continents are presented according to different units employed (items/kg in Table 2.7 and items/m³ in Table 2.8) (modified from Fauziah *et al.*, 2018).

In Malaysia primarily, there has been only one study concerning the monitoring of microplastics in freshwater environment, henceforth making it challenging to understand the extent of microplastics pollution. It was a preliminary analysis conducted in the sediment of urban rivers in Johor, specifically the Skudai and Tebrau rivers (Sarijan *et al.*, 2018). Both were observed to be polluted with microplastics, whereby Tebrau River reported greater concentration of 680 ± 140 particles/kg, as compared to Skudai River of 200 ± 80 particles/kg.

Continent	Country	Location	Concentration (items/kg)	Sample type	Composition	References
Asia	China	Shanghai	802 ± 594 (dw)	Sediment	PP, PE, rayon, cotton + viscose, phenoxy resin, poly(vinyl stearate), 76% rayon + 24% PES	Peng et al. (2018)
Asia	China	Three Gorges Reservoir	25 to 300 (ww)	Sediment	PE, PP, PS	Di & Wang (2018)
Asia	China	Beijiang River	178 ± 69 to 544 ± 107	Sediment	PE, PP, Copolymer, Paint particle	Wang <i>et al.</i> (2017a)
Asia	China	Taihu Lake	11.0 - 234.6 (dw)	Sediment	CP, PET, PE, PA, PP	Su et al. (2016)
Europe	United Kingdom	Edgbaston Pool, Birmingham	250 - 300	Sediment	NA	Vaughan <i>et al.</i> (2017)
Europe	Netherlands	Meuse River	1400 dw	Sediment	NA	Leslie <i>et al.</i> (2017)
Europe	Italy	Lake Chiusi	234 dw	Sediment	PE, PP, PET, PVC	Fischer <i>et al.</i> , (2016)
Europe	United Kingdom	River Thames Basin	660	Sediment	PP, PES, PET, PS, PE, PVC, Polyarylsulphone	Horton <i>et al</i> . (2017)
North America	Canada	Ontario Lake	760 (dw)	Tributary sediment	PE, PS, PU, PP, PVC, PSS, PET, PMMA, polyvinyl/vinyl acetate copolymer, PMMA-PS copolymer or mixture, ABS, Nylon, phenoxy/epoxy Resin, Polymethylsiloxane (silicone)	Ballent <i>et al.</i> (2016)

Table 2.7: Several relevant studies on microplastics abundance in freshwater matrices across continents (items/kg).

CP-Cellophane: PS – Polystyrene: PA – Polyamide: PES - Polyester: PP – Polypropylene: PE – Polyethylene: PET - Polyethylene Terephthalate: PVC - Polyvinyl Chloride: ABS - Acrylonitrile Butadiene Styrene: PSS: Poly(Styrenesulfonate): dw – dry weight: ww – wet weight.

Continent	Country	Location	Concentration (items/m ³)	Sample type	Composition	References
Asia	China	Three Gorges Reservoir	1597 to 12,611	Water	PE, PP, PS	Di & Wang (2018)
Asia	China	Hanjiang River	2933 ± 305.5	Water	PA, PE, PET, PP, PS	Wang <i>et al.</i> (2017b)
Asia	China	Yangtze River	2516.7 ± 911.7	Water	PA, PE, PET, PP, PS	Wang <i>et al.</i> (2017b)
Asia	China	Sha Lake	6390 ± 862.7	Water	PA, PE, PET, PP, PS	Wang <i>et al.</i> (2017b)
Asia	China	Nantaizi Lake	6162.5 ± 537.5	Water	PA, PE, PET, PP, PS	Wang <i>et al.</i> (2017b)
Asia	China	Nan Lake	5745 ± 901.6	Water	PA, PE, PET, PP, PS	Wang <i>et al.</i> (2017b)
Asia	China	Taihu Lake	0.0034 - 0.0258	Water	CP, PET, PE, PA, PP	Su et al. (2016)
Europe	United Kingdom	Itchen River	1155	Water	PE, PP, CP	Gallagher <i>et al.</i> (2016)
Europe	Italy	Lake Chiusi	2.68 to 3.36	Water	PE, PP, PET, PVC	Fischer <i>et al.</i> , (2016)
Europe	Austria	Danube River	0.3168 ± 4.6646	Water	NA	Lechner et al. (2014)
Europe	Sweden	Lysekil	8.25 ± 0.85	WWTP effluent	PE, PP	Magnusson & Norén (2014)
North America	USA	Los Angeles River	12,932	Water	NA	Moore <i>et al.</i> (2011)
North America	USA	WWTPs across United States	0.00005 ± 0.000024	Effluent	NA	Mason <i>et al.</i> (2016)
North America	USA	San Gabriel River	411	Water	NA	Moore <i>et al.</i> (2011)

Table 2.8: Several relevant studies on microplastics abundance in freshwater matrices across continents (items/m³).

PE – Polyethylene: PP - Polypropylene: CP-Cellophane: PS – Polystyrene: PA – Polyamide: PET - Polyethylene Terephthalate

2.3.4.3 Fate and Impacts of Microplastics

Adverse impacts resulting from microplastics exposure have been observed in several freshwater animals which include invertebrate and vertebrate species such as *Corbicula fluminea* (Asian clams) (Su *et al.*, 2016), *Daphnia magna* (crustacean) (Pacheco *et al.*, 2018, Martins & Guilhermino, 2018), and *Danio rerio* (zebrafish) (Lei *et al.*, 2018). For instance, *Daphnia magna* was found to ingest 2 μ m/L when exposed to 20 nm and 1000 nm of fluorescent polystyrene (Rosenkranz *et al.*, 2009). Similarly, in a laboratory assessment of freshwater invertebrates, five species were found to have ingested microplastics (Imhof *et al.*, 2013).

Alarmingly, microplastics are ingested more commonly and are available to a wider variety of species, as compared to macroplastics, due to their smaller dimensions (Possatto *et al.*, 2011; Slootmaekers *et al.*, 2019). Ingestion of microplastics can be found in almost all trophic levels and once ingested, aquatic organisms are exposed to numerous toxicity effects derived from the microplastics (Wright *et al.*, 2013). Table 2.9 delineates the potential toxicological effects of microplastic that impact the freshwater species (modified from Strungaru *et al.*, 2019).

For example, the presence of microplastics in the digestive tract may inhibit nutrient absorption and reduce; (i) consumption of resources, (ii) growth, (iii) reproduction, and (iv) survival (Lee *et al.*, 2013; Au *et al.*, 2015; Lei *et al.*, 2018). Due to large surface area and intrinsic hydrophobicity, the potential of hydrophobic chemical adsorption onto the surface of microplastics has caused great concern (Horton *et al.*, 2017). Microplastics may harbour POPs and other xenobiotic pollutants that adsorb onto their surfaces, thereby providing routes for secondary toxicity (Besseling *et al.*, 2013; Ziccardi *et al.*, 2016), and potentiating the effects of toxic chemicals (Syberg *et al.*, 2017).

Organism	Microplastic	Exposure duration	Interactions and toxicological effects	Reference
	concentration			
Danio rerio	5, 50, 500 μg/L	10 and 20 days	Present in intestinal lumen but with no	Karami <i>et al.</i> (2017)
(zebrafish larvae)			morphological changes	
Danio rerio	20 μg/L, 200 μg/L,	3 weeks	 Inflammatory responses; 	Lu et al. (2016)
(zebrafish adults)	2000 µg/L		• Increased the oxidative stress;	
			• Low feeding activity;	
			• Affected the lipid metabolism;	
			Reducing energy;	
Pomatoschistus microps	-	24 h	Reduction of the predatory performance and	De Sá et al. (2015)
(common goby juvenile)		• •	efficiency;	
Chironomus tepperi	500 MP/kg	10 days	• Significant decreased survival;	Ziajahromi et al.
(rice midge larvae)			• Reduction of adult emerged number;	(2018)
Corbicula fluminea	2.8, 3.2, 4.1 and	28 days	Histological changes in digestive glands;	Rochman et al. (2017)
(Asian clam)	4.2 mg/L		• Severe tubular dilatation in combination of	
			all types;	
Gammarus fossarum	100, 540, 2680,	24 h	Longer exposure time was responsible for	Blarer & Burkhardt-
(freshwater amphipod)	13,380 fibers/cm		assimilation efficiency reduction and body	Holm (2016)
			mass reduction	
Daphnia magna	12.5–400 mg/L	96 h	1 μm were easily ingested in guts and were	Rehse et al. (2016)
(water fleas)			responsible for immobilization of daphnids	
Chlorella pyrenoidosa	10, 50 and 100 mg/L	30 days	Algal growth inhibition;	Mao <i>et al.</i> (2018)
(fresh water algae)			• Decreasing of photosynthetic parameters	
			activity and cell wall thickness	

Table 2.9: Overview of the potential toxicological effects of microplastic particles in freshwater species, from 2015 to 2019.

2.3.4.4 Analytical Methods in Monitoring Microplastics Pollution

Numerous studies on microplastics occurrence have been conducted globally with various methods and reporting units, arising from different techniques and methodologies applied (Fauziah *et al.*, 2018). Analytical methods for monitoring microplastics in environmental samples consist of sampling, extraction (or separation), as well as, identification and quantification (or classification). In general, the selection of sampling method largely depends on the matrices to be sampled and the size limitation of microplastics targeted.

There are three main sampling strategies documented specifically to collect microplastic samples, namely selective sampling, bulk sampling and volume-reduced sampling (Hidalgo-Ruz *et al.*, 2012). Selective sampling is where the microplastics are extracted directly from the aquatic environment, applicable in cases where the microplastic items are large enough for identification with the naked eye. This brings to the main drawback of this method of high size limitation of detectable microplastics, as they are easily overlooked when mixed with other debris (Craig, 2018). Bulk sampling on the other hand involves the collection of samples, of predetermined volume or weight, which may negatively affect the representativeness of the sample (Tsang *et al.*, 2017).

As for the volume-reduced approach, it is advantageous for covering large quantities or areas of samples, as the method comprises of reducing the entire volume of bulk sample by fast filtration, such as from the use of nets (Güven *et al.*, 2017). Mesh size is a critical point because it determines the minimum size of microplastics to be detected. The most common mesh size employed is 300 μ m, including lower mesh sizes of 150 and 80 μ m (Dris *et al.*, 2016). The use of lower mesh size improves microplastics detection, but, the smaller the net mesh size, the greater is the likelihood of clogging due to suspended organic matter (Pico *et al.*, 2019). Of the three methods, selective method is usually applied in beach sampling, bulk method is mainly used to collect sediment samples and occasionally water samples, while volume-reduced method seems to be the most popular approach for water samples (Wang & Wang, 2018).

As for biota samples, microplastics have been detected in several organisms under natural and laboratory conditions (Rezania *et al.*, 2018). Organisms can be collected in grasps, traps, creels or bottom crawling (benthic invertebrates), by manta or bongo nets (planktonic and nektonic invertebrates), by trawls in different water levels (fish), by hand (e.g. bivalves or crustaceans) or by electrofishing.

The collected organisms are mostly pre-treated with chemical or enzymatic digestion to destroy the organic matter. It is noteworthy that there is a risk of damaging the microplastics due to mechanical friction or degradation, as well as, loss due to heating of the samples (Table 2.10) (Lusher *et al.*, 2017).

Method	Advantages	Disadvantages
Acidic digestion	HNO ₃ : Most organics	HNO ₃ : Dissolution of PS and
(HNO ₃ , HCl)	destroyed	PE possible; HCl: incomplete destruction of organics
Alkaline digestion (NaOH)	Most organics destroyed	Some polymers degraded (e.g. PC, CA, PET; PVC)
Alkaline digestion	Most organics destroyed;	-
(KOH)	most polymers resistant	
Oxidizing digestion (H ₂ O ₂)	Most organics destroyed	Polymers might be affected
Enzymatic degradation	Most organics destroyed,	Time-consuming, partly
(cellulose, lipase,	not hazardous	expensive, different enzymes
chitinase, protease,		for different sample
proteinase-K)		

Table 2.10: Comparison of organic digestion methods of biota samples (Lusher *et al.*, 2017).

As for the microplastics extraction procedure, density separation is the most common method utilized to separate microplastics from sediment or other inorganic matter which was not destroyed during the enzymatic or chemical digestion. Owing to the lower density of the microplastics (0.8–1.4 g/cm³), they float on the surface and are further retrieved with a separating funnel. Several of the high-density solutions utilized in this method are described in (Table 2.11)

Density solution	Chemical formula	Density (g/cm ³)
Sodium chloride	NaCl	1.0–1.2
Sodium tungstate dihydrate	$Na_2WO_4 \cdot 2 H_2O$	1.4
Sodium polytungstate	$3 \text{ Na}_2 \text{WO}_4 \cdot 9 \text{ WO}_3 \cdot 2 \text{ H}_2 \text{O}$	1.4
Potassium formate	K(HCOO)	1.6
Zinc chloride	ZnCl ₂	1.6-1.8
Sodium iodide	NaI	1.8

Table 2.11: Overview of the solutions used in density separation (Frias et al., 2014).

Apart from that, sieving is another frequently used method to isolate microplastics from water and sediment matrices. The mesh size of sieves mainly depends on the desired size range of microplastics to be extracted, with the majority ranging from 0.035 to 4.75 mm (Crawford & Quinn, 2017). Multi-tier sieving has been successfully employed in numerous studies to separate microplastics into several size categories, by using a series of sieves with a decreasing mesh size. New analytical methods in separating microplastics from sediments began to emerge like the electrostatic separation method, which takes advantage of differences in electrostatic behaviors between microplastics and sediments, where samples are charged at a high voltage (Felsing *et al.*, 2018).

With regards to identification and quantification techniques, a variety of methods have been employed to characterize microplastics such as through visual examination (by naked eye or using a microscope), Raman Spectroscopy (Di & Wang, 2018; Zhang *et al.*, 2016), Fourier Transform Infrared Spectroscopy (FTIR) (Fuller & Gautam, 2016; Lourenco *et al.*, 2017) and Thermal Desorption Pyrolysis Gas Chromatography/Mass Spectrometry (TD-Pyr -GC/MS) (Dekiff *et al.*, 2014). Recent literatures documented that visual examination remains as the most used technique, up to 79% of the studies, whereas techniques such as Raman and FTIR were used only by 28% and 14% studies, respectively, and for GC-MS, regardless of using pyrolysis or thermal extraction, is used by only 7% studies (Renner *et al.*, 2017).

Additionally, detecting microplastic pollution levels in real time such as through remote sensing is becoming increasingly important, in which to cater this, a new generation of sensors is being developed to measure microplastics faster and at various depths (Garaba & Dierssen, 2018; Erik, 2019).

2.3.4.5 Global Initiatives in Tackling Microplastics Pollution

The increased awareness on the grave threats of microplastics gives rise to the commitment of numerous stakeholders, including main industrial plastic producers, in developing strategies to conserve the oceans by reducing the prevalence of plastics and subsequently, microplastics in the environment. Legally, there are numerous instruments that have been proposed and/or put into effect at the national, regional and international levels aiming at curtailing this issue. These include action plans, strategies, regulations, conventions, guidelines and agreements that contain specific management measures, which may either be voluntary or non-voluntary, and can be classified as either preventive, removal, mitigative, and educational awareness (da Costa, 2018).

Table 2.12 summarizes the international conventions that promote the management of marine debris. Furthermore, marine debris has also been recognized in the decisions of the 11th Conference of the Parties to the Convention on Biological Diversity (CBD COP 11 Decision XI/18) and the 10th Conference of the Parties to the Convention on the Conservation of Migratory Species of Wild Animals (Gall & Thompson, 2015).

Year	Initiatives	Description
1972	London Convention	Prohibits the discharge or intentional dumping at sea of plastic waste in all maritime zones globally.
1982	United Nations Convention on the Law of the Sea (UNCLOS)	Protects and preserves the marine environment from both sea- and land-based sources of pollution.
1992	Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and Their Disposal	Basel Convention was amended on 29 April 2019 to include plastic waste in a legally-binding framework which will make global trade in plastic waste more transparent and better regulated, whilst also ensuring that its management is safer for human health and the environment.
2011	Honolulu Strategy	Planning framework intended to prevent and manage marine debris. Goals on reduction in generation and impacts of marine debris from land-based, sea-based sources, and also reduction in accumulation of marine debris
2012	Global Partnership on Marine Litter (GPML)	Developed from Honolulu strategy, a multi-stakeholder coordination in reducing and managing marine debris
2013	MARPOL Annex V	Addresses ocean-based litter pollution and prohibits the discharge of all plastics from ship.
2015	G7 Summit	Ocean Plastic charter on making all plastics recyclable, reducing single-use plastics and promoting use of recyclable plastics.
2016	United Nations Environment Assembly's resolution on Marine Debris	Declared marine debris as serious global issue and motioned countries to put marine plastic pollution high on environmental policy agenda.
UNEP-MAP (1995), OSPAR (2010), HELCOM (2015)	United Nations Environment Program/Mediterranean Action Plan (UNEP–MAP) and Oslo/Paris Convention	Formulated guidelines for evaluation of marine debris including microplastics.

Table 2.12: International conventions that promote the management of marine debris.

Apart from that, sustainable development goals (SDGs) are also applicable in tackling marine debris issue that directly or indirectly prevent, mitigate and encourage appropriate management of marine debris through the four SDGs namely Clean Water and Sanitation (Goal 6), Sustainable Communities and Cities (Goal 11), Responsible Consumption and Production (Goal 12) and Life Below Water (Goal 14).

In addition to the aforementioned initiatives, there are also directives outlined by the European Union that are of importance with respect to marine plastic debris which are the Marine Strategy Framework Directive (MSFD) (2008/56/EC), the Water Framework Directive (WaFD) (2000/60/EC) and the Port Reception Facilities Directive (PRFD) (2000/59/EC) (Steensgaard *et al.*, 2017). The MSFD focusses on restoring the marine environment by preventing the increase of marine debris. From this directive, Denmark has established "the Danish Marine Strategy" in 2012 (Lassen *et al.*, 2015), which includes monitoring programmes that investigate marine debris on beaches and the sea floor, including the analysis of microplastics in sediments, as well as, ingestion by marine biota (Strands *et al.*, 2014). Additionally, the WaFD aims to achieve "good water status" through both "good ecological status" and "good chemical status", that applies to surface waters such as lakes, rivers, estuaries and coastal waters up to one nautical mile from land (Klauer *et al.*, 2017). Since shipping is recognized to contribute 6.5 million tonnes of marine plastic debris, PRFD is relevant as it aims to reduce pollution of both seas and coastlines caused by shipping and cargo residues (Kaika & Page, 2003).

Furthermore, there are also monitoring research on marine debris by the nongovernmental organizations (NGOs) that engage in awareness campaigns (Pettipas *et al.*, 2016) such as the 5 Gyres Institute and the Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection. Also, the International Coastal Cleanup (ICC), overseen by the Ocean Conservancy encourages other NGOs and volunteer groups in cleaning up coastal areas worldwide. Meanwhile, the Honolulu Strategy outlines strategies for the prevention and management of marine debris which has been adapted across the globe (Prata, 2018). Two of the strategies that are of particular interests are the implementation of market-based instruments (e.g., levies on new plastic bags) and the formulation of policies, regulations and legislations like bans on microbeads and plastic bag.

Levying of taxes is one of the commonly adapted intervention strategies to reduce single-use plastic bag albeit with varied success (Dikgang *et al.*, 2012). Some other types of interventions that have been implemented in different countries include an outright ban on plastic bags in Bangladesh and China (Zhu, 2011), a plastic 'producer tax' in Italy, and a 'weight-based tax' (i.e. a tax based on the weight of one's wastage) in Denmark (Convery *et al.*, 2007). Compared to plastic bags, there have been limited interventions to reduce microbeads, but there has been a rapid proliferation in policies to reduce the use of microbeads globally (Figure 2.15) (Xanthos & Walker, 2017).



Figure 2.15: The number and trend of global plastic bags and microbeads interventions globally (Xanthos & Walker, 2017).

Many initiatives and campaigns have been undertaken by the Government of Malaysia in tackling plastic pollution (Table 2.13). In 2018, the Government has charted a zerowaste plan that aims to abolish single-use plastics by 2030, making Malaysia the first country in Southeast Asia to take bold action to tackle this issue (Zafirah, 2018). The Malaysia's Roadmap toward Zero Single-Use Plastics 2018-2030 includes among which a nationwide charge on plastic bags and to only serve plastic straws upon customer request, while suggesting manufacturers on how they can go to alternatives such as reusable straws (MESTECC, 2018).

Table 2.13: Initiatives and campaigns undertaken by the Government to beat plastic pollution.

Year	Initiative	Description	Reference
2009	Banning of single- use plastic.	The use of single-use plastic bag is banned on every Saturday.	Sang <i>et al.</i> (2019)
2017	Banning of single- use plastic.	The ban is applied on all days. Consumers are required to use their own recyclable shopping bags.	Sang <i>et al.</i> (2019)
2017	Restrict the use of conventional plastics products.	The use of conventional plastics products that are based on hydrocarbons has been restricted since 2016 and is enforced on 1 September 2017 by the Ministry of Federal Territories.	Sang <i>et al.</i> (2019)
2018	Malaysia's Roadmap toward Zero Single- Use Plastics 2018- 2030.	Introduced by the Ministry of Energy, Science, Technology, Environment and Climate Change (MESTECC) to take a phased, evidence-based and holistic approach by involving all stakeholders in jointly addressing single-use plastics pollution in Malaysia.	MESTECC (2018)
2019	Banning of plastic straws	The use of plastic straws is banned in the Federal Territories (Kuala Lumpur, Putrajaya & Labuan) starting on 1st January 2019 before the ban is fully enforced on licensed traders and food operators in 2020.	Nair (2018)
2019	WWF-Malaysia's goal to stop plastic leakage into nature by 2030.	Advocate producers and businesses to design products and packaging materials with recovery and circularity in mind.	WWF (2019)

Globally, addressing this issue requires a collective action along the entire life cycle of plastic - production, consumption and disposal. It requires a joint effort such as policies and stronger enforcement from the Government, innovative and sustainable solutions from industry players, as well as, behavioral change among the consumers. If addressed, these may actively help in reducing the pervasiveness of microplastics.

2.4 Persistent Organic Pollutants (POPs)

The rapid economic expansion and the ever-growing population have led to adverse environmental impacts, and pollution arising from POPs is one of them (Han *et al.*, 2016). Subsequent sections elaborate the sources and occurrences of POPs, their associated impacts, the analytical methods in the determination process, followed by the international environmental agreements pertaining to POPs.

2.4.1 Sources and Occurrences of POPs in Freshwater Environment

POPs are priority pollutants, comprised of predominantly man-made chemicals consisting of pesticides, industrial chemicals (PCBs, PBDEs, PFOS etc.) and by-products of industrial processes (dioxins and furans). Additionally, natural processes are also responsible for adding these pollutants into our ecosystem, such as from volcanic activities and vegetation fires (El-Shahawi *et al.*, 2010).

There are different types of POPs recognized by the United Nations Environment Programme (UNEP), Inter-governmental Negotiating Committee, Montreal, Canada, International POPs Elimination Network (IPEN) and Stockholm Convention. Among these, POPs that are of global concern are the ones being targeted by the Stockholm Convention. They are persistent, mutagenic, carcinogenic, toxic, and/or having endocrine disrupting properties (Cruz-Martinez *et al.*, 2015; Smalling *et al.*, 2015). The pollutants are classified into four categories (i.e. (A) subject to elimination of production and use, (B) restricted in production and use, (C) unintentionally produced and (D) chemicals under investigation) (Table 2.14) (Alharbi *et al.*, 2018).

Table 2.14: POPs recognized in Stockholm Convention (Alharbi et al., 2018).

Class	POPs	
Α	Aldrin, hexachlorobenzene, mirex, endrin, chlordecone, chlordane,	
	dieldrin, heptachlor, toxaphene, lindane, hexa- and penta-	
	bromodiphenyl ethers (commercial octabromodiphenyl ether), tetra-	
	and penta-bromodiphenyl ethers (commercial pentabromodiphenyl	
	ether), PCBs, α - and β -hexachlorocyclohexane, α - and β -	
	endosulfans (technical endosulfan and its isomers),	
	pentachlorobenzene, hexabromobiphenyl.	
В	DDT, PFOS and PFOSF.	
С	Pentachlorobenzene, hexachlorobenzene, PCDDs, polychlorinated	
	dibenzofurans PCDFs, PCBs.	
D	Chlorinated naphthalenes, HBCD, short-chained chlorinated	
	paraffins, hexachlorobutadiene, pentachlorophenol.	

Phthalate acid esters (PAEs), most commonly used as plasticizers in the polymer industry have been classified as another major group of POPs, by the U.S. Environmental Protection Agency, the European Union and the China National Environmental Monitoring Center due to its mutagenicity, teratogenicity, along with carcinogenicity of these compounds (Agamuthu & Kee, 2016; Zhang *et al.*, 2018). The worldwide production of plasticizers is in billions of dollars with the majority of plasticizers being used across Asia, mainly in China, and about 85% of these are PAEs (Godwin, 2017; Luo *et al.*, 2018).

In the aquatic environment, sediment is considered to be the ultimate sink while water column acts as the main carrier of POPs, both serving as reservoirs for the cycling of these contaminants (Liu *et al.*, 2017). Table 2.15 highlights some studies that have been carried out globally to monitor the extent of POPs pollution.
Type of POPs	Location	Mean value (ng/L)	Reference
PAHs	Yellow River, China	18,663	Li et al. (2016)
	Liaohe River Basin, China	4,021	Guo et al. (2007)
	Minjiang River Estuary, China	72,400	Zhang <i>et al.</i> (2004)
PAEs	Jiulong River Estuary, China	2,625	Li et al. (2017)
	Pearl River, China	5,340	Liu et al. (2014)
	Moscow River, Russia	85	Eremina et al.
			(2016)
	Epe and Lagos Lagoons,	180	Adeogun et al.
	Nigeria		(2015)
DDT	El-Rahawy, Egypt	229	El Bouraie et al.
			(2011)
	Chenab River, Pakistan	81	Eqani et al. (2012)
PCBs	Minjiang River Estuary, China	1,338	Zhang et al. (2004)
	Haihe River, China	1,565	Han & Currell
			(2017)

Table 2.15: Mean values of POPs in freshwater environment reported worldwide.

2.4.2 Fate and Impacts of POPs

POPs typically exist in the environment for more than 20 years to as long as a century, due to their persistence against degradation (Alharbi *et al.*, 2018; Markowitz & Rosner, 2018). POPs with higher water solubility are mainly present in the aqueous phase, while POPs that are of medium and low solubility are able to interact with suspended particles and sediments or accumulate in the biological tissues of aquatic biota, and subsequently biomagnified (Pérez-Parada *et al.*, 2018).

Despite bans or restrictions on production and use of POPs, they are widely distributed within environmental compartments and continue to be reported at toxic concentrations in organisms of various trophic levels (Johnson *et al.*, 2013). The accumulation of POPs might lead to toxic effects in affected organisms via the alteration of the biochemical, physiological, histological and morphological parameters (Da Cuña *et al.*, 2013, Hued *et al.*, 2013). On top of that, global warming may further enhance the impact of POPs as evidence showed that elevated temperatures may alter the biotransformation of

contaminants to a more bioactive metabolites and thus impair homeostasis (Noyes *et al.,* 2009).

Concerning human food safety, the alarm on the presence of POPs in aquatic biota has arisen due to their occurrence in muscle tissue of edible species, as ingestion is the main route of non-occupational exposure to POPs in humans (Vestergren *et al.*, 2012). Research showed that POPs specifically DDT concentration in human is rising due to the exposure from fish species and poultry meats (Shoeb *et al.*, 2016).

In addition to that, humans are also exposed to POPs through inhalation and dermal contact as these compounds are widely used in various industrial and consumer products, such as in cosmetics, plastics and paints (Han & Currell, 2017). These compounds have adverse health effects and have been associated with an increased risk of breast cancer, testicular cancer, liver cancer, and colorectal cancer (Table 2.16) (Alharbi *et al.*, 2018).

No.	POPs	Health Problems	Reference
1	PCBs, OCDD and	Cardiovascular problems, increased	Penell et al.
	one flame retardant	blood pressure, increase in total	(2014)
	brominated	cholesterol, HDL, LDL, total serum	
	compound (BDE47)	lipids, ventricular systolic and	
		diastolic dysfunction, kidney cancer	
		and anorexia-cachexia syndrome.	
2	PAHs	Breast cancer in human, endocrine	Cabaravdić
		disruption.	(2006)
3	Phthalates	Testicular cancer, endocrine	Virtanen et al.
		disruption, disorders of	(2005)
		neurodevelopment and cardiovascular	
		systems.	
4	Oxychlordane and	Endocrine disruptors and obesity,	Elobeid et al.
	DDT	weight gain, advance puberty, and	(2010)
		induce changes in gene expression	
		associated with steroid hormones.	
5	Dioxins and furans	Type 2 diabetes, abdominal obesity.	Zeliger (2013)
6	PCDDs and PBDEs	Dhyroid hormone signalling.	Zoller (2008)

Table 2.16: Diseases and health problems reported due to the pollution of POPs.

2.4.3 Analytical Methods in Determining POPs

It is fundamental in developing effective analytical methods in the determination of POPs to well study their abundances, fates, together with their potential sources (Xu *et al.*, 2013). The detection of free concentrations of POPs, such as in the surface water or sediments is an issue of great importance, as in many cases, bioaccumulation and toxicity of pollutants were not related to the total, but rather to the free concentration of each matrices (Stefaniuk & Oleszczuk, 2016; Bartolomé *et al.*, 2018)

Table 2.17 presents a brief summary of the available extraction methods of POPs, from solid and liquid samples (Xu *et al.*, 2013; Lorenzo *et al.*, 2018). As recognized in the USEPA 1613, among all methods, the conventional Soxhlet extraction and Liquid–liquid extraction has been the standard approaches in the analysis of POPs from solid and liquid samples, respectively (Luque de Castro & Priego-Capote, 2010).

Sample	Extraction	Solvent	References
Solid	Soxhlet extraction	Toluene for environmental samples and hexane/DCM for biota tissue.	Charlestra (2008)
	Microwave-assisted extraction	Same solvents as those for Soxhlet extraction	Kot-Wasik <i>et al.</i> (2007)
	Pressurized liquid extraction	Same solvents as those for Soxhlet extraction; solvent is filled with 60% volume of PLE cell	Degger <i>et al.</i> (2011)
	Supercritical fluid extraction	CO ₂ supercritical fluid	Mugnai <i>et al.,</i> (2011)
Liquid	Liquid–liquid extraction	DCM	Hubert <i>et al.</i> (2000)
	Solid-phase extraction	C ₁₈ disk; washed with 5 mL acetone; eluted with 15 mL DCM for OCPs; eluted with 20 mL ACN for PCBs	Helaleh <i>et al.</i> (2012)
	Solid-phase microextraction	-	Saadati <i>et al.</i> (2013)

Table 2.17: POPs extraction methods (Xu et al., 2013; Lorenzo et al., 2018).

In order to achieve accurate quantification, compounds must be fully resolved from each other through chromatographic separation, whereby in most cases, this may be possible by the use of selective detection (Megson *et al.*, 2016). While several chromatographic techniques are potentially useful, gas chromatography (GC) and highperformance liquid chromatography (HPLC) are undoubtedly the most widely used techniques for environmentally-relevant separations (Darnerud *et al.*, 2011; Wittsiepe *et al.*, 2014). Additionally, GC coupled with mass spectrometry (MS) has been the most commonly used detector for the separation and identification of organic pollutants including a wide range of pesticides (Kim *et al.*, 2019).

2.4.4 International Environmental Agreements Pertaining to POPs

POPs have been the focus of several multilateral environmental agreements and conventions, which have been enacted to control the release, production and their usage (Table 2.18). These initiatives generally share the common objective of protecting human health and the environment from hazardous chemicals and wastes, in which they assist countries to better manage chemicals at different stages of their life-cycle (Torre *et al.*, 2016).

The Basel Convention emerged as a result of the claims made by developing countries, especially African countries, due to waste being improperly disposed of in their territories, nevertheless the effectiveness is still unclear (Kellenberg & Levinson, 2014; Núñez-Rocha & Martínez-Zarzoso, 2018). The Stockholm Convention initially addressed 12 priority POPs and the list have been extended to 28, as of 2017 (Rigét *et al.*, 2019; Zhao *et al.*, 2019a).

Malaysia became a signatory to the Stockholm Convention on POPs on 16 May 2002 and is one of the 12 countries selected to implement a GEF/UNEP-funded project for the development of a National Implementation Plan (NIP) for POPs management (IPEP, 2005). Ratification efforts are tied to Malaysia's ratification of the Minamata Convention on mercury, designed to protect human health and the environment from anthropogenic emissions and releases of mercury and mercury compounds (TAIEF, 2016).

The United Nations Industrial Development Organization (UNIDO) is responsible for supporting industries in developing countries and countries with economies in transition, to implement the Stockholm Convention. UNIDO's strategic programmes focus on its mandate on inclusive and sustainable industrial development, as guided by the Sustainable Development Goal 9 (i.e. Infrastructure, Industry and Innovation) (UNIDO, 2019).

As a whole, despite adaptation and entry into force of these agreements, reports still confirm elevated POPs concentrations (Bruce-Vanderpuije *et al.*, 2019) and this may be partly due to the long-range transport of these pollutants, as well as, impacts from improper waste disposal, mostly in developing countries (Gioia *et al.*, 2012). On top of that, the key challenge identified by the conventions' secretariats is the inadequate implementation of national-level commitments, concerning adaptation and compliance mechanisms.

Year	International agreement	Objectives	Descriptions	Reference
1992	Basel Convention on the Control of Transboundary Movements of Hazardous Wastes and their Disposal	To control international shipments of hazardous waste and to develop appropriate management techniques.	Also provides for the establishment of regional or sub-regional centres for training and technology transfers to cater to the specific needs of different regions and subregions.	Lucier & Gareau (2016)
1998	UNECE Convention on Long-Range Trans-boundary Air Pollution (CLRTAP)	To limit and, as far as possible, gradually reduce and prevent air pollution including long-range transboundary air pollution.	Focuses on a list of 16 substances comprising eleven pesticides, two industrial chemicals and three unintentional by-products, which later included seven new substances.	Bull (2013)
2004	Stockholm Convention	To promote global action, with an overall objective to protect human health and the environment from POPs.	Each party is required to eliminate the production, export, import and use of POPs listed in Annex A, and to restrict the production and use of those listed in Annexes B and C.	Secretariat of the Stockholm Convention (2017)
2004	Rotterdam Convention	To promote shared responsibilities in relation to importation and use of hazardous chemicals.	Contain legally binding obligations in implementing Prior Informed Consent (PIC) procedure.	Barrios (2003)
2007	Union Implementation Plan (UIP)	To fulfil legal obligations, lay down strategy and action plan for further measures related to POPs included in the Stockholm Convention and/or in the UNECE Protocol on POPs.	Developed an Implementation Plan on POPs, which also covers the substances that fall under the UNECE Protocol on POPs.	Vijgen <i>et al</i> . (2019)

 Table 2.18: Multilateral environmental agreements enacted to regulate POPs.

CHAPTER 3: METHODOLOGY

3.1 Study sites

Six rivers were selected based on their location to represent Peninsular Malaysia. They comprised of three rivers from the West Coast (Sepetang River, Serkam River and Ayer Masin River) and three rivers from the East Coast (Sedili Besar River, Cherating River and Semerak River). Eight sampling sites were selected within each river based on river accessibility and status.

The distance between the sites depends on the overall distance of each river, upon which the total distance is divided by eight. However, the said distance relies on accessibility. Hence, the distance revolves around the addition or reduction of maximum 500m from the predetermined distance due to the presence of human intervention (i.e. concrete riverbank).

Meanwhile, river status refers to the observable condition of river during the time where the sites were established. For instance, when establishing two consecutive sites of predetermined distance and the said sites are within an area of open dumping, the determination of the second site is done as much as possible at the location without open dumping, to better investigate the extent of microplastics and POPs pollutions. However, the deviation in the distance was within 500m from the predetermined distance.

Three sampling events were conducted for each sampling site within November 2017 to August 2018. Table 3.1 delineates the coordinates of the sampling sites, while the location of the sampling sites is presented in Figure 3.1.

River	State	Site	Location	Sampling Coordinates	
				Latitude	Longitude
Sepetang	Perak	1	upstream	4°55'39.4"N	100°41'54.2"E
River		2	upstream	4°55'38.1"N	100°41'51.9"E
		3	upstream	4°55'34.8"N	100°41'43.6"E
		4	middle stream	4°55'19.4"N	100°41'00.7"E

Table 3.1: The coordinates of the sampling sites.

River	State	Site	Location	Sampling	Coordinates
				Latitude	Longitude
Sepetang	Perak	5	middle stream	4°54'45.6"N	100°40'15.8"E
River		6	downstream	4°54'45.3"N	100°40'09.9"E
		7	downstream	4°54'33.4"N	100°39'59.7"E
		8	downstream	4°54'25.5"N	100°39'47.6"E
Serkam	Malacca	1	upstream	2°08'24.6"N	102°22'49.8"E
River		2	upstream	2°08'22.2"N	102°22'55.9"E
		3	middle stream	2°08'19.5"N	102°22'59.2"E
		4	middle stream	2°08'10.3"N	102°22'58.2"E
		5	downstream	2°08'08.5"N	102°22'58.4"E
		6	downstream	2°08'06.2"N	102°22'59.0"E
		7	downstream	2°08'04.3"N	102°22'58.7"E
		8	downstream	2°08'03.1"N	102°22'58.5"E
Aver Masin	Johor	1	upstream	1°20'44.8"N	103°27'20.8"E
River		2	upstream	1°20'37.5"N	103°27'17.6"E
		3	upstream	1°20'36.5"N	103°27'16.8"E
		4	upstream	1°20'35.1"N	103°27'13.6"E
		5	middle stream	1°20'34.0"N	103°27'11.6"E
		6	middle stream	1°20'32.9"N	103°27'05.1"E
		7	downstream	1°20'31.5"N	103°26'59.4"E
		8	downstream	1°20'32.0"N	103°26'54.7"E
Sedili Besar	Johor	1	upstream	1°55'34.9"N	104°05'34.9"E
River		2	upstream	1°55'36.3"N	104°05'36.1"E
		3	middle stream	1°55'39.0"N	104°05'38.7"E
		4	middle stream	1°55'41.9"N	104°05'38.8"E
		5	downstream	1°55'44.5"N	104°06'20.4"E
		6	downstream	1°55'44.7"N	104°06'20.4"E
		7	downstream	1°55'44.9"N	104°06'20.5"E
		8	downstream	1°55'45.1"N	104°06'20.6"E
Cherating	Pahang	1	upstream	4°07'30.3"N	103°21'39.7"E
River		2	upstream	4°07'30.9"N	103°21'41.7"E
		3	upstream	4°07'51.0"N	103°23'37.9"E
		4	middle stream	4°07'49.7"N	103°23'37.3"E
		5	middle stream	4°07'44.4"N	103°23'34.3"E
		6	downstream	4°07'40.7"N	103°23'32.1"E
		7	downstream	4°07'38.8"N	103°23'30.8"E
		8	downstream	4°07'37.6"N	103°23'27.6"E
Semerak	Kelantan	1	upstream	5°49'53.6"N	102°28'22.4"E
River		2	upstream	5°49'54.4"N	102°28'22.4"E
		3	upstream	5°49'56.1"N	102°28'22.7"E
		4	middle stream	5°49'56.3"N	102°28'23.0"E
		5	middle stream	5°49'57.3"N	102°28'23.2"E
		6	downstream	5°51'30.5"N	102°30'23.1"E
		7	downstream	5°51'30.5"N	102°30'24.4"E
		8	downstream	5°52'04.6"N	102°29'32.8"E

Table 3.1, continued.



Figure 3.1: The location of sampling sites, with numbers correspond to location in Table 3.1.

3.2 Sampling Method

3.2.1 River sediment

Two replications of the top 5 cm of the sediment in a 0.04 m² range area between the shoreline and riverbank was collected using a stainless-steel shovel, and kept in sealed plastic bags (Jiang *et al.*, 2019). Triplicates of 200g of river sediments were also collected randomly for the analysis of POPs. All sediment samples were stored at 4°C prior to analysis.

3.2.2 River water

Microplastics in water were collected using conical nylon plankton net (100 μ m; 0.3m in diameter; 1m long) (modified from Tsang *et al.*, 2017) at two sampling locations (i.e. one at the upstream, one at the downstream). The survey was done by passing flows of water through the net for one hour (Plate 3.1). The microplastics retained in the net were washed into a container for further laboratory analysis (modified from Zhao *et al.*, 2014).



Plate 3.1: Microplastics sampling of river water using the net (as circled) at one of the sampling sites in Semerak River, Kelantan.

The volume of sample collected was calculated by taking the product of river surface velocity, cross sectional area of the submerged portion of the net opening, and sample collection time (Eqn. 3.1) (Estahbanati & Fahrenfeld, 2016). For the analysis of POPs, triplicates of 200 ml of water were collected randomly per sampling location and stored at 4°C prior to analysis.

 $Volume \ sampled = Water \ valocity \ \times \ Net \ cross \ sectional \ area \ \times \ Time$

(Eqn. 3.1)

3.3 Sample Extraction and Laboratory Analyses

3.3.1 River sediment

3.3.1.1 Soil Particle Analysis

Sediment samples were dry sieved using Tyler Sieves of 1.0 mm mesh sizes. The sediment samples were then tested using Beckman Coulter (LS 13, 320) to determine its soil type (i.e. silt, clay and sand). The results obtained were then calculated by using percentage, and the soil triangle from the U.S. Department of Agriculture (USDA) was then utilized to identify the soil texture (USDA, 1984).

3.3.1.2 Microplastics

All sediment samples were dried at 50 °C for at least 48 h. 300 g dried samples were analyzed. Microplastics were extracted from each sample based on a density separation method modified by Thompson *et al.* (2004). Each 300 g sediments were mixed with 750 mL of concentrated NaCl solution in a glass beaker for 2 min by stirring with a glass rod. The mixture was left standing overnight and the resulting supernatant were wet sieved through a set of Tyler Sieves with 5.0 mm, 1.0 mm and 0.1 mm mesh sizes. The microplastics that were retained on the sieves were separated using steel tweezers, and were then treated with 20% alcohol solution overnight.

3.3.1.3 POPs

USEPA method 3540 (Soxhlet extraction) modified by Gaylor *et al.* (2015) was used for extracting POPs from sediments. Sediment samples were air dried, grounded and passed through a 2 mm sieve. 20 g of each sample were then extracted with dichloromethane (DCM) for 3h via Soxhlet. Filtered solution was evaporated by rotary evaporator with water temperature set at 40°C to total dryness. Round bottom flask was rinsed with 3 ml of DCM and shaken well. Lastly, the solution was transferred into GC-MS/MS vial and stored in a freezer until analysis.

3.3.2 River water

3.3.2.1 Physicochemical Analysis

(A) Conductivity, temperature, total dissolved solids (TDS), pH and salinity

100 ml of river water sample was poured into a glass beaker. Conductivity, temperature, TDS, pH and salinity were identified using YSI 550A Multiparameter and the values were tabulated.

(B) Turbidity and total suspended solids (TSS)

Samples of 25 ml of river water and deionized water were placed into different spectrophotometer glass cuvettes. Deionized water was used as blank. Turbidity and TSS were determined with the use of spectrophotometer (HACH DR/4000).

(C) Biochemical oxygen demand (BOD)

Biochemical oxygen demand (BOD) is a commonly used parameter for water biodegradability. This analysis measured the oxygen required by microorganisms for the biochemical degradation of organic material (Simon *et al.*, 2011). Reagents for BOD test were prepared as follows: a) Phosphate buffer solution: 8.5 g of potassium dihydrogen phosphate (KH₂PO₄), 21.75 g of dispotassium hydrogen phosphate (K₂HPO₄), 33.4 g of disodium hydrogen phosphate heptahydrate (NA₂HPO₄.7H₂O) and 1.7 g ammonium chloride (NH₄CL) were dissolved in 500 ml of distilled water and diluted to 1L. The pH of this buffer was 7.2.

b) Magnesium sulphate solution: 22.5 g of magnesium sulphate (MgSO₄.7H₂O) was dissolved in distilled water and diluted to 1L.

c) Calcium chloride solution: 27.5 g of anhydrous calcium chloride (CaCl₂) was dissolved in distilled water and diluted to 1L.

d) Ferric chloride solution: 0.25 g of ferric chloride (FeCl₃.6H₂O) was dissolved in distilled water and diluted to 1L.

e) Acid and alkali solutions (1N): Acid solution was prepared by added 28 ml of concentrated sulfuric acid slowly into distilled water and diluted to 1L. 40 g of sodium hydroxide was added in distilled water and diluted to 1L to prepared alkali solution.

Procedure:

- 1. 350 ml of river water sample was prepared for each BOD bottle.
- 2. River water sample was diluted (50x) with prepared BOD dilution water in the beakers and pH was adjusted between 6.5-7.5 by adding acid or alkali solution.
- BOD bottles were filled up with diluted samples and DO₀ readings were taken using DO meter YSI Model 57.
- 4. The remaining portion of BOD bottles were topped up with prepared BOD dilution water to avoid trapping air bubbles in the bottles.
- 5. Stopper was placed tightly and BOD bottles were incubated at 20 °C for 5 days.
- 6. Bottles were taken out after 5 days and DO₅ were determined.

7. BOD₅ was calculated using the formula: $[(DO_0-DO_5) \times dilution factor]$ (Eqn. 3.2)

(D) Chemical oxygen demand (COD)

COD is defined as the amount of specified oxidant that reacts in the water under controlled conditions. River water sample was diluted (50x) with distilled water. Then, 2 ml of diluted river water was pipetted into COD HACH vial and tightly capped. Vial was shaken vigorously and the outer wall of the vial was wiped dry. Vial was then placed into TECATOR COD digestion unit and digested for two hours at 150 °C. Vial was cooled to room temperature and COD was measured using spectrophotometer (HACH COD HR Program).

3.3.2.2 Microplastics

Twenty liters of water samples containing microplastics from the washing of nets were poured to pass through a set of Tyler sieve of 5.0 mm, 1.0 mm and 0.1 mm mesh sizes. The microplastics that were retained on the sieves were separated using steel tweezers, and were then treated with 20% alcohol solution overnight.

3.3.2.3 POPs

Liquid-liquid Extraction (LLE) process was used to extract POPs from water samples. Method of extraction was modified from Marinho *et al.* (2010), Botalova *et al.* (2011), and Li *et al.* (2013). 200 ml sample was poured into measuring cylinder and transferred into separating funnel. 50 ml of Dichloromethane (DCM) was then added into the separating funnel and shaken well for five minutes. The solution was left for three minutes until two layers were formed. Lower layer was dried by filtration using 20 g of anhydrous granulated Sodium Sulphate (Na₂SO₄) into round bottom flask. The steps were repeated twice. Filtered solution was evaporated by rotary evaporator with water bath set at 40°C to total dryness. Round bottom flask was rinsed with 3 ml of DCM and shaken well. Lastly, the solution was transferred into GC-MS/MS vial for analysis.

3.4 Identification, Classification and Quantification

3.4.1 Anthropogenic activity

Anthropogenic activities that may contribute to the generation of microplastics and POPs were identified through observation of 5 km radius area along each river. The classification of anthropogenic activities was conducted according to the categories established by the Department of Survey and Mapping Malaysia (JUPEM) from the Geospatial Image Online Services (GIOS) (JUPEM, 2019). As for the assessment, each river was categorized based on total number of anthropogenic activities (Table 3.2).

Table 3.2: River category with respect to the assessment of anthropogenic activities.

No.	Category	No. of anthropogenic activity (N)
1	Low	$N \leq 5$
2	Moderate	5 < N < 10
3	Hotspot	$N \ge 10$

3.4.2 Microplastics

Identification of microplastics was conducted based on the morphological characteristics (i.e. type, size and color) (Table 3.3), using a binocular dissection microscope equipped with digital eye-piece camera (Dino-Eye, AM4023X, 1.3 megapixels).

Table 3.3: The morphological characteristics of microplastics.

Category	Classification	Reference
Туре	Line (fibrous), Fragment (hard, jagged), Film	Sutton <i>et al.</i> (2016)
	(thin, flimsy), Foam (lightweight, sponge-like),	
	Pellet (hard, rounded).	
Size	<0.1 mm, 0.1 – 0.5 mm, 0.5 – 1.0 mm, 1.0 –	Wang <i>et al.</i> (2018)
	5.0 mm	
Colour	Transparent, Black, Blue, Red, Yellow, White,	Su et al. (2016)
	Others	

3.4.3 POPs

The determination of POPs in DCM extracts was performed using the Agilent Technologies-7890 Gas Chromatograph coupled with Tandem Mass Spectrometry Agilent 7.000 (GC-MS/MS) (Agilent Technologies, USA). The following conditions were set for all extracted samples (Table 3.4). The concentration of POPs was calculated based on peak area percent and retention time.

Programme	Condition
Column	A HP-5MS fused silica capillary column (30 m x 0.25 mm, Agilent Technologies, Malaysia)
Carrier gas	Nitrogen
Oven temperature	60 °C for two minutes, followed by linear increase of 10 °C/minute to 120 °C, and from 120 to 300 °C at a rate of 3 °C/minute and held at 300 °C for 10 minutes.
Injector temperature	280 °C
Detector temperature	300 °C
Column flow rate	1.5 ml/minute
Split flow ratio	Splitless

Table 3.4: Programme for GC-MS/MS analysis.

3.5 Statistical Analyses

All data collected were tabulated and statistically analyzed using the Microsoft Excel 2016 and IBM SPSS Statistics version 20.0 software. Two units of microplastics abundance were applied, namely number of part/mass (particles/kg) for river sediment, and part/volume (particles/m³) for river water. As for POPs, the unit parts per million (ppm) was used to report concentration (in sediment and water). Descriptive analysis was performed on the abundance of microplastic particles and POPs concentration in the sediment and water, i.e. maximum value, minimum value, mean value, and standard deviation. The differences between the abundance of microplastics and POPs concentration of the six rivers were performed with the one-way analysis of variance (ANOVA, p < 0.05), while the correlation between microplastics and POPs concentration of sediment and water was tested with the Pearson correlation analysis.

CHAPTER 4: RESULTS AND DISCUSSION

4.1 Description of Selected Rivers

4.1.1 Sepetang River, Perak

Sepetang River emerges from Kerian District and traverses the northeast region of Perak into the Straits of Malacca. The river is approximately 33 km long with an area of 248 km². The Sepetang River defines part of the border between Kerian and Larut Matang District in Perak, which runs parallel to the river. The estimated total amount of rainfall was between 2,400 and 2,600 mm per annum, with average yearly temperature of 25°C to 26°C (Malaysian Meteorology Department, 2017). The river experiences minimum rainfall during the Southwest Monsoon season (i.e. from end of May to September).

4.1.2 Serkam River

Serkam River is located within Jasin District of Malacca, which occupied an area of 2,606 ha. The river is 17 km long with the estimate terrain elevation above sea level of 4 m. The temperature ranging from 25°C to 26°C, with rainfall ranging between 2,200 and 2,400 mm per annum (Malaysian Meteorology Department, 2017).

4.1.3 Ayer Masin River

Ayer Masin River, with approximate length of 16.6 km, flows towards the Straits of Malacca. The river is situated within Ayer Masin Mukim of 3,725 ha in Pontian District. The average yearly temperature is 25.5 °C, with average rainfall of 2,300 mm per annum (Malaysian Meteorology Department, 2017).

4.1.4 Sedili Besar River

Sedili Besar River is located on the north eastern side of Johor and empties into the South China Sea. The river is 66 km in length with the river mouth situated at the northern end of Teluk Mahkota bay. The river has a total drainage basin area of 271 km², within

the Kota Tinggi District. The annual rainfall is between 1,800 and 2,400 mm per annum, with annual temperature ranging from 25°C to 26°C (Malaysian Meteorology Department, 2017).

4.1.5 Cherating River

Cherating River is situated in Pahang, located about 47 km north of Kuantan. The river is 16.1 km long and approximately 43 m wide, that empties into the South China Sea. The estimated total amount of rainfall is between 2,200 to 2,500 per annum, with heavy rainfall during the Northeast Monsoon season (i.e. November to March) (Malaysian Meteorology Department, 2017). The annual average temperature is between 25°C and 26°C.

4.1.6 Semerak River

Semerak River is located within the Pasir Puteh District, in an area of 116.5 km², in the south east of Kelantan. The river is approximately 27.7 km in length, and drains into the South China Sea. The estimated total amount of rainfall is between 3,200 and 3,500 mm per annum with heavy rainfall during the Northeast Monsoon season (November to March) (Malaysian Meteorology Department, 2017). This area is reported to have the highest amount of rainfall with the lowest average annual temperature (i.e. $23^{\circ}C - 24^{\circ}C$) among all of the study sites.

4.2 Background Study of Selected Rivers

4.2.1 Sepetang River, Perak

The sediment of Sepetang River is predominantly sand particles in 87.5% of the soil samples, with an average of $62.59\% \pm 21.80$ sand, followed by $25.77\% \pm 12.30$ silt, and $11.65\% \pm 9.81$ clay (Table 4.1). According to the USDA soil classification, the soil texture of the sediment is dominated by sandy loam, followed by loamy sand and silty clay loam (USDA, 1984).

Site	Clay (%)	Silt (%)	Sand (%)	Soil texture
1	10.60 ± 0.64	29.50 ± 2.34	59.90 ± 1.70	Sandy loam
2	34.60 ± 6.79	51.30 ± 1.56	14.10 ± 5.23	Silty clay loam
3	3.01 ± 2.13	10.89 ± 1.36	86.10 ± 3.49	Loamy sand
4	7.31 ± 0.56	14.69 ± 0.71	78.00 ± 1.27	Loamy sand
5	13.00 ± 1.34	30.30 ± 4.38	56.70 ± 3.04	Sandy loam
6	11.20 ± 1.80	24.90 ± 4.06	63.90 ± 2.26	Sandy loam
7	5.78 ± 2.51	23.42 ± 0.32	70.80 ± 2.82	Sandy loam
8	7.68 ± 0.68	21.12 ± 1.02	71.20 ± 1.83	Sandy loam
Average	11.65 ± 9.81	25.77 ± 12.30	62.59 ± 21.80	Sandy loam

Table 4.1: Soil texture of Sepetang River.

In terms of physicochemical characteristics of river water, high BOD₅ were recorded across all sampling sites, ranging from average value of 9.30 ± 13.37 to 31.00 ± 15.78 mg/L (Table 4.2). Meanwhile, salinity was similar in all sampling sites, with average value of 0.04 ± 0.01 ppt. The pH values were in the range of pH 4.46 ± 1.62 to pH 7.57 \pm 1.22, which showed that the river water of Sepetang River was acidic to slightly basic across all sampling sites.

Site	BOD ₅	COD	T (°C)	TDS	Sal (ppt)	рH
	(mg/L)	(mg/L)		(mg/L)	U I <i>)</i>	1
1	24.0 ±	$50.75 \pm$	27.8 ± 0.2	5702.21 ±	$0.04 \pm$	4.46 ±
	15.9	16.37		7.23	0.01	1.62
2	22.00 ±	34.0 ±	27.7 ± 0.1	$5600.55 \pm$	$0.04 \pm$	7.54 ±
	13.07	16.7		7.44	0.01	1.82
3	30.0 ±	31.25 ±	27.9 ± 0.2	$5604.55 \pm$	$0.04 \pm$	7.57 ±
	14.3	12.39		7.45	0.01	1.22
4	$16.0 \pm$	$48.50 \pm$	27.8 ± 0.3	5212 ±	$0.04 \pm$	$7.46 \pm$
	16.2	11.30		7.44	0.01	1.82
5	31.00 ±	36.0 ±	27.8 ± 0.2	$5503.9 \pm$	$0.04 \pm$	4.45 ±
	15.78	13.6		7.34	0.01	1.92
6	19.0 ±	61.25 ±	27.5 ± 0.1	$4101.6 \pm$	$0.03 \pm$	7.41 ±
	12.9	16.00		6.34	0.01	1.34
7	$23.40 \pm$	47.0 ±	27.2 ± 0.2	$4145.6 \pm$	$0.03 \pm$	4.47 ±
	14.67	14.0		7.54	0.01	1.25
8	9.30 ±	$68.0 \pm$	27.4 ± 0.1	4124.6 ±	$0.03 \pm$	4.53 ±
	13.37	12.5		3.84	0.01	1.89
Augroga	$21.84 \pm$	$47.09 \pm$	$27.64 \pm$	$4999.38 \pm$	$0.04 \pm$	5.98 ±
Average	17.37	16.9	0.24	738.87	0.01	1.62

 Table 4.2: Physicochemical characteristics of Sepetang river water.

4.2.2 Serkam River, Malacca

The soil composition primarily composed of silt particles with an average value of $46.72\% \pm 10.12$ silt, followed by sand and clay particles with $38.46\% \pm 13.48$, and $14.40\% \pm 2.40$, respectively (Table 4.3). There is a trend in the soil texture of sediment particles across sampling sites which comprised of sandy loam in the upstream, followed by silt loam in most of the sampling sites, and loam towards the downstream.

Site	Clay (%)	Silt (%)	Sand (%)	Soil texture
1	12.00 ± 1.27	31.50 ± 1.63	56.50 ± 2.90	Sandy loam
2	7.46 ± 0.91	31.34 ± 2.91	61.20 ± 3.82	Sandy loam
3	18.30 ± 1.90	54.40 ± 3.18	27.30 ± 5.09	Silt loam
4	17.70 ± 2.33	54.10 ± 4.81	28.20 ± 7.21	Silt loam
5	15.70 ± 2.69	52.70 ± 5.30	31.60 ± 7.99	Silt loam
6	16.50 ± 2.69	51.80 ± 4.67	31.70 ± 7.35	Silt loam
7	16.50 ± 2.19	54.60 ± 4.81	28.90 ± 7.00	Silt loam
8	14.40 ± 2.40	43.30 ± 4.03	42.30 ± 6.43	Loam
Average	14.82 ± 3.57	46.72 ± 10.12	38.46 ± 13.48	Silt loam

Table 4.3: Soil texture of Serkam River.

In regard to the physicochemical characteristics of river water, BOD₅ was within the same range across all sampling sites ranging between 2.66 ± 2.78 and 7.70 ± 1.26 mg/L (Table 4.4). COD readings recorded at Site 2 and Site 7 were among the highest as compared to the other sampling sites, with $1,062 \pm 27.6$ mg/L and $1,050 \pm 23.3$ mg/L, respectively. Serkam River was revealed to be slightly acidic with the lowest pH recorded at Site 4 of pH 4.48 ± 0.73 .

Site	BOD ₅	COD	T (°C)	TDS	Sal (ppt)	pН
	(mg/L)	(mg/L)		(mg/L)		
1	7.20 ±	74.0 ±	34.7 ± 2.5	$260.95 \pm$	0.19 ±	6.40 ±
	1.76	25.3		49.83	0.01	0.67
2	5.40 ±	$1062.0 \pm$	25.3 ± 3.6	11732.32	0.29 ±	6.15 ±
	1.32	27.6		± 41.23	0.01	0.97
3	3.15 ±	86.0 ±	22.6 ± 3.5	241.15 ±	$0.18 \pm$	6.18 ±
	1.78	29.2		41.87	0.01	0.26
4	5.04 ±	303.0 ±	25.1 ± 3.3	$7228.00 \pm$	$0.40 \pm$	$4.48 \pm$
	1.12	21.2		49.34	0.01	0.73
5	2.66 ±	163.0 ±	25.6 ± 3.7	$7728.00 \pm$	0.62 ±	5.59 ±
	2.78	20.2		42.90	0.01	0.23

 Table 4.4: Physicochemical characteristics of Serkam river water.

Site	BOD5	COD	T (°C)	TDS	Sal (ppt)	pH
	(mg/L)	(mg/L)		(mg/L)		
6	54 ± 3.86	$167.0 \pm$	25.1 ± 2.5	11693.50	$0.39 \pm$	6.18 ±
		21.8		± 42.13	0.01	0.67
7	$7.70 \pm$	$1050.0 \pm$	25.6 ± 1.5	12199.00	$0.99 \pm$	6.54 ±
	1.26	23.3		± 49.79	0.01	0.07
8	3.33 ±	67.0 ±	25.3 ± 3.6	21853.00	$0.18 \pm$	6.37 ±
	1.77	29.5		± 44.37	0.01	0.26
Average	5.00 ±	371.0 ±	26.16 ±	9116.99 ±	0.41 ±	5.99 ±
	1.86	429.0	3.58	7049.73	0.28	0.67

Table 4.4, continued.

4.2.3 Ayer Masin River, Johor

The sediment of Ayer Masin River comprised of mostly silt particles (55.30% \pm 8.84), followed by 30.29% \pm 10.55 sand, and 14.36% \pm 2.40 clay (Table 4.5). In terms of soil texture, the sediments at the majority of the sampling sites were silt loams, from Site 3 towards the downstream.

Site	Clay (%)	Silt (%)	Sand (%)	Soil texture
1	11.40 ± 0.07	41.40 ± 5.73	47.20 ± 5.66	Loam
2	14.80 ± 4.00	48.20 ± 2.64	37.00 ± 6.65	Loam
3	11.30 ± 1.82	54.60 ± 5.25	34.10 ± 7.07	Silt loam
4	12.80 ± 2.09	50.80 ± 5.91	36.40 ± 7.99	Silt loam
5	18.30 ± 4.67	60.70 ± 2.76	21.00 ± 7.42	Silt loam
6	15.90 ± 1.41	69.70 ± 1.34	14.40 ± 2.76	Silt loam
7	15.60 ± 0.57	54.90 ± 8.56	29.50 ± 7.99	Silt loam
8	14.80 ± 1.13	62.10 ± 5.94	22.70 ± 7.07	Silt loam
Average	14.36 ± 2.40	55.30 ± 8.84	30.29 ± 10.55	Silt loam

Table 4.5: Soil texture of Ayer Masin River.

BOD₅ values across all sampling sites were within the range of moderately polluted river, with average value between $1.80 \pm 1.22 \text{ mg/L}$ (Site 4) and $8.23 \pm 2.28 \text{ mg/L}$ (Site 6) (Table 4.6). Literatures have shown that BOD range of 2 to 8 mg/L reflects that the river water is moderately polluted (WWAP, 2015). Average COD readings were reported to be $36.1 \pm 10.6 \text{ mg/L}$, with average salinity of 0.62 ± 0.14 ppt. Ayer Masin River was revealed to be slightly acidic at the upstream and slightly basic towards the downstream.

Site	BOD5	COD	T (°C)	TDS	Sal (ppt)	pН
	(mg/L)	(mg/L)		(mg/L)		-
1	5.805 ±	30.0 ±	23.5 ± 1.7	$26097 \pm$	$0.59 \pm$	4.51 ±
	2.42	11.6		22.28	0.05	1.10
2	5.85 ±	$24.0 \pm$	26.6 ± 1.2	$24140 \pm$	0.73 ±	6.92 ±
	2.12	10.9		31.98	0.07	1.70
3	3.47 ±	$41.0 \pm$	24.8 ± 2.0	$26078 \pm$	$0.81 \pm$	$6.83 \pm$
	2.77	10.4		21.88	0.02	1.13
4	$1.80 \pm$	23.0 ±	27.2 ± 1.4	$25889.5 \pm$	$0.34 \pm$	4.51 ±
	1.22	11.6		23.26	0.05	1.56
5	4.46 ±	54.0 ±	23.1 ± 1.8	27131 ±	0.69 ±	$7.03 \pm$
	8.32	14.6		11.28	0.03	1.24
6	8.23 ±	33.0 ±	27.3 ± 1.1	$27046.5 \pm$	0.61 ±	7.15 ±
	2.28	16.0		15.78	0.08	1.99
7	3.33 ±	$44.0 \pm$	26.1 ± 1.0	27056.5 ±	$0.60 \pm$	$7.06 \pm$
	2.33	10.1		21.76	0.04	1.11
8	4.73 ±	40.0 ± 9.8	27.4 ± 2.7	$27046.5 \pm$	$0.61 \pm$	7.6 ± 1.20
	2.69			11.28	0.05	
Average	4.83 ±	36.1 ±	25.75 ±	26310.63	$0.62 \pm$	6.43 ±
	2.23	10.6	1.73	± 1021.28	0.14	1.21

Table 4.6: Physicochemical characteristics of Ayer Masin river water.

4.2.4 Sedili Besar River, Johor

The soil composition of Sedili Besar River is predominantly composed of sand $(56.01\% \pm 20.65)$ (Table 4.7). Clay particles contribute the least percentage with an average of $11.75\% \pm 6.83$. Overall, the soil texture of the river is dominated by sandy loam, from Site 3 towards the downstream.

Site	Clay (%)	Silt (%)	Sand (%)	Soil texture
1	28.30 ± 3.11	64.70 ± 4.60	7.00 ± 7.71	Silt loam
2	11.70 ± 0.99	36.20 ± 2.33	52.10 ± 3.32	Loam
3	10.00 ± 1.34	23.90 ± 1.91	66.10 ± 3.25	Sandy loam
4	10.40 ± 0.80	23.80 ± 1.53	65.80 ± 2.33	Sandy loam
5	8.28 ± 1.72	22.62 ± 1.96	69.10 ± 3.68	Sandy loam
6	7.18 ± 1.34	23.02 ± 2.20	59.80 ± 10.61	Sandy loam
7	8.95 ± 2.77	21.75 ± 0.76	69.30 ± 3.54	Sandy loam
8	9.17 ± 1.46	24.73 ± 1.44	58.90 ± 7.99	Sandy loam
Average	11.75 ± 6.83	30.09 ± 14.72	56.01 ± 20.65	Sandy loam

 Table 4.7: Soil texture of Sedili Besar River.

The BOD₅ for Sedili Besar River ranged between 1.22 ± 1.55 mg/L and 7.74 ± 1.90 mg/L, at an average of 4.30 ± 2.50 mg/L (Table 4.8). The values were comparable to the other rivers, except for Sepetang River which recorded substantially high BOD₅ values. The pH values were between pH 4.70 ± 1.11 and pH 7.29 ± 1.01 , which showed that the river was slightly acidic at an average of pH 6.50 ± 1.09 .

Site	BOD ₅	COD	T (°C)	TDS	Sal (ppt)	pH
	(mg/L)	(mg/L)		(mg/L)		
1	1.85 ±	60.0 ±	26.6 ± 1.2	7332.00 ±	0.37 ±	7.01 ±
	2.50	11.0		25.63	0.02	1.09
2	4.01 ±	93.0 ±	24.7 ± 1.3	6753.50 ±	0.31 ±	7.04 ±
	2.60	12.0		26.66	0.01	1.02
3	2.39 ±	83.0 ±	26.9 ± 2.2	$5180.00 \pm$	0.42 ±	7.29 ±
	2.53	10.0		24.99	0.02	1.01
4	1.22 ±	99.0 ±	27.1 ± 1.0	$6324.50 \pm$	0.44 ±	7.23 ±
	1.55	11.1		23.93	0.02	1.20
5	3.69 ±	30.0 ±	26.6 ± 1.7	$741.00 \pm$	$0.56 \pm$	4.81 ±
	2.30	14.0		22.63	0.01	1.10
6	7.74 ±	160.0 ±	28.3 ± 1.2	$1222.00 \pm$	$0.96 \pm$	7.15 ±
	1.90	12.5		24.00	0.01	1.09
7	7.11 ±	39.0 ±	24.6 ± 2.0	6305.00 ±	$0.48 \pm$	6.77 ±
	2.59	11.3		26.56	0.02	1.07
8	6.39 ±	$388.0 \pm$	26.6 ± 1.2	7335.00 ±	0.51 ±	4.70 ±
	2.34	11.0		25.12	0.20	1.11
Average	4.30 ±	119.0 ±	$26.43 \pm$	5149.13 ±	0.51 ±	6.50 ±
-	2.50	116.0	1.23	2664.30	2.44	1.09

 Table 4.8: Physicochemical characteristics of Sedili Besar river water.

4.2.5 Cherating River, Pahang

The soil composition of Cherating River primarily composed of sand particles with average of $86.56\% \pm 9.65$, followed by silt and clay particles at $11.14\% \pm 7.15$, and $2.65\% \pm 2.94$, respectively (Table 4.9). In terms of soil texture, the sediments at the majority of the sampling sites were loamy sand.

Across all sampling sites, Site 5 were documented to have the highest BOD₅, COD and TDS readings as compared to the others (Table 4.10). However, salinity was similar for all sites with average of 0.29 ± 0.25 ppt. As for pH, the upstream of the river (i.e. Site 1 to Site 4) was slightly acidic and the river water became slightly basic downstream (i.e. Site 5 to Site 8).

Site	Clay (%)	Silt (%)	Sand (%)	Soil texture
1	0	4.21 ± 1.00	95.79 ± 1.00	Sand
2	0	0	100	Sand
3	0	6.58 ± 0.89	93.42 ± 0.89	Sand
4	4.45 ± 0.42	19.25 ± 1.98	76.90 ± 2.40	Loamy sand
5	5.09 ± 0.80	16.81 ± 1.58	78.10 ± 0.78	Loamy sand
6	4.63 ± 1.58	12.60 ± 2.38	85.00 ± 3.96	Loamy sand
7	0	10.50 ± 1.06	89.50 ± 1.06	Loamy sand
8	7.06 ± 0.99	19.14 ± 1.62	73.80 ± 0.64	Sandy loam
Average	2.65 ± 2.94	11.14 ± 7.15	86.56 ± 9.65	Loamy sand

Table 4.9: Soil texture of Cherating River.

 Table 4.10: Physicochemical characteristics of Cherating river water.

Site	BOD ₅	COD	T (°C)	TDS	Sal (ppt)	pH
	(mg/L)	(mg/L)		(mg/L)		
1	5.8 ± 1.78	97.0 ±	25.2 ± 3.5	21.45 ±	0.01 ±	6.89 ±
		23.1		45.78	0.07	0.46
2	7.3 ± 2.01	275.0 ±	25.9 ± 3.4	19.50 ±	0.01 ±	5.82 ±
		32.1		45.55	0.03	0.23
3	8.6 ± 1.99	490.0 ±	28.1 ± 2.9	17030.00	$0.55\pm$	6.83 ±
		19.2		± 44.75	0.01	0.24
4	9.5 ± 2.18	722.0 ±	28.3 ± 3.1	18271.50	0.2 ±	6.99 ±
		20.1		± 43.45	0.08	0.47
5	10.75 ±	127.0 ±	29.4 ± 3.3	18343.00	0.28 ±	7.21 ±
	1.33	22.5		± 45.90	0.07	0.53
6	9.75 ±	37.0 ±	262 1 2 4	13227.50	$0.07 \pm$	7.21 ±
0	1.24	22.3	30.3 ± 2.4	± 45.22	0.05	0.12
7	7.75 ±	310.0 ±	20.0 ± 1.0	11732.50	0.61 ±	7.25 ±
	2.08	20.1	29.0 ± 1.9	± 42.00	0.02	0.09
o	5.7 ± 1.07	210.0 ±	20.0 ± 2.4	11674.00	0.55 ±	7.09 ±
0	3.7 ± 1.07	30.1	29.0 ± 2.4	± 41.75	0.02	0.14
A 11000 000	8.27 ±	283.0 ±	280 1 2 4	11289.93	0.29 ±	6.91 ±
Average	1.98	227.1	20.9 ± 3.4	\pm 7459.75	0.25	0.45

4.2.6 Semerak River, Kelantan

Sand particles dominated the soil composition of Semerak River at $65.30\% \pm 25.08$ (Table 4.11). Clay particles contribute the least with an average of $8.22\% \pm 7.61$. As for the soil texture, no clear trend was observed. However, sandy loam was identified in most of the sampling sites.

Site	Clay (%)	Silt (%)	Sand (%)	Soil texture
1	6.82 ± 0.16	30.01 ± 2.95	63.40 ± 3.11	Sandy loam
2	17.80 ± 2.97	65.00 ± 3.96	21.40 ± 6.93	Silt loam
3	20.00 ± 3.61	44.00 ± 5.59	41.10 ± 9.19	Loam
4	11.70 ± 1.48	30.99 ± 7.71	59.40 ± 9.19	Sandy loam
5	3.94 ± 1.56	17.27 ± 1.90	81.00 ± 3.46	Loamy sand
6	5.47 ± 0.42	26.03 ± 1.70	69.10 ± 2.12	Sandy loam
7	0	6.60 ± 3.29	93.40 ± 3.29	Sand
8	0	6.38 ± 0.54	93.62 ± 0.54	Sand
Average	8.22 ± 7.61	28.29 ± 19.57	65.30 ± 25.08	Sandy loam

 Table 4.11: Soil texture of Semerak River.

BOD₅ across all sampling sites fall within the range of moderately polluted river, with the highest (9.80 \pm 2.00 mg/L) recorded at Site 7 (Table 4.12). High COD values were evident across all sites, with average value of 208.4 \pm 125.5 mg/L, while the salinity ranged from 0.05 \pm 0.01 to 0.67 \pm 0.03 ppt. Semerak River was found to be slightly basic upstream, and slightly acidic downstream.

Site	BOD ₅	COD	T (°C)	TDS	Sal (ppt)	pH
	(mg/L)	(mg/L)		(mg/L)		
1	3.45 ±	$140.0 \pm$	30.9 ± 1.2	23315.5±	$0.48 \pm$	7.78 ±
	2.54	11.5		16.0	0.01	0.57
2	$2.70 \pm$	$258.0 \pm$	30.9 ± 1.1	23419.5 ±	$0.59 \pm$	7.32 ±
	3.04	12.4		18.2	0.04	0.65
3	8.75 ±	$66.0 \pm$	29.5 ± 1.0	21677.5 ±	$0.57 \pm$	7.38 ±
	2.56	12.6		16.0	0.02	0.16
4	$8.65 \pm$	$139.0 \pm$	29 ± 1.0	3074.5 ±	$0.51 \pm$	7.16 ±
	2.23	11.3		15.8	0.01	0.23
5	$7.50 \pm$	396.0 ±	29.5 ± 1.5	21677.5 ±	$0.67 \pm$	7.38 ±
	1.53	13.5		11.7	0.03	0.89
6	6.65 ±	$185.0 \pm$	29.6 ± 1.2	66.3 ±	$0.05 \pm$	5.66 ±
	2.04	12.6		11.0	0.01	0.44
7	9.80 ±	383.0 ±	28.1 ± 1.2	65.0 ±	$0.05 \pm$	6.85 ±
	2.00	12.2		14.9	0.02	0.12
8	$7.30 \pm$	$100.0 \pm$	27.5 ± 1.2	65.0 ±	$0.05 \pm$	6.85 ±
	1.26	12.0		15.0	0.01	0.56
Average	6.85 ±	$208.4 \pm$	$29.38 \pm$	$11670.1 \pm$	0.37 ±	6.92 ±
	2.53	125.5	1.20	11661.0	0.27	0.57

Table 4.12: Physicochemical characteristics of Semerak river water.

4.3 Anthropogenic Activities Surrounding Selected Rivers

It is of utmost importance to evaluate the anthropogenic activities that may have influenced the generation of microplastics and POPs into the selected rivers in determining their potential sources, concentrations as well as in assessing future mitigation plans. Table 4.13 highlights the anthropogenic activities that are present along selected rivers in Peninsular Malaysia.

Type of	River							
activities	Sepetang	Serkam	Ayer Masin	Sedili Besar	Cherating	Semerak		
Human	1	/	1	1		/		
settlement								
Tourism activities								
Resorts / Chalets	1			1	/	1		
Boat ride (eg:	1				1			
fireflies and								
eagle sighting)								
Mangrove river	1				/			
cruise								
River kayaking					1			
Camping	1				1			
Recreational	1		/	/	1			
centre								
Fisheries								
Fishing village		/	/	/	/	1		
Jetty	1	/	/	/				
Pre-fishing	1	/	/	/				
Leisure fishing	1	/	/	/	/	/		
Fishing pond					/			
Agriculture			/			/		
Aquaculture				/		1		
Eateries								
Restaurants	1	/	/		/			
Food stalls		/	/	/	/	1		
Total	15**	8*	10**	9*	11**	7*		

Table 4.13: The anthropogenic activities that are present along selected rivers.

**hotspots of anthropogenic activities (anthropogenic activities ≥ 10) *moderate anthropogenic activities (5 < anthropogenic activities < 10)

Sepetang River substantially demonstrated the highest number of anthropogenic activities among all selected rivers. Site 1 and Site 2 were surrounded by human settlement, and alongside this, open dumping of MSW was visible at several locations

along the river (Plate 4.1). Industrial activities such as charcoal, plastic, rubber, and aluminium factories are present close to Site 3 and Site 4 which may further exacerbate the pollution. Industries have larger tendency to generate greater extent of pollution as reports have shown that environments with mass industries are considered as key areas for contaminants, such as for microplastics and organic pollutants (Browne *et al.*, 2011). The downstream of the river (i.e. Site 5, Site 6, Site 7, and Site 8) were close to fishing villages (Plate 4.2) with busy fishing activities, along with heavy tourism activities.



Plate 4.1: Open dumping observed at the riverbank along the river.



Plate 4.2: Fishing villages along Sepetang River.

Cherating River (Plate 4.3) that lies within a resort town on the east coast of Peninsular Malaysia were recorded to have the second highest number of anthropogenic activities. Tourism and fishing activities are the main anthropogenic activities occurring in the area. The aforementioned high values of BOD₅, COD and TDS readings recorded at Site 5 are partly due to the presence of commercial fishing pond just adjacent to the river. The tourists or even locals may have discarded litter on the riverbank, which can easily enter the aquatic environment (Lytle, 2010), as well as, fishermen deliberately or accidentally released plastic wastes into the river environment (Hammer *et al.*, 2012).



Plate 4.3: The jetty for tourism-related activities in Cherating River.

Ayer Masin River, along with Serkam River and Sedili Besar River are classified to be profoundly affected by fishing activities, as well as, from human settlement. Human settlement was observed to be concentrated at Site 1, Site 2 and Site 3 of these rivers, with several fishermen's jetties observed from the middle region further downstream. Aquaculture farming of several types of fish was observed to be practiced downstream of Sedili Besar River (Plate 4.4). Additionally, houses were built adjacent to the aforementioned rivers with inefficient solid waste collection system. Due to that, open dumping of MSW were apparent along the riverbank (Plate 4.5 and Plate 4.6).



Plate 4.4: Aquaculture farms located downstream of Sedili Besar River, Kelantan.



Plate 4.5: Open dumping observed near the fishermen's jetty at Ayer Masin River.

The lowest number of anthropogenic activities was identified in Semerak River. Yet, it is noteworthy that this particular river is considered to share a moderate anthropogenic activity even though it holds the lowest number. The upstream (Site 1, Site 2, Site 3 and Site 4) was located within the area of human settlement and agricultural area of paddy cultivation.



Plate 4.6: Litters observed on the riverbank of Serkam River.

Several resorts were present towards the downstream (Site 5 and Site 6) to cater for tourists who may enjoy the scenic view of the river (Plate 4.7). Meanwhile, Site 7 and Site 8 were within a fishermen's village with the presence of aquaculture farms (Plate 4.8), specifically freshwater prawn farming.



Plate 4.7: The scenic view of Semerak River captured at one of the resorts.



Plate 4.8: Aquaculture farms located downstream of Semerak River, Kelantan.

4.4 Microplastics Abundance of Selected Rivers

4.4.1 Sepetang River, Perak

4.4.1.1 Total Abundance of Microplastics

Microplastics were ubiquitous in all river sediment samples along Sepetang River (Table 4.14). The abundance in river sediment ranged from 57.22 ± 36.75 to 149.44 ± 77.0 particles/kg, with an average of 101.39 ± 54.69 particles/kg. Statistically significant difference of microplastics abundance in river sediment was observed between the sampling sites [ANOVA, F(2,21)=16.99, p=0.000].

Greater microplastics abundance in sediments were observed at the locations of intense industrial activities (i.e. Site 3 and Site 4). This is not surprising since microplastic inputs are expected to be much higher in industrialized parts of a river. Similar finding was documented in a study conducted in the Pearl River, along Guangzhou city, China (Yan *et al.*, 2019). However, it is important to note that inefficient plastic waste management strategy is the central idea of the consequent microplastics pollution. If the waste management is good, plastic waste can be reduced, even if the area is highly industrialized (Rajmohan *et al.*, 2019). Meanwhile for river water, the downstream were documented to hold greater abundance, with average abundance of 0.0072 ± 0.0028 particles/m³ as compared to the upstream, with an average of 0.0051 ± 0.0017 particles/m³. Higher abundance of microplastics in river water downstream is significantly contributed by the substantial anthropogenic pressures from fisheries and tourism activities that are present in the area. A study in the Snake and Lower Columbia Rivers revealed similar findings (Kapp & Yeatman, 2018). However, no significant difference was observed in the abundance of microplastics in river water between the sampling sites.

Site	Microplastics abundance in river sediment (particles/kg)	Microplastics abundance in river water (particles/m ³)
1	57.22 ± 36.75	
2	85.56 ± 47.18	0.0051 + 0.0017
3	149.44 ± 77.00	0.0031 ± 0.0017
4	120.00 ± 41.63	
5	96.67 ± 59.32	
6	83.89 ± 52.95	0.0072 + 0.0028
7	70.56 ± 57.26	0.0072 ± 0.0028
8	114.44 ± 92.86	
Average	101.39 ± 54.69	0.0062 ± 0.0022

Table 4.14 : The abundance of microplastics in river sediment and river water of Sepetang River.

4.4.1.2 Abundance of Microplastics According to Type

Same trend of types dominancy was observed at both river sediment and river water samples in Sepetang River (Figure 4.1 and Figure 4.2). Film type was the most dominant with 50% in river sediment and 63% in river water samples. These could possibly originated from discarded plastics by the local villagers of nearby human settlement or littered by the tourists and visitors that came for recreational purposes. According to literatures, film microplastics may be derived from the decomposition of plastic packaging materials (Antunes *et al.*, 2013).



Figure 4.1: Percentage of microplastic particles by type in river sediment of Sepetang River.



Figure 4.2: Percentage of microplastic particles by type in river water Sepetang River.

Line constituted the second most dominant type with a proportion of 35% in river sediment samples, and 21% in river water samples. This probably sourced from fishing activities that were present along the river. In addition, atmospheric deposition and surface runoff are also potential sources of line microplastics (Browne *et al.*, 2011). The amount of microplastics of foam type is the least, accounting for only 1% which indirectly indicates the insignificant usage of foam type packaging by nearby restaurants and villagers. In overall, no significant difference was observed in the abundance of microplastics by type between river sediment and river water.

4.4.1.3 Abundance of Microplastics According to Size

Different proportions of microplastic sizes were observed for river sediment and river water samples. Microplastics in the sediments encompassed of larger size range in which the amount of microplastics decreased as the length decreased (Figure 4.3). 29% of the microplastics in river sediments were in the range of 1.0 to 5.0 mm, followed by 0.5 to 1.0 mm, 0.1 to 0.5 mm, and <0.1 mm at 27%, 26%, and 18%, respectively. The results were contradictory to most freshwater studies which recorded higher proportions of smaller microplastics such as in Wei River, China and Lake Garda, Italy (Imhof *et al.*, 2013; Ding *et al.*, 2019).



Figure 4.3: Percentage of microplastic particles by size in river sediment of Sepetang River.

On the other hand, a higher portion of smaller size microplastics were identified in river water, dominated by microplastics of < 0.1 mm in size at 37% (Figure 4.4). The dominance of smaller microplastics in surface water were also observed in a study conducted along rivers in the Tibet Plateau (Jiang *et al.*, 2019). The high number of small-sized microplastics may be reasoned by greater decomposition rate of larger plastic wastes into smaller plastic particles (Wu *et al.*, 2018). Overall, significant difference of microplastics abundance by size was observed between river sediment and river water [ANOVA, F(1,4)= 39.690, p=0.003].





4.4.1.4 Abundance of Microplastics According to Colour

River sediments were documented to be dominated by white microplastics with 39%, as compared to 13% in river water (Figure 4.5 and Figure 4.6). The high proportion of white colour category is partly due to the contribution of white thread-like user plastics mainly of weaved plastic bag and rope monofilaments, while transparent microplastics mainly originate from the weathering of sheet-like user plastics such as plastic bags and food wrapping (Yaghmour *et al.*, 2018). Meanwhile, transparent microplastics were the most dominant in river water at 38%, as compared to 23% in river sediment.



Figure 4.5: Percentage of microplastic particles by colour in river sediment of Sepetang River.



Figure 4.6: Percentage of microplastic particles by colour in river water of Sepetang River.

Smaller proportions were observed in both matrices for red- (3% in river sediment, 8% in river water), yellow- (4% in river sediment, 8% in river water), others (11% in river sediment, 8% in river water) and blue-coloured microplastics (10% in river sediment, 4% in river water). A significant difference of microplastics abundance by colour was observed between river sediment and river water of Sepetang River [ANOVA, F(1,4)= 39.690, p=0.003].

4.4.2 Serkam River, Malacca

4.4.2.1 Total Abundance of Microplastics

Microplastics were detected in all samples collected along Serkam River, with average abundance of 31.88 ± 10.95 particles/kg and 0.0028 ± 0.0014 particles/m³ in river sediment and river water, respectively (Table 4.15). In general, high abundance of microplastics in both river sediment and river water were detected downstream, all of which were observed to be caused by fishing and recreational activities, as well as, from several eateries present nearby the river. Literatures have shown that intensive human activities may increase the abundance, since more diverse sources of inputs of waste plastics are associated (Zhang *et al.*, 2017). Overall, no statistical difference of microplastics abundance was observed between the sampling sites in both river sediment and river water.
Site Microplastics abundance in river sediment (particles/kg)		Microplastics abundance in river water (particles/m ³)		
1	24.45 ± 16.19			
2	22.78 ± 3.85	0.0020 + 0.0012		
3	22.22 ± 8.39	0.0020 ± 0.0012		
4	23.33 ± 12.02			
5	40.56 ± 19.46			
6	36.67 ± 7.26	0.0026 + 0.0016		
7	31.11 ± 25.84	0.0030 ± 0.0010		
8	53.89 ± 46.97			
Average	31.88 ± 10.95	0.0028 ± 0.0014		

Table 4.15: The abundance of microplastics in river sediment and river water of Serkam

 River.

4.4.2.2 Abundance of Microplastics According to Type

Film type was prevalent, with a proportion of 43% in river sediment, followed by fragment at 34% of microplastics (Figure 4.7), and at 42% in river water, followed by fragment at 27% (Figure 4.8). Similar to this study, high proportions of film and fragment were also recorded in the sediment and water from Lake Hovsgol, Mongolia (Free *et al.*, 2014) and Tamar Estuary, UK (Sadri & Thompson, 2014). Overall, no significant difference was observed in the types of microplastics between the sediment and river water samples of Serkam River.



Figure 4.7: The abundance of microplastics in river sediment and river water of Serkam River.



Figure 4.8: Percentage of microplastic particles by type in river water of Serkam River.

4.4.2.3 Abundance of Microplastics According to Size

River sediments was identified to contain greater abundance of larger microplastics, with the dominance of 1.0 to 5.0 mm in size (Figure 4.9). The larger size fractions of microplastics were abundant since they tend to be trapped in the sediment. Hurley & Nizzetto (2018) reported that small particles can easily be carried away by runoff, thus larger ones remained in the sediment. The dominance of similar size fractions was recognized in a study conducted in Pearl River along Guangzhou City (Yan *et al.*, 2019).



Figure 4.9: Percentage of microplastic particles by size in river sediment of Serkam River.

Meanwhile, the smaller size fractions of 0.5 to 1.0 mm were the most dominant in river water, which accounted for about 38% of microplastics (Figure 4.10). This is highly attributed to the low density of smaller microplastics which tend to float in the water. It is reported that particle size showed significant effect on the fate and retention of microplastics in river (Besseling *et al.*, 2017). A significant difference of microplastics abundance by size was observed between river sediment and river water [ANOVA, F(1,4)=39.690, p=0.003].



Figure 4.10: Percentage of microplastic particles by size in river water of Serkam River.

4.4.2.4 Abundance of Microplastics According to Colour

Transparent and white microplastics were prevalent in river sediment, at 37% and 30%, respectively (Figure 4.11). This could be derived from fragmentation of plastic bags and food wrapping. River water samples on the other hand, were dominated by black microplastics, with a proportion of 34% (Figure 4.12).



Figure 4.11: Percentage of microplastic particles by colour in river sediment of Serkam River.



Figure 4.12: Percentage of microplastic particles by colour in river water of Serkam River.

High proportions of black microplastics was believed to originate from old tyres used as fenders on the wooden fishermen's jetty. Tyres are not only made of rubber but they are a complex blend of various materials and chemicals, including different types of plastic (Paul, 2018). The finding is in accordance with the microplastics identified in the Charleston Harbor and Winyah Bay, USA (Gray *et al.*, 2018). A significant difference of microplastics abundance by colour was observed between river sediment and river water [ANOVA, F(1,6)=8.265, p=0.028].

4.4.3 Ayer Masin River, Johor

4.4.3.1 Total Abundance of Microplastics

Microplastics were widely distributed in all samples of river sediment, with average abundance of 42.92 ± 20.19 particles/kg (Table 4.16). Statistically significant difference of microplastics abundance in river sediment was observed between the sampling sites [ANOVA, F(2,21)=8.28, p=0.002]. The highest abundance of microplastics was recorded at Site 3, in the upstream of Ayer Masin River, with average abundance of 61.67 ± 55.30 particles/kg. Similarly, greater abundance of microplastics in samples of river water was also recorded upstream, averaged at 0.0112 ± 0.0092 particles/m³. This is nearly twice as much of that as compared to the downstream, with an average abundance of 0.0090 ± 0.0012 particles/m³.

Site	Microplastics abundance in river sediment (particles/kg)	Microplastics abundance in river water (particles/m ³)		
1	42.22 ± 9.77			
2	33.89 ± 12.62	0.0112 + 0.0002		
3	61.67 ± 55.30	-0.0112 ± 0.0092		
4	27.78 ± 12.06			
5	52.22 ± 18.36			
6	46.11 ± 19.32			
7	37.78 ± 30.57	0.0090 ± 0.0012		
8	41.67 ± 33.29			
Average	42.92 ± 20.19	0.0101 ± 0.0052		

Table 4.16: The abundance of microplastics in river sediment and river water of Ayer

 Masin River.

The high abundance of microplastics is highly associated with heavy local fishery industries occurring within the area, aggravated by the presence of illegal dumping of waste observed at Site 3. It has been reported that the intrusion of plastic wastes into the river environment is vastly intensified by inefficient waste management system (Jambeck *et al.*, 2015). A similar pattern was observed in the Douro River, Portugal where higher microplastics occurrence was found in the middle part of the river close to greater

anthropogenic activities, with lower microplastics concentrations further the downstream (Rodrigues *et al.*, 2019).

4.4.3.2 Abundance of Microplastics According to Type

Dominant type of microplastics was film in both river sediment and river water samples, constituting 63% and 44%, respectively (Figure 4.13 and Figure 4.14). The film microplastics originate from the fragmentation of single-use plastic bags and food wrappers which were probably littered by villagers from nearby human settlement. Plastic items are indisputably abundant since they are lightweight, strong and cheaply available (Jayasiri *et al.*, 2013). Line constituted the second most dominant type with 21% in river sediment samples, and 27% in river water samples, followed by fragment, with 14% and 22%, respectively.

Similar finding was observed in the Ciwalengke River, Indonesia in which line particles were found more often (65%) than the fragment (35%) in both river sediment and river water (Alam *et al.*, 2019). The occurrence of line microplastics may come from the degradation of fishing gears, as well as, sewage that contains lines from washing of textiles (Claessens *et al.*, 2011) while fragments might be, to a great extent, attributed to the decomposition of many plastic wastes, such as agricultural tools, plastic packaging materials, plastic woven bags and plastic seed bags (Antunes *et al.*, 2013).

River water were observed to hold a greater percentage of foam and pellet, at 2% and 5%, respectively, as compared to river sediment, which constituted of only 1% for each type. This is possibly due to the lightweight properties of these plastics which tend to float and accumulate on the water surface (Galgani *et al.*, 2015). Foam microplastics may come from the degradation of polystyrene packaging materials littered during leisure fishing while pellets may come from industrial effluent or domestic sewage as they are widely used as material for plastic production, and cosmetic scrubbers in many personal

care products (Napper *et al.*, 2015). Overall, no statistical difference was observed in the abundance by type of microplastics between river sediment and river water of Ayer Masin River.



Figure 4.13: Percentage of microplastic particles by type in river sediment of Ayer Masin River.



Figure 4.14: Percentage of microplastic particles by type in river water of Ayer Masin River.

4.4.3.3 Abundance of Microplastics According to Size

River sediment and river water of Ayer Masin River were revealed to have microplastics of comparable size fractions, mostly within 0.5 to 1.0 mm in size, constituting 33% in river sediment and 40% in river water (Figure 4.15 and Figure 4.16).

The trend of size fractions was continued by the size range of 0.1 to 0.5 mm in river sediment and river water, constituting 25% and 23%, respectively. Larger microplastics, of 1.0 to 5.0 mm were the least recorded in both matrices, comprising of 18% in river sediment and 17% in river water. The higher number of small-sized microplastics may be reasoned by the decomposition of larger plastic wastes into smaller microplastics particles. These trends were similar to the discoveries in the Three Gorges Reservoir and Qinghai Lake, China (Di & Wang, 2018; Xiong *et al.*, 2018; Ding *et al.*, 2019). Overall, no statistical difference was observed for microplastic abundance by size between river sediment and river water of Ayer Masin River.



Figure 4.15: Percentage of microplastic particles by size in river sediment of Ayer Masin River.



Figure 4.16: Percentage of microplastic particles by size in river water of Ayer Masin River.

4.4.3.4 Abundance of Microplastics According to Colour

White microplastics clearly prevailed in both river sediment and river water, at 36% and 29%, respectively, followed by transparent microplastics at 25% in river sediment, and 18% in river water (Figure 4.17 and Figure 4.18). Greater abundance of yellow microplastics of 23% were observed in river water, as compared to 7% in river sediment. White, transparent, and yellow microplastics may come from the fragmentation of widely used plastic bags and food wrapping. In addition, blue constituted 14% of microplastics in river sediment, while in contrast, blue colour was the least identified in river water, accounting for only 3% of microplastics. Blue-coloured microplastics probably originate from the degradation of multiple plastic products that are widely used in daily life such as in clothing and packaging, as well as, from the blue rope monofilaments utilized in fishing and tourism activities (Zhang *et al.*, 2015; Wang *et al.*, 2017). Significant difference was observed for microplastic abundance by colour between river sediment and river water of Ayer Masin River [ANOVA, F(1,6)=8.265, p=0.028].







Figure 4.18: Percentage of microplastic particles by colour in river water of Ayer Masin River.

4.4.4 Sedili Besar River, Johor

4.4.4.1 Total Abundance of Microplastics

Microplastics were widely present in all samples of river sediment collected along Sedili Besar River, with an average abundance of 32.36 ± 14.03 particles/kg (Table 4.17). The abundance varied from 15.00 ± 2.87 particles/kg (Site 3) to 42.78 ± 43.15 particles/kg (Site 5). However, no significant difference of microplastics abundance in river sediment was observed between the sampling sites. In samples of river water, greater abundance of microplastics were recorded downstream, with average abundance 0.0051 ± 0.0062 particles/m³, which is nearly three times greater than that upstream (0.0013 ± 0.0004 particles/m³). This may have been contributed by the fisheries, aquaculture and tourism activities which are recognized to be the main anthropogenic activities in the area. Similarly, greater anthropogenic factors were known to exert greater impacts on the abundance of microplastics in the Hanjiang River and Yangtze River in China (Wang *et al.*, 2017b), which is in agreeable to findings from this study.

Site	Microplastics abundance in river sediment (particles/kg)	Microplastics abundance in river water (particles/m ³)		
1	40.00 ± 24.04			
2	32.78 ± 31.51			
3	15.00 ± 2.87	0.0013 ± 0.0004		
4	21.11 ± 9.18			
5	42.78 ± 43.15			
6	34.45 ± 17.98	0.0051 ± 0.0062		
7	32.22 ± 14.37			
8	40.56 ± 35.84			
Average	32.36 ± 14.03	0.0032 ± 0.0031		

Table 4.17: The abundance of microplastics in river sediment and river water of Sedili

 Besar River.

4.4.4.2 Abundance of Microplastics According to Type

Film type which most likely arises from the breakdown of many plastic products such as plastic carry bags, packaging materials and plastic containers, was the dominant type found in both river sediment and river water, constituting 63% and 72% of microplastics, respectively (Figure 4.19 and Figure 4.20).



Figure 4.19: Percentage of microplastic particles by type in river sediment of Sedili Besar River.



Figure 4.20: Percentage of microplastic particles by type in river water of Sedili Besar River.

Line was the second most abundant type identified, constituting 20% in river sediment and 24% in river water. This is in accordance with the finding in Atoyac River basin, Mexico (Shruti *et al.*, 2019). The presence of line type microplastics has been commonly attributed to the release of synthetic fibers from textiles and garments during washing, or from the nets used in fishing activities (Browne *et al.*, 2011; Almroth *et al.*, 2018). Additionally, fragment type microplastics which may also originate from the fragmentation of many plastics products were recorded in samples of river sediment, with a proportion of 15%. Fragment type was absent in samples of river water. A small proportion of foam type microplastics were evident, with only 4% abundance in river water samples. Overall, no statistical difference was observed in the types of microplastics between these two matrices.

4.4.4.3 Abundance of Microplastics According to Size

Different microplastic size fractions were observed between river sediment and river water samples of Sedili Besar River. In river sediment, greater dominance was of larger-sized microplastics of 1.0 to 5.0 mm in size (Figure 4.21).



Figure 4.21: Percentage of microplastic particles by size in river sediment of Sedili Besar River.

The greater dominance of larger-sized microplastics in the sediment of Sedili Besar River can be due to higher densities of larger microplastics, causing the tendency of settling in the sediment. This is aggregable to findings in other researches (Nizzetto *et al.*, 2016; Di & Wang, 2018). In the present study, the proportion decreased with the decreased in size, in which microplastics of < 0.1 mm in size constituted the least, with a proportion of 17%. In contrast to river water, most microplastics were in the range of 0.1 to 0.5 mm in size of 39% in proportion, followed by 32% of microplastics that were 0.5 to 1.0 mm in size (Figure 4.22).



Figure 4.22: Percentage of microplastic particles by size in river water of Sedili Besar River.

A study conducted in rivers of Shanghai, China recorded similar findings in which 31% of microplastics identified were smaller than 0.01 mm in size (Peng *et al.*, 2018). Furthermore, microplastics with largest-sized fractions (i.e. 1.0 to 5.0 mm) constituted the least in river water samples, which accounted for only 7%. A statistical difference was observed for microplastic abundance by size between these two matrices [ANOVA, F(1,4)=39.690, p=0.003].

4.4.4.4 Abundance of Microplastics According to Colour

River sediment of Sedili Besar River encompass great abundance of transparent microplastics (50%) which could come from fishing activities since transparent plastic fishing lines and nylon nets were the commonly used fishing tools in the area. Other dominant colours identified were white (15%), blue (9%), black (8%), and others (10%) (Figure 4.23). Lower proportion were observed for red- and yellow-coloured microplastics in river sediment, at only 5% and 3%, respectively.



Figure 4.23: Percentage of microplastic particles by colour in river sediment of Sedili Besar River.

On the other hand, white microplastics were prevalent in river water samples, which accounted for 32%, with other dominant colours being blue (18%), black (21%), yellow (11%) and transparent (11%) (Figure 4.24). The finding was in accordance with the study conducted in Ebro River, Spain which documented higher concentration of white microplastics in river water (Simon-Sánchez *et al.*, 2019). Overall, significant difference was observed for microplastic abundance by colour between river sediment and river water of Sedili Besar River [ANOVA, F(1,6)=8.265, p=0.028].



Figure 4.24: Percentage of microplastic particles by colour in river water of Sedili Besar River.

4.4.5 Cherating River, Pahang

4.4.5.1 Total Abundance of Microplastics

All sampling sites along Cherating River were recorded to be polluted with microplastics, with average abundance of 32.15 ± 20.32 particles/kg in river sediment, and 0.0038 ± 0.0015 particles/m³ in river water (Table 4.18). A statistical significant difference of microplastics abundance in river sediment was observed between the sampling sites [ANOVA, F(2,21)=4.08, p=0.003].

Table 4.18: The abundance of microplastics in river sediment and river water of Cherating River.

Site	Microplastics abundance in river sediment (particles/kg)	Microplastics abundance in river water (particles/m ³)		
1	14.44 ± 10.05			
2	22.22 ± 8.55	0.0005 + 0.0002		
3	23.33 ± 7.26	0.0005 ± 0.0003		
4	37.78 ± 50.40			
5	51.11 ± 57.16			
6	50.00 ± 38.44	0.0070 + 0.0022		
7	27.22 ± 25.46	0.0070 ± 0.0033		
8	31.11 ± 42.34			
Average	32.15 ± 20.32	0.0038 ± 0.0015		

Site 5 of the river which was located adjacent to a commercial fishing pond, recorded the highest abundance of microplastics, with an average of 51.11 ± 57.16 particles/kg. Likewise, greater microplastics pollution in river water was identified downstream as compared to the upstream, with average abundance of 0.0070 ± 0.0033 and 0.0005 ± 0.0003 particles/m³, respectively. Nevertheless, no significance difference of microplastics abundance in river water was observed between these two sections (i.e. the upstream and the downstream).

4.4.5.2 Abundance of Microplastics According to Type

Film was the most dominant type of microplastics in both river sediment (64%) and river water (48%) of Cherating River (Figure 4.25 and Figure 4.26). High abundance of

film type microplastics may be generated from the fragmentation of large plastic litters such as plastic bags, confectionary and convenience plastic food wrappings that were carelessly discarded from various tourism activities carried along the river. These litters can be deposited directly into the river or onto the riverbank and then washed into the river by surface runoff after heavy rain (Eo *et al.*, 2019).

Microplastics of line type was the second most abundant in river sediment samples, with a proportion of 18%. Fishing activities, which also have a relevant role in the study area, are the potential sources of line type microplastics. Fishing materials that are commonly made of synthetic fibers are extensively used and their degradation or direct disposal in the river environment might lead to their degradation to microsize (Andrady, 2011).



Figure 4.25: Percentage of microplastic particles by type in river sediment of Cherating River.



Figure 4.26: Percentage of microplastic particles by type in river water of Cherating River.

On the other hand, fragments, possibly originated from the degradation of hard plastic items such as food containers and drinking bottles, was the second most abundant type in samples of river water, with 31%, followed by line at 17%. Foam which may come from polystyrene packaging materials, constituted the least in samples of river sediment and river water, constituting 1% and 4%, respectively. Overall, no significant difference was observed in the types of microplastics in samples of river sediment and river water of Cherating River.

4.4.5.3 Abundance of Microplastics According to Size

The most dominant size fraction of microplastics present in samples of river sediment of Cherating River was < 0.1 mm in size, which accounted for 31%, while the aforementioned size fraction was the least identified in river water samples, with a proportion of 7% (Figure 4.27 and Figure 4.28).



Figure 4.27: Percentage of microplastic particles by size in river sediment of Cherating River.



Figure 4.28: Percentage of microplastic particles by size in river water of Cherating River.

This is believed to be attributed by the interactions of aggregates, biofouling, and faecal matter that lead to the increased in density of smaller microplastics, and along with decreased buoyancy, thus enhance their settling. Similar findings were also reported by other researches (Long *et al.*, 2015; Porter *et al.*, 2018).

Other studies have also observed smaller-sized microplastics in freshwater sediments such as in the St. Lawrence River, America (Castañeda *et al.*, 2014) and in the sediment

of Lake Ontario, Canada (Corcoran *et al.*, 2015). Other dominant size fractions of microplastics in the sediment were found to vary from 0.5 to 1.0 mm, 0.1 to 0.5 mm, and 1.0 to 5.0 mm, constituting 25%, 24%, and 20%, respectively.

As for river water, a high proportion of microplastics within the size of 0.5 to 1.0 mm were recorded, which accounted for 45% of microplastics. Similar finding was observed in the lower reaches of Yangtze River, China (Xiong *et al.*, 2019).

Overall, significant difference was observed for microplastic abundance by size between river sediment and river water of Cherating River [ANOVA, F(1,4)= 39.690, p=0.003].

4.4.5.4 Abundance of Microplastics According to Colour

Black microplastics clearly prevailed in river sediment samples, with 32%, while only 14% recorded in samples of river water (Figure 4.29 and Figure 4.30). The finding was in accordance with microplastic study in the Antuã River, Portugal (Rodrigues *et al.,* 2018).

On the other hand, white microplastics were prevalent in river water samples, which accounted for 54% of microplastics, while only 17% recorded in river sediment samples. Additionally, samples of river sediment and river water recorded a similar proportion of transparent microplastics of 25%.









Colours like blue, yellow, red and others constituted less than 10% in river sediment samples, whereas colours grouped under 'others' such as green, brown, and purple, constituted the least in river water samples, constituting 7% of microplastics. The results were in contrast with a study in the Saigon River, Vietnam which documented a high variety of colours with a predominance of blue microplastics (Lahens *et al.*, 2018). Overall, significant difference was observed for microplastic abundance by colour

between river sediment and river water of Cherating River [ANOVA, F(1,6)=8.265, p=0.028].

4.4.6 Semerak River, Kelantan

4.4.6.1 Total Abundance of Microplastics

Microplastics were widely present in all river sediment samples along Semerak River, with an average abundance of 22.64 \pm 12.21 particles/kg (Table 4.19). Statistically significant difference of microplastics abundance in river sediment was observed between the sampling sites [ANOVA, F(2,21)=8.42, p=0.002]. The abundance in river sediment varied from 13.33 \pm 7.64 particles/kg at the downstream (Site 8), to 32.78 \pm 16.86 particles/kg at the upstream (Site 3). Correspondingly, greater abundance of microplastics in river water samples were also recorded in the upstream, of average abundance 0.0137 \pm 0.0046 particles/m³.

 Table 4.195: The abundance of microplastics in river sediment and river water of Semerak River.

Site	Microplastics abundance in river sediment (particles/kg)	Microplastics abundance in river water (particles/m ³)		
1	26.11 ± 19.32			
2	30.56 ± 23.35	0.0127 + 0.0046		
3	32.78 ± 16.86	-0.0137 ± 0.0046		
4	15.00 ± 14.43			
5	16.67 ± 12.02			
6	31.11 ± 17.66			
7	15.56 ± 5.85	0.0037 ± 0.0033		
8	13.33 ± 7.64			
Average	22.64 ± 12.21	0.0097 ± 0.0050		

High microplastics abundance identified at the upstream of the river was partly due to the presence of human settlement and agricultural activities, which brought in plastics into the river from the result of improper waste management. It is worth noting that improper waste management and excessive agricultural activities might enhance the release of microplastics into the aquatic environment, as evident in Wei River, China (Ding *et al.*, 2019). As rice crop production in the area involved practices such as ploughing, flooding, sowing, re-flooding, and draining the fields before the harvest, the practice of ploughing might release microplastics that are trapped in soils, while the practices of flooding and draining will act similar to rainfall run-off, dragging microplastics into the river. This is similar to findings of other researches (Simon-Sánchez *et al.*, 2019).

4.4.6.2 Abundance of Microplastics According to Type

Film microplastics were identified to be the dominant type of microplastics in both river sediment and river water samples, constituting 45% and 44%, respectively (Figure 4.31 and Figure 4.32). Films can originate from leisure fishing activities, as well as, from nearby human settlement and chalets, through fragmentation of widely used plastic items like plastic bags and food wrapping. Films may also be transported by wind and subsequently deposited in aquatic environments (Cole *et al.*, 2011; Dris *et al.*, 2016).



Figure 4.31: Percentage of microplastic particles by type in river sediment of Semerak River.



Figure 4.32: Percentage of microplastic particles by type in river water of Semerak River.

Fragments, which are widely used in food packaging, drinking bottle, and durable bags, were the second most abundant in samples of river sediment, at 32%, followed by line at 21%. In contrary, lines which may originate from the fragmentation of fishing nets and ropes or from sewage containing fibers, was the second most abundant type in samples of river water, with 28%. This is followed by fragment at 16%.

Meanwhile, pellet constituted the least in both river sediment and river water samples, constituting 1% and 4% of microplastics, respectively. The low abundance of pellet recorded in this study was in contrary with several freshwater studies such as in the Great Lakes, USA, as well as, in the Rhine River and Danube River in Europe (Eriksen *et al.*, 2013; Lechner *et al.*, 2014; Mani *et al.*, 2015). Overall, no significant difference was observed in the types of microplastics between river sediment and river water samples of Semerak River.

4.4.6.3 Abundance of Microplastics According to Size

Different microplastic sizes were observed in samples of river sediment and river water (Figure 4.33 and Figure 4.34). Most microplastics extracted from the sediments were 0.5 to 1.0 mm, at 40%, followed by 0.1 to 0.5 mm, at 33%. In contrast, 0.1 to 0.5 mm size fraction was the most dominant in river water, which accounted for about 48% of

microplastics, followed by 0.5 to 1.0 mm, constituting 31% of microplastics. This is highly attributed to the lower densities of smaller microplastics which tend to float in water while larger microplastics of higher densities tend to be retained in the sediment (Alam *et al.*, 2019).



Figure 4.33: Percentage of microplastic particles by size in river sediment of Semerak River.



Figure 4.34: Percentage of microplastic particles by size in river water of Semerak River.

A study in the Nakdong River, South Korea recorded a similar finding with the peak size range 0.15 mm in river water (Eo *et al.*, 2019). Also, the preponderance of small microplastics (< 0.5 mm) has been reported in many other studies worldwide (Dikareva & Simon, 2019). Overall, a statistical difference was observed for microplastic abundance

by size between river sediment and river water of Semerak River [ANOVA, F(1,4)= 39.690, p=0.003].

4.4.6.4 Abundance of Microplastics According to Colour

Transparent- and white-coloured microplastics were the two most dominant colours in both sediment and water of Semerak River (Figure 4.35 and Figure 4.36).



Figure 4.35: Percentage of microplastic particles by colour in river sediment of Semerak River.



Figure 4.36: Percentage of microplastic particles by colour in river water of Semerak River.

Transparent- and white-coloured microplastics are often the resulting product of weathering of multiple plastic products that are widely used in daily life such as in clothing and packaging. This is also reported by other researches (Zhang *et al.*, 2015; Wang *et al.*, 2017b). Smaller proportions of less than 10% were observed for blue, black, and yellow microplastics in both river sediment and river water. Microplastic study in the rivers of Shanghai, China presented similar finding where these colours were the minority among all microplastics extracted (Peng *et al.*, 2018). Additionally, red-coloured microplastics accounted for only 2% in river sediment, while no red microplastic was identified in river water samples. In general, no significant difference was observed in the colours of microplastics between river sediment and river water of Semerak River.

4.4.7 Comparative Study of the Rivers

Figure 4.37 depicts the abundance of microplastics in river sediment and river water of selected rivers in Peninsular Malaysia.





Sepetang River clearly showed the dominance of microplastics abundance in river sediment of the six rivers studied, with average abundance of 101.39 ± 54.69 particles/kg.

The highest incidence of contamination is not surprising since this river flows through some extensive industrial areas including manufacturers of plastic products, as well as, areas with intense fishing and tourism activities.

Another potential source of microplastics includes the contribution from the point source of microplastics input through improper solid waste management, observed near the sampling sites. Similar sources of microplastics input are evident in Citarum River, Indonesia where plastic manufacturers and defective waste management promote waste plastic entering into the river ecosystem (Alam *et al.*, 2019).

Globally, the finding in Sepetang River $(101.39 \pm 54.69 \text{ particles/kg})$ was in accordance with rivers in the Tibet Plateau, China (50 to 195 particles/kg) (Jiang *et al.*, 2019) and in the Bloukrans River, South Africa (6.3 to 160.1 particles/kg) (Nel *et al.*, 2018), but was relatively lower than that in the Xiangjiang River, China (27 to 866 particles/kg) (Wen *et al.*, 2018) and 30 times lower than in the Rhine River, Germany (228 to 3,763 particles/kg) (Klein *et al.*, 2015).

Semerak River recorded the lowest microplastics abundance in river sediment, with an average of 22.64 ± 12.21 particles/kg. Nevertheless, in this river, a remarkably high abundance of microplastics was detected in river water. This is highly attributed to the aquaculture activities that are present in the area. As reported in other studies, elevated microplastic concentrations were observed in countries with high levels of urbanization (Graca *et al.*, 2017) and human activities (Nor & Obbard, 2014) or close to fresh water discharges and aquaculture facilities (Vianello *et al.*, 2013).

Ayer Masin River, characterized by having moderate anthropogenic activities, with a predominance towards fisheries and tourism activities, was recognized to hold the greatest abundance of microplastics in river water. In comparison, the average microplastics abundance of 0.0101 ± 0.0052 particles/m³, was significantly lower than that to other freshwater microplastic studies in Asia such as in the Yangtze River Estuary (i.e. 2,516.7 ± 911.7 particles/m³) (Zhao *et al.*, 2014) and Hangjiang River in China (i.e. 2,933 ± 305.5 particles/m³) (Wang *et al.*, 2017a) but was comparable with marine microplastics studies in Southern Europe such as in Aveiro (i.e. 0.002 ± 0.001 particles/m³) and Lisbon in Portugal (i.e. 0.033 ± 0.021 particles/m³) (Frias *et al.*, 2016).

A statistically significant difference of microplastics abundance in river sediment was observed between the rivers [ANOVA, F(5,6)=45.01, p=0.000]. However, one-way analysis of variance showed statistically non-significant difference of microplastics abundance in river water between the rivers. Overall, the variation in microplastics concentration among sampling sites may have been a result of differences in anthropogenic impacts, point sources of microplastic input, as well as, the influences of natural factors such as currents and winds (Gray *et al.*, 2018).

4.5 POPs Accumulation of Selected Rivers

Figure 4.38 illustrates the concentrations of POPs found in river sediment of selected rivers in Peninsular Malaysia. In total, four types of POPs were reported to be present in river sediment namely 1,2-Benzenedicarboxylic acid, mono(2-ethylhexyl) ester (MEHP); 1,2-Benzenedicarboxylic acid, butyl decyl ester (BDP); Decanedioic acid, bis(2-ethylhexyl) ester (DEHS); and 1,2-Benzenedicarboxylic acid, diisooctyl ester (DIOP). DEHS and DIOP were the two dominant POPs present in river sediment in majority of the selected rivers.



Figure 4.38: Concentrations of POPs found in river sediment of selected rivers in Peninsular Malaysia.

The highest concentration of DEHS was recorded in Serkam River (60.15 ppm) while the highest concentration of DIOP was found in Sedili Besar River (677.49 ppm). Meanwhile, BDP, which was present in the lowest concentration among the other identified POPs (2.33 ppm), was only detected in the river sediment of Sepetang River.

Figure 4.39 illustrates the concentrations of POPs found in river sediment of selected rivers in Peninsular Malaysia.



Figure 4.39: Concentrations of POPs found in river water of selected rivers in Peninsular Malaysia.

High concentrations of DIOP were also reported in the majority of water samples collected from the studied rivers, with concentrations varied from 61.6 ppm in Sepetang River to 153.41 ppm in Cherating River. Meanwhile, DEHS and MEHP were present at low concentrations in river water. DEHS was only found in Cherating River (6.42 ppm) while MEHP was only detected in Sedili Besar River (19.13 ppm).

All of the four POPs identified were grouped in Phthalate esters (PAEs), and their presence was observed in rivers with moderate to high anthropogenic activities. Numerous studies indicate that the widespread of these contaminants, is due to the discharge of untreated effluents from industrial, agricultural and municipal activities (Abbassy *et al.*, 2018). PAEs which are widely used as plasticizers in households and industrial products, such as in children's toys, food packaging, lubricants, adhesives, paints, building materials, pharmaceuticals, and personal care products may have gradually released and migrate from the host polymers into the environment as they are not chemically bound to the polymer molecules, consequently accumulating (Mi *et al.*, 2019).

Table 4.20 summarizes the comparison of PAEs concentrations (ppm) with those measured in global rivers. In comparison with worldwide investigations in freshwater systems, the highest concentrations of PAEs recorded in river sediment of the present study (677.49 ppm) was comparable to Changjiang River, China and was two or three orders of magnitude higher than the rivers tabulated. Additionally, the value of PAEs in sediment of Sedili Besar River was almost 3,000 times more than the Kaveri River in India and the Chaohu Lake in China. The highest concentration of PAEs recorded in the river water (159.83 ppm) was significantly higher than those found in most river studies worldwide. Astonishingly, the value was almost 10,000 times more than the Klang River and almost 400,000 times more than the Rhone River in France.

Location	Concentrations	Reference	
	(ppm)		
River sediment			
Yellow River	9.29 - 50.69	Sha et al. (2007)	
HaiHe River	0.31–2.73	Chi (2009)	
Changjiang River, China	729.20–1545.8	Du <i>et al.</i> (2013)	
Qiantang River	1.56	Sun <i>et al.</i> (2013)	
Chaohu Lake, China	0.30	Kang et al. (2016)	
Jiulong River, Southeast China	0.0043-0.3947	Li et al. (2017)	
Kaveri River, India	0.28	Selvaraj et al. (2015)	
Sepetang River	423.52	Present study	
Serkam River	70.85	Present study	
Ayer Masin River	299.74	Present study	
Sedili Besar River	677.49	Present study	
Cherating River	344.62	Present study	
Semerak River	73.04	Present study	
River water			
Songhua River, China	0.00226 - 0.0116	Gao <i>et al.</i> (2014)	
Jiulong River, China	0.00062 - 0.01243	Li et al. (2017)	
Rhone River, France	0.000407	Paluselli et al. (2018)	
Klang River Basin, Malaysia	0.0166	Tan (1995)	
Sepetang River	61.60	Present study	
Serkam River	90.50	Present study	
Ayer Masin River	122.98	Present study	
Sedili Besar River	19.13	Present study	
Cherating River	159.83	Present study	
Semerak River	128.13	Present study	

Table 4.20: Comparison of PAEs concentrations (ppm) with those measured in global rivers.

4.6 Comparative Study of the Rivers

4.6.1 Comparison of Microplastics Distribution of Selected Rivers along the West Coast and East Coast of Peninsular Malaysia

The rivers in the West Coast demonstrated greater abundance of microplastics, as compared to the East Coast, in both river sediment and river water, as illustrated in Figure 4.40 (river sediment) and Figure 4.41 (river water).



Figure 4.40: Abundance of microplastics in river sediment of selected rivers along the West Coast and the East Coast of Peninsular Malaysia.



Figure 4.41: Abundance of microplastics in river water of selected rivers along the West Coast and the East Coast of Peninsular Malaysia.

The average abundance of microplastics identified in the sediment from the West Coast and the East Coast were 176.19 ± 37.35 particles/kg and 87.15 ± 5.55 particles/kg, respectively. A significant difference was observed on the abundance of microplastics in river sediment between the West Coast and the East Coast of Peninsular Malaysia [ANOVA, F(5,42)=26.945, p<0.05].

As for river water, an average abundance of 0.0190 ± 0.0037 particles/m³ of microplastics were recorded in rivers of the West Coast, while an abundance of 0.0167 ± 0.0036 particles/m³ of microplastics were recorded in rivers of the East Coast. However, no significant difference was observed on the abundance of microplastics in river water between the West Coast and the East Coast of Peninsular Malaysia.

The higher abundance recorded in rivers of the West Coast as compared to the East Coast may be reasoned to their geographical locations which drained into the Straits of Malacca. It is one of the most important shipping waterways in the world, which serve part of a major maritime trade route between the Indian and the Pacific Ocean (Chong & Lam, 2013). The Straits which support a vast volume of merchant shipping with more than 200 vessels and thousands of fishing boats passing through the Straits on a daily basis, undoubtedly contribute to a tremendous plastics litter load in adjoining rivers and seas (Lebreton *et al.*, 2012; Qu & Meng, 2012).

On top of that, it is also attributed to the proximity to a greater anthropogenic source of microplastics in the West Coast, particularly along Sepetang and Ayer Masin rivers. As previously mentioned, studies have shown that sampling areas with extensive anthropogenic activities (i.e. high industrialization and urbanization) are expected to host high levels of microplastics contamination (Lambert *et al.*, 2014; Duis & Coors, 2016; Yan *et al.*, 2019).

Table 4.21 depicts the characteristics of microplastics along the West Coast and the

East Coast of Peninsular Malaysia.

Table 4.21:	The characte	eristics of 1	nicroplastics	along the	West Coast	t and the	East	Coast
of Peninsula	ar Malaysia.							

Characteristics	West Coast		East Coast		
	River	River water	River	River water	
	sediment (%)	(%)	sediment (%)	(%)	
Туре	Туре				
Line	18	29	18	22	
Fragment	12	29	12	17	
Film	68	42	69	61	
Foam	2	0	1	0	
Pellet	0	0	0	0	
Size					
< 0.1	13	30	7	21	
0.1 - 0.5	18	28	15	42	
0.5 - 1.0	33	40	33	32	
1.0 - 5.0	36	2	45	5	
Colour					
Transparent	14	12	40	8	
Black	15	33	4	13	
Blue	14	5	11	16	
Red	1	0	3	0	
Yellow	9	12	3	10	
White	32	26	22	42	
Others	15	12	17	11	

*highest percentage marked bold

Overall, film was revealed to be the dominant type of microplastics in rivers along the West Coast and the East Coast, recorded in both river sediment (i.e. 68% in the West Coast and 69% in the East Coast) and river water (i.e. 42% in the West Coast and 61% in the East Coast).

Microplastics in river sediment of the West Coast and the East Coast were revealed to have a similar dominancy of size fraction of 1.0 to 5.0 mm in size, with a percentage of 36% in the West Coast and 45% in the East Coast. However, most of the microplastics in river water of the West Coast were revealed to be within 0.5 to 1.0 mm in size (40%), whereas microplastics of 0.1 to 0.5 mm in size were prevalent in the East Coast (42%).

White microplastics clearly prevailed in river sediment across rivers of the West Coast (32%), while transparent microplastics were prevalent in river sediment across rivers of the East Coast (40%). Microplastics in river water on the other hand showed a dominance towards black microplastics along the West Coast (33%), and with a dominance towards white microplastics along the East Coast (42%) of Peninsular Malaysia.

4.6.2 Comparison of POPs Distribution in Selected Rivers along the West Coast and East Coast Peninsular Malaysia

Table 4.22 highlights the concentrations of POPs along the West Coast and the East Coast of Peninsular Malaysia.

No.	Compound name	Concentration (ppm)			
	• X	West	Coast	East	Coast
		River sediment	River water	River sediment	River water
1	1,2-Benzenedicarboxylic acid, mono(2-ethylhexyl) ester (MEHP)	10.7	NA	50.2	19.1
2	1,2-Benzenedicarboxylic acid, butyl decyl ester (BDP)	2.3	NA	NA	NA
3	Decanedioic acid, bis(2- ethylhexyl) ester (DEHS)	83.4	NA	44.9	6.4
4	1,2-Benzenedicarboxylic acid, diisooctyl ester (DIOP)	697.6	275.1	1,000.0	281.5
Tota	1	794.1	275.1	1,095.2	307.1

Table 4.22: The concentrations of POPs along the West Coast and the East Coast of Peninsular Malaysia.

In general, rivers in the East Coast were detected with greater concentrations of POPs (1,402.3 ppm), as compared to the rivers in the West Coast (1,069.2 ppm). Among the four types of POPs identified, DIOP were found to be present in all rivers, with the highest concentration reported in river sediment of the East Coast (1,000 ppm).

High concentration of DIOP also has been reported in China's rivers such as in the Guanting Reservoir, including the lakes in Shichahai and the lakes in Summer Palace,
Beijing (Meng *et al.*, 2014; Zheng *et al.*, 2014). DIOP which is a congener of PAEs, is continuously being released to the atmosphere, waters, soils, and garbage from indiscriminate disposal of phthalate-containing products, which then enter river environment through urban surface runoff, municipal effluent and dust deposition from agricultural fields.

PAEs contamination levels are strongly influenced by pervasive anthropogenic sources. Even though rivers in the East Coast were categorized to have moderate anthropogenic activities, greater PAEs pollution reported in the East Coast may be linked to the atmospheric deposition of these pollutants which is exacerbated by the presence of the Northeast (NE) Monsoon.

During the NE Monsoon, (i.e. from November to March), heavy rain and strong northeasterly winds could result in a greater atmospheric transport of the PAEs from source to sink (Zuraire *et al.*, 2018). It is primarily due to the fact that PAEs or POPs in general, exist in a free mobile state and may be transported to long distances due to their persistent in nature (Magdouli *et al.*, 2013; Wang *et al.*, 2015). Southwest (SW) Monsoon (i.e. from May to September) on the other hand, is dry and with the absence of strong wind (Daryabor *et al.*, 2015), hence, leading to a lower extent of PAEs pollution in rivers of the West Coast.

4.6.3 Comparison of Microplastics Abundance and POPs Concentration with Anthropogenic Activities of Selected Rivers

Figure 4.42 illustrates the relationship between the abundance of microplastics and concentration of POPs in river sediment, along with the hotspots of anthropogenic activities in selected rivers of Peninsular Malaysia.



Figure 4.42: The relationship between the abundance of microplastics and concentration of POPs in river sediment, along with the hotspots of anthropogenic activities in selected rivers of Peninsular Malaysia.

In general, the trend in the accumulation of microplastics in river sediment was coherent with the increase in anthropogenic activities, which were observed across all selected rivers. For instance, Sepetang River which was investigated to be most polluted with microplastics (101.39 ± 54.69 particles/kg), was due to the intense anthropogenic activities. A study conducted in rivers of Japan offered more evidence in which microplastic concentrations were significantly correlated with urbanization and population density, indicating that microplastic concentrations in river vastly depend on anthropogenic activities (Kataoka *et al.*, 2019).

Nevertheless, the greatest concentrations of POPs in river sediment was detected in Sedili Besar River (677.49 ppm), associated with having moderate anthropogenic activities. This may be explained by the presence of aquaculture activity which was observed to be a significant source of POPs pollution. This is agreeable to findings in other studies (Tsapakis *et al.*, 2010; Russell *et al.*, 2011). Undoubtedly, aquaculture has strongly contributed to local economic growth; however, it has also resulted in the rapid deterioration of aquatic ecosystems.

On top of that, the highest concentration of POPs reported in the sediment (i.e. 677.49 ppm) was approximately four times greater in magnitude as compared to the highest concentration reported in the river water (i.e. 159.83 ppm) (Figure 4.43).





The higher concentration of POPs recorded in the sediment as compared to in the river water is attributed to the hydrophobicity of POPs where the particles tend to accumulate in the sediments. Qiu *et al.* (2020) have quoted that POPs are absorbed rapidly by suspended particulate matter, some of which precipitate and accumulate in the sediments once delivered into the water column.

Furthermore, in the present study, it was found that aquaculture activity was not only evidenced to accumulate POPs in river environment, but also contribute to microplastic pollution. This was observable in Semerak River which held great abundance of microplastics and POPs concentrations in river water, despite having moderate anthropogenic activities. Such observation can be due to the fact that plastics are widely used in aquaculture which is intensified by constant exposure of river to this activity. In aquaculture, the species are grown on plastic polypropylene lines while polyethylene is extensively used as floating rigs and ropes (Andrady, 2011), hence deliberating microplastics to river environment (Mathalon & Hill, 2014). A study at an aquaculture site at Xiangshan Bay, China reported similar findings (Wu *et al.*, 2020).

As previously mentioned, POPs tend to accumulate in the sediment. In contrast, it was detected that microplastics concentration at aquaculture site, in this case Semerak River was less to accumulate in river sediment. This is due to the relatively low density of microplastic types used which tends to float on river water rather than to sink in the sediment. Specifically, polypropylene and polyethylene have a density of 0.88 - 0.96 g/cm³, therefore these particles tend to float on the water surface (Suaria & Aliani, 2014) or in suspension in the water column (Fossi *et al.*, 2012).

Apart from aquaculture activity, the presence of fishing pond in Cherating River, may correspondingly contributes to the high concentration of microplastics and POPs recorded in the river. In fact, the concentration of microplastics in the sediment and river water of Cheating and Sedili Besar rivers were almost similar of astounding 32.15 ± 20.32 particles/kg and 32.36 ± 14.03 particles/kg, respectively in river sediment, while 0.0038 ± 0.0015 particles/m³ and 0.0032 ± 0.0031 particles/m³ in river water, respectively. A study conducted in rivers of the Tibet Plateau concluded that fisheries, in general, were measured to be the critical sources of these contaminants (Jiang *et al.*, 2019).

4.7 Correlation Studies of Selected Rivers

4.7.1 Correlation between Microplastics Abundance and Soil Texture

Since microplastics were observed to be more concentrated in river sediment as compared to in river water, correlation studies were conducted to understand the relationship of these microplastics between different soil texture of the sediments.

Based on the results, no correlation (0.164) with $R^2 = 0.004$ was found between the abundance of microplastics and clay soil texture in rivers of Peninsular Malaysia. A similar finding was explored in previous studies such as in the Changjiang Estuary, China (Peng *et al.*, 2017) and the Singapore's coastal mangrove ecosystems (Nor & Obbard, 2014), which showed no significant relationship between the abundance of microplastics and the clay soil texture.

As for correlation between the abundance of microplastics and soil texture of silt in the selected rivers, no correlation (-0.078) with $R^2 = 0.006$ was also observed. This was in accordance with a study conducted in Tamar Estuary, UK (Browne *et al.*, 2010) where the fine grain size distribution showed no significant relationship with microplastic distribution.

Furthermore, the abundance of microplastics also showed no significant relationship with sand soil texture of the rivers (0.035) with $R^2 = 0.001$. This may be attributed by the high probability of remobilization of microplastic particles in loose sandy river sediment once deposited is favored, due to their small size. This was agreeable to a study in the Warnow estuarine sediments, Germany (Enders *et al.*, 2019) and in addition to that, the study found significant correlation between high-density polymer size fractions (\geq 500 µm) and sediment grain size.

4.7.2 Correlation between Microplastics Abundance and POPs Concentration

Since the only type of POPs detected in the present study was the phthalic acid esters (PAEs), the correlation study between the abundance of microplastics and POPs was calculated based on the concentrations of PAEs detected. However, no correlation was observed between the abundance of microplastics and POPs concentration in both river sediment ($R^2 = 0.052$) and river water ($R^2 = 0.024$) of Peninsular Malaysia.

This is in contrast with findings documented in previous studies, such as in rivers of the southern Jiangsu Province, China (Wang *et al.*, 2016), South American estuaries (Barletta *et al.*, 2019), and in the Xiamen coastal areas (Tang *et al.*, 2018). However, the absence of relationship detected in the present study between microplastics abundance and POPs concentration could be reasoned by the influence of external factors such as from water currents, temperature and wind. This is supported by Wang *et al.* (2018) in which environmental factors may control the extent of microplastics and POPs pollutions.

4.8 General Discussion

Microplastics are emerging anthropogenic contaminants, yet their accumulation in the freshwater environment has been receiving less attention, as compared to that in the marine environment. The investigation on the extent of microplastics pollution in the present study revealed that microplastics were present in all river sediment and river water samples, with an astonishing amount of 263.34 ± 28.89 particles/kg in river sediment, and 0.0358 ± 0.0033 particles/m³ in river water, collected from six rivers in Peninsular Malaysia.

Of that amount, the highest incidence of microplastics abundance in river sediment was discovered in Sepetang River with average abundance of 101.39 ± 54.69 particles/kg, followed by Ayer Masin River, with 42.92 ± 20.19 particles/kg. The other four rivers

reported lower microplastics abundance which varied from 22.64 ± 12.21 particles/kg to 32.36 ± 14.03 particles/kg. On the other hand, Ayer Masin River was revealed to hold the greatest microplastics abundance in river water, of average abundance 0.0101 ± 0.0052 particles/m³, followed by Semerak and Sepetang rivers, with an average abundance of 0.0097 ± 0.0050 particles/m³ and 0.0062 ± 0.0022 particles/m³, respectively.

The other three rivers reported comparable abundances which ranged from 0.0028 ± 0.0014 particles/m³ to 0.0038 ± 0.0015 particles/m³. As for the characteristics, most of the extracted microplastics were films and white in colour, with a dominant size fraction of 1.0 to 5.0 mm in river sediment, and 0.1 to 0.5 mm in river water.

There is an urgent need to look into POPs accumulation as their concentrations in global waterways continues to grow and that rivers transported significant concentrations of these contaminants. Additionally, microplastics are known to sorb these compounds from the surrounding environment, which may further act as carriers or vectors to transport these contaminants to biota.

This study revealed that POPs accumulation in all six rivers reached an astounding amount of 1889.26 \pm 229.10 ppm in river sediment, and 582.17 \pm 50.89 ppm in river water. Four types of POPs were discovered and they were generally grouped in Phthalate esters (PAEs), namely MEHP, BDP, DEHS, and DIOP. DIOP was the dominant POPs identified, with the highest concentration in river sediment that was observed in Sedili Besar River of 677.49 ppm, while the highest concentration in river water was recorded in Cherating River of 153.41 ppm. A further point to highlight was that the highest concentration of POPs reported in the sediment was approximately four times greater in magnitude as compared to the highest concentration reported in the river water, highly attributed to the hydrophobicity of POPs where the particles tend to accumulate in the sediments. Generally, microplastics and POPs were prevalent in rivers with hotspots of anthropogenic activities. Sepetang River which flows through some extensive industries including manufacturers of plastic products, as well as through extensive fishing and tourism activities, indisputably recorded the highest microplastics abundance in river sediment. Additionally, intense fishing and tourism activities also contribute to greater extent of microplastics pollution in the other rivers.

It should also be emphasized that despite of having moderate anthropogenic activities, as observed in Sedili Besar and Semerak rivers, these rivers however showed great abundance of microplastics and POPs concentrations, highly linked to the presence of aquaculture activities. Such observation can be due to the fact that plastics are widely used in aquaculture, as the species are grown on plastic polypropylene lines while polyethylene is extensively used as floating rigs and ropes. The impact is intensified by constant exposure of rivers to this activity.

In general, the variation in microplastics concentration among sampling sites may have been a result of differences in anthropogenic impacts, point sources of microplastics input like improper management of MSW, as well as, the influence from natural factors such as currents and winds. Correlation study revealed no relationship on the abundance of microplastics and POPs concentration in both river sediment ($R^2 = 0.052$) and river water ($R^2 = 0.024$) of Peninsular Malaysia.

4.9 Limitation of Study

The present study had several limitations. As far as the microplastics detection is concerned, it is vital to accept that there is an intrinsic instrumental size limitation associated with the detection and quantification of particles by visual inspection using a microscope. Apart from that, organic matter is mentioned in some studies as a nuisance for observing and counting of microplastics. Hence, FTIR and Raman are often used to validate the detected microplastics by identifying their compositions (Filella, 2015). In the present study, without the use of the aforementioned analyses makes it challenging to identify and validate the extracted particles.

4.10 Recommendations

The recommendations for future study of microplastics and POPs pollutions are as follows:

1. As the present study revealed the significance of fisheries notably aquaculture in the generation of microplastics and POPs, studies should be established to investigate the concentration of these contaminants in culture organisms.

2. Since PAEs are the important components that make up plastics and is the only type of POPs discovered in the present study, a detailed research should be conducted to study the concentration of PAEs throughout the entire life cycle of plastics (i.e. the production, application and removal phases). This is to better investigate the contribution of plastics in the generation of POPs.

CHAPTER 5: CONCLUSION

This study reveals the microplastics and POPs pollutions in sediments and waters of selected rivers in Peninsular Malaysia, alongside the possible anthropogenic sources associated with their abundances. Sepetang River, Cherating River, and Ayer Masin River substantially demonstrated greater number of anthropogenic activities, characterized as the hotspots of anthropogenic activities, followed by Sedili Besar River, Serkam River and Semerak River, which were revealed to have moderate anthropogenic activities.

Sepetang River was revealed to have the highest number of anthropogenic activities mainly industries, fishing and tourism activities. Tourism activities were observed to be the main anthropogenic activities in Cherating River, while Ayer Masin River and Sedili Besar River were classified to be profoundly affected by fishing activities. Semerak River, characterized by having the least number of anthropogenic activities, was mainly dominated by agriculture and aquaculture activities. It is noteworthy that this particular river is considered to share a moderate anthropogenic activity even though it holds the lowest number. Additionally, the presence of human settlements and eateries, as well as, open dumping spotted in most of the selected rivers may fairly contribute to the generation of microplastics and POPs.

Results demonstrate that microplastics and POPs are abundant and are widely distributed across the selected rivers. Average abundance of microplastics ranged from 32.36 ± 14.03 particles/kg to 101.39 ± 54.69 particles/kg in river sediment, with average microplastics abundance ranging from 0.0038 ± 0.0015 particles/m³ to 0.0101 ± 0.0052 particles/m³ in river water. The highest abundance of microplastics in river sediment was discovered in Sepetang River, while Ayer Masin River held the greatest number of microplastics in river water. In terms of the morphology, films and white-coloured were the predominant microplastics in both river sediment and river water of the selected

rivers, with a dominant size fraction of 1.0 to 5.0 mm in river sediment, and 0.1 to 0.5 mm in river water.

Four types of POPs were discovered and they were generally grouped in Phthalate esters (PAEs), namely Benzenedicarboxylic acid, mono(2-ethylhexyl) ester (MEHP), 1,2-Benzenedicarboxylic acid, butyl decyl ester (BDP), Decanedioic acid, bis(2ethylhexyl) ester (DEHS), and 1,2-Benzenedicarboxylic acid, diisooctyl ester (DIOP). Overall, DIOP was the dominant POPs identified, with the highest concentration in river sediment was observed in Sedili Besar River of 677.49 ppm, while the highest concentration in river water was recorded in Cherating River of 153.41 ppm.

Furthermore, rivers that were identified to be the hotspots of anthropogenic activities were observed to hold greater abundance of microplastics and POPs concentrations. Fisheries, notably aquaculture activities were measured to be among the critical sources of these contaminants, as evidenced in Sedili Besar, Cherating, and Semerak rivers. However, the present study reveals no correlation between the abundance of microplastics and POPs concentration in both river sediment ($R^2 = 0.052$) and river water ($R^2 = 0.024$) of Peninsular Malaysia.

Overall, this study provides baseline data for the monitoring of microplastics and POPs in selected rivers of Peninsular Malaysia which over time, serves as a foundation for understanding the fate and hazards associated with these contaminants. It is of vital importance that the regulatory authorities should implement and enforce appropriate strategies to monitor, regulate, and protect the rivers, in safeguarding the overall environment.

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