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CHARACTERIZATION AND MODELLING OF COLLOIDAL PARTICLES TRANSPORT IN RELATION TO BISPHENOL A IN BENTONG RIVER, MALAYSIA

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By

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

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Bisphenol A is considered one of the most prominent endocrine disrupting chemicals worldwide. Upon entering aquatic environments, BPA can adsorb onto solids or colloidal particles which play a significant role in determining its behaviour, distribution and fate due to their unique characteristics, and may inhibit its degradation in water and aid in its transport to distant places posing a severe threat to the ecosystem. This issue was highlighted in Bentong River, which is exposed to sewerage discharge and is a main water supply for the Semantan water intake, a part of the Pahang-Selangor Raw Water Transfer (PSRWT) project that provides millions of people with potable water across the two states, rendering this river a critical importance for public safety. The emphasis was directed toward the possibility of colloidal-bound BPA reaching the water intake downstream of Bentong River. Analytical work consisted of identifying particle size distribution in water while BPA analysis involved isolation from water samples through solid phase extraction followed by analysis using LC-MS/MS. Experimental work involved illustrating the interaction and relationship between BPA and colloidal particles in terms of recovery and competitive sorption. The final step was incorporating the obtained figures and results into an environmental model (WASP) to simulate the fate and transport of colloidal-bound BPA in Bentong River. Results showed that BPA levels in Bentong River varied between 1.13-5.52 ng L^{-1} in the soluble phase while the highest BPA concentration in the colloidal phase was 2.06 ng L^{-1}. Experimentally, BPA recovery rate declined by 17% with increasing colloidal organic concentration, and BPA’s extraction via SPME was hampered by inhibition interactions with colloidal particles causing an apparent decrease of 16% in recovery rate. Modelling results demonstrated the significance of spatial detail and highlighted the effects of colloidal particles’ concentration and density on BPA’s removal from the water column. All scenarios showed that after 7.5-10 km mark BPA’s concentration started to reach a steady state with very low

concentrations which indicated that a downstream transport of colloidal-bound BPA was less likely due to minute BPA levels. The various model scenarios implied that particles with low densities, flowing in aqueous conditions that generate low attachment efficiency milieus and with low colloids, while SPM concentrations have a higher tendency to stay suspended within the water column, and consequently have higher propensity to get transported or relocated to farther distances away from the emission point. Even though a downstream transport of colloidal-bound BPA was less likely due to minute BPA levels as proven in this study, the possibility still exists especially if these levels were to increase later on.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

PENCIRIAN DAN PEMODELAN PERGERAKAN ZARAH BAHAN KOLOID MELALUI HUBUNGAN DENGAN BISPHENOL DI SUNGAI BENTONG MALAYSIA

Oleh

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Bisphenol A dianggap sebagai salah satu bahan kimia gangguan endokrin yang paling terkenal di seluruh dunia. Setelah memasuki persekitaran akuatik, BPA dapat dijerap kepada pepejal atau zarah koloid yang berperanan penting dalam menentukan tingkah laku, taburan dan nasibnya kerana ciri-ciri uniknya, terhalang daripada didegradasi dalam air serta membantu pengangkutannya ke tempat yang lebih jauh, menimbulkan ancaman terhadap ekosistem. Isu ini diketengahkan di Sungai Bentong, yang terdedah kepada pembuangan air pembetungan dan merupakan sumber bekalan air utama untuk pengambilan air Semantan, iaitu sebahagian daripada projek Pemindahan Air Mentah Pahang-Selangor (PSRWT) untuk bekalan kepada berjuta penduduk di kedua-dua negeri, menjadikan sungai ini sangat penting untuk keselamatan orang ramai. Penekanan kajian adalah bagi analisis kemungkinan BPA terikat-koloid tiba di titik pengambilan air di hilir Sungai Bentong. Pengenalpastian taburan saiz zarah di dalam air dijalankan menerusi kajian analitik sementara analisis BPA melibatkan pengasingan dari sampel air melalui pengekstrakan fasa pepejal diikuti dengan analisis menggunakan LC-MS / MS. Eksperimen yang dijalankan menunjukkan interaksi dan hubungan antara partikel BPA dan koloid dari segi pemulihan dan penjerapan yang kompetitif. Langkah terakhir adalah kemasukan data ke dalam model alam sekitar (WASP) bagi memodelkan nasib dan pengangkutan BPA yang terikat-koloid di Sungai Bentong. Hasil kajian menunjukkan bahawa tahap kepekatan BPA di Sungai Bentong berjulat antara 1.13-5.52 ng L^{-1} pada fasa larut sementara kepekatan BPA tertinggi dalam fasa koloid adalah 2.06 ng L^{-1}. Secara eksperimen, kadar pemulihan BPA menurun sebanyak 17% dengan peningkatan kepekatan organik koloid, dan pengekstrakan BPA melalui SPME terhambat oleh interaksi perencatan dengan zarah koloid menyebabkan penurunan ketara dalam kadar pemulihan 16%. Hasil pemodelan menunjukkan kepentingan perincian ruang dan menyoroti kesan kepekatan dan ketumpatan zarah koloid pada penyingkiran BPA dari

badan air. Semua senario menunjukkan bahawa setelah mencapai jarak 7.5-10 km, kepekatan BPA mula mencapai keadaan stabil dengan kepekatan yang sangat rendah yang menunjukkan bahawa pengangkutan BPA di hilir sungai tiada berkaitan dengan kepekatan BPA yang terlalu kecil. Berbagai senario model menyiratkan bahawa zarah dengan ketumpatan rendah, mengalir dalam keadaan akues akan menghasilkan tahap kecekapan jerapan milius yang rendah, dengan kepekatan koloid rendah, manakala kepekatan pepejal terampai mempunyai kecenderungan yang lebih tinggi untuk tetap terampai dalam jasad air, dan akibatnya memiliki kecenderungan yang lebih tinggi untuk diangkut atau dipindahkan ke jarak yang lebih jauh dari titik pelepasan. Walaupun pengangkutan BPA terikat-koloid tidak dapat dikaitkan dengan kepekatan BPA yang sangat rendah seperti yang dibuktikan dalam kajian ini, kemungkinan masih ada terutama jika tahap ini akan meningkat di kemudian hari.

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Date: 08 July 2021

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LIST OF ABBREVIATIONS

BPA	Bisphenol A
EDCs	Endocrine disrupting compounds
PD	Patch density
ED	Edge density
SHDI	Shannon's diversity index
CONTAG	Contagion
LPI	Largest patch index
COHE	Cohesion index
SHMN	Shape index
AI	Aggregation index
ENNMN	Mean Euclidean nearest neighbour index
SPME	Solid phase micro-extraction
SPE	Solid phase extraction
LC/MS-MS	Liquid chromatography tandem mass spectrometry
CFUF	Cross flow ultrafiltration
PSD	Particle size distribution
DOC	Dissolved organic carbon
TOC	Total organic carbon
COC	Colloidal organic carbon
KMO	Kaiser-Meyer-Olkin test
Kd	Sorption coefficient
Kcol	Colloidal sorption coefficient
Kcoc	Colloidal organic carbon partition coefficient

Kow	Octanol/water partition coefficient
DVLO	Derjaguin, Verway, Landau and Overbeek theory
Kagg	Aggregation rate constant
α	Attachment efficiency
kcoll	Collision rate
μ	Water's absolute viscosity
g	Gravitational acceleration
ρ	Particles' density
r	Particle's radiu
vs	Settling velocity
NSE	Nash-Sutcliffe efficiency coefficient
PBIAS	Percent bias
RMSE	Root Mean Square Error
WASP	Water quality analysis simulation program

CHAPTER 1

INTRODUCTION

1.1 Emerging Pollutants and Endocrine Disrupting Compounds

Water is one of the most indispensable elements of life's existence and continuation on earth. The demand for clean and safe water is constantly on the rise especially today with the massive increase of world population and the growing necessities of the modern world. Water demand management, water resource planning and ensuring minimum water for daily consumption are the most common concerns in urbanized regions around the globe. The management of water resources comes across many obstacles that arise due to the critical importance of water and its many applications. These various applications and practices are regularly in conflict with one another and the fulfilment of one usually impedes the realization of the other. One of the challenging problems that obscures all these applications and management practices is water pollution. Water pollution is a global issue that causes scarcity of useful water all around the world.

For many decades, heavy metals were the topic of interest for most researchers in the aquatic environment. However, the focus has somewhat shifted toward other types of substances that have had less awareness about them and had emerged with some environmental problems, these substances are commonly known as emerging pollutants. Emerging pollutants are substances not presently monitored or censored in the environment nor regulated by law, but are believed to bring about severe effects on human health and ecosystems. These pollutants comprise a wide array of compounds that are not unknown necessarily and might actually have been in use for years, but have not been identified till novel detection techniques were employed (Ragav et al., 2013). These compounds are chiefly industrial products that are used every day, such as detergents, cosmetic products, pesticides, plasticizers, etc. One of their distinctive characteristics is persistency in the environment due to their resistance to degradation. Still, some of these compounds may not be as persistent as others, but their incessant introduction poses a possible risk to the environment (Pignotti, 2018).

Emerging pollutants include various compounds that exhibit different features. Therefore, they are often categorized into several classes according to their characteristics and use. Among these classes are the Endocrine Disrupting Compounds (EDCs), which represent a major concern due to the threat and danger they pose to the endocrine system of organisms. World Health Organization (WHO) defines endocrine disruptors and potential endocrine disruptors as "an exogenous substance of mixture that alters function(s) of the endocrine system and consequently causes adverse effects in an intact organism, or its progeny, or (sub) population" (Damstra et al., 2002). Despite the limited current knowledge regarding the actual effects of these compounds on human health and aquatic species, there is evidence that EDCs can cause harmful effects at extremely low concentrations. Moreover, there have been growing concerns recently regarding the potential harmfulness of these compounds posed on human health and organisms (Ribeiro et al., 2017). These chemicals can disrupt the endocrine system and interfere with its functions in different forms. EDCs have the ability to mimic typical hormone functions by binding with and ultimately triggering endocrine receptors in a certain cell or tissue, which leads to unpredicted endocrine stimulation. By contrast, other EDCs have the ability to bind to certain receptors without activating them which prevents natural hormones from binding with these receptors and results in a lack of endocrine response.

1.2 Bisphenol A (BPA)

Among those emerging pollutants, also considered as one of the most environmentally prevalent chemicals is Bisphenol A (BPA). BPA, one of the endocrine disruptors, is a high manufactured chemical used extensively today in plastics' production and in epoxy resins. BPA is an organic chemical comprised of two phenolic rings linked by a single carbon carrying two methyl groups. Originally identified as a possible synthetic estrogen in the 1930s, it is currently considered a potential endocrine disrupting chemical and is used extensively nowadays in plastics' production (Allard, 2014). This compound is found in everyday consumer goods from water bottles to can linings and is considered a cause for public health concern due to its extensive human exposure (Collica et al., 2018). Due to the growing demand for plastic products, BPA's production has increased steadily in past years, making it one of the most produced volume substances worldwide. In fact, the global demand has risen from 3.9 million metric tonnes in 2006 to 5.5 million metric tonnes in 2011 (Flint et al., 2012), and was estimated at 7.7 million metric tonnes in 2015 (Almeida et al., 2018). Accordingly, the global market for BPA is forecast to reach 10.6 million metric tons by 2022 (Research and Markets, 2016). BPA's release into the environment is attributed to many sources such as disposal of industrial wastewater from industrial production facilities, breakdown and leaching of many domestic products containing BPA and burning of plastics (Clarke and Smith 2011). It may as well be released into the environment along with untreated landfill leachates (Masoner et al., 2014). This compound is often detected in sewage effluent from industrial and urban sources at concentrations varying between 0.23-149 µg L^{-1} (Höhne & Püttmann, 2008; Sanchez-Avila et al., 2009). Research work conducted in Spain and Greece has revealed that only around 68-87% of BPA is removed in conventional sewage treatment processes (Stackelberg et al., 2007; Stasinakis et al., 2008) while the rest ends up in receiving coastal and surface waters posing a probable risk to these receiving waters (Sanchez-Avila et al., 2009). Thus, BPA's investigation in water, particularly when it's linked with human consumption is extremely vital. Rivers in Malaysia supply over 98% of the country's potable water which gets typically treated by conventional water treatment processes. Yet, many of these rivers are still exposed to untreated sewage discharge, industrial waste and municipal effluents (Fulazzaky et al., 2010).

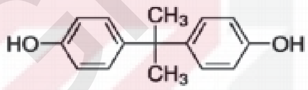
Furthermore, biomonitoring studies indicated that humans are at hazard from internal exposure to BPA, even though this chemical is biotransformed quickly and excreted in urine (Vandenberg et al., 2010). Although BPA has a low affinity for estrogen receptors in comparison with other compounds such as 17β-estradiol, it still causes similar alternations in some cell functions. Moreover, BPA in nanogram ranges was found to be more effective at preventing the release of a vital adipokine that shields human beings from metabolic syndrome (Menale et al., 2008). Chronic health conditions such as cardiovascular disease and diabetes in people may also be associated with higher exposure to BPA compounds (Mercogliano & Santonicola, 2018). Several studies have also reported adverse health complications in wildlife exposed to BPA including changes in reproductive systems and sexual behaviour, but only few studies tackled BPA's effect on human sexual functions (Collica et al., 2018).

The presence of emerging pollutants such as personal care products, pharmaceuticals, pesticides and EDCs in water bodies is a critical issue in many countries. However, the environmental fate of emerging pollutants including BPA and their behaviour are largely unknown. These pollutants primarily enter the aquatic environment, specifically rivers via industrial and domestic wastewater effluent. The conventional treatment plants are ineffective in removing these compounds entirely, resulting in their release undegraded or partially degraded into the aquatic environment (Sarkar et al., 2019). Certain species of these emerging pollutants can undergo partial degradation due to sunlight influence or bioactivity, however, the resulting product can be even more hazardous than the original compound (Naidu et al., 2016). Runoff from agricultural areas sprayed with pesticides or containing biosolids and landfill leachate is considered among the secondary sources of these chemicals (Ragav et al., 2013).

Once introduced into river environments, these pollutants take several paths within the water system due to their physiochemical characteristics (Yan et al., 2015a). BPA biodegrades rather quickly in receiving waters and partially adsorbs onto solid particles. These suspended solid particles associated with BPA may settle down in deposition zones subjected to extra biodegradation under oxic conditions (Staples et al., 2018). However, less degradation is expected to occur in anoxic conditions, though recent research has shown that abiotic processes facilitate mineralization and transformation when oxygen is unavailable, which suggests that BPA is susceptible to degradation under these conditions (Staples et al., 2018). Furthermore, photolysis is another route of degradation for BPA which depends on a specific waveband lengths (Im and Loeffler, 2016).

Despite all the latest efforts in reducing the exposure to BPA and using BPA-free plastics, recent studies state that BPA levels in human beings are actually underestimated (Gerona et al., 2020). Lately, several studies have focused particularly on the occurrence and behaviour of BPA in riverine aquatic environments, which is attributed in part to rivers being often severely impacted by human activities due to the large population densities around rivers and the rapid pollutant transport down the river's hydrologic gradients (Yan et al., 2015a). BPA has been reported in surface water, runoff, urban snow, wastewater and landfill leachate. The ubiquitous occurrence of this pollutant in municipal streams reflects its extensive production and use in society. Ultimately, this compound can be released to aquatic systems either directly or indirectly wherein interactions with other water constituents are to be expected (Ribeiro et al., 2017). BPA can migrate and move about among river compartments and matrices. It can get adsorbed onto sediments or suspended particles, reach groundwater, or accumulate in aquatic organisms and intensify through the food chain until it eventually reaches humans (Ragav et al., 2013).

Table 1.1 : Main characteristics of Bisphenol A

Chemical structure	
Solubility (mg/L)	300
Molecular Weight (g/mol)	228.3
logK_{ow}	3.4
pK_a	9.6

1.3 Pollutants' Transport

The fate and transport of pollutants in river systems are controlled by their reactivity and hydrodynamic transport. Rivers have distinct hydrodynamic characteristics that are different compared to estuaries and lakes. Pollutants' transport in water takes place under dissolved or particulate forms. In surface waters, solid particles introduced into streams can move either in a particulate state downstream (bed-load transport) and get deposited farther downstream, or move in a suspended form within the water column. This transport depends on the particle's size, shape and density, as well as flow rate, velocity, and turbulence (Durães et al., 2018). The speed and turbulence of currents enable transportation of particles with different sizes. When the river flow or riverbed gradient diminishes, particles tend to settle down and this often occurs when river flow reaches reservoirs (Auel et al., 2017). Moreover, the dissolved transport in water is also significant since it is related with more available forms of contaminants with substantial environmental concern. This relates mainly to leaching or detachment processes, by which contaminants are released from the solid phase into the aqueous phase under the effect of desorption and dissolution of contaminants from their support-phases. This relies on numerous factors including pH, redox conditions, temperature, biotic action, partition constant, etc. (Ji, 2012).

1.4 Pollutants' Transport Mechanisms

1.4.1 Advection, Dispersion, and Diffusion

Dissolved contaminants may be transmitted by advection, dispersion, and diffusion in water. Advection involves movement along the direction of flow, which is related to the fluid's average velocity. Darcy's Law which links the hydraulic gradient with the bulk properties of the materials (porosity) explains this form of transport. Advection is the easiest way to describe the transport of contaminants, which can be viewed as a chemical transport induced by a hydraulic gradient. On the other hand, pollutants can be spread and dispersed during flow movement due to changes in fluid velocity. This spreading is the result of dispersion, which is comprised of mechanical dispersion when water velocity is high, and diffusion when the water velocity is low. Diffusion is a process where contaminants move under the effect of kinetic energy in the direction of the concentration gradient (Durães et al., 2018).

1.4.2 Colloid-Facilitated Transport

This term describes a transport process by which colloidal particles serve as a transport vector for various pollutants in surface water and in underground water circulating in fissured rocks (Bedrikovetsky et al., 2011). This relates to the movement of small size particles (<1μm) to which pollutants are attached to by sorption or ionic exchange. Such particles are distributed in the aqueous phase and may act as mobile sorbents, thus allowing pollutants to disperse easily and move to other locations or environmental compartments away from the source of pollution (Durães et al., 2018). Colloids may be of organic or inorganic origin, and they do not tend to settle out of suspensions due to their small size, succumbing to the Brownian motion effect and minor currents in the bulk solutions (Pédrot et al. 2008). The fate of pollutants and their impact on the environment are strongly dependant on the nature and behaviour of mobile colloids, whose fate is affected by their size and stability and the size and connectivity of its pores. Following mobilization, the lifetime of colloidal particles is mostly determined by convective transport, particle deposition or resuspension and aggregation behaviour, which may take place simultaneously (Ilina et al. 2008). However, the significance of each one of these processes highly depends on the chemical and physical characteristics of the aquatic system.

Experimental and analytical evidence illustrated that many pollutants in water are not only transported in a dissolved form, but also sorbed to mobile colloidal particles. That has led to numerous studies centred around colloidal facilitated transport of many pollutants such as heavy metals, pesticides, pharmaceuticals, radionuclides and hormones (Gavrilescu, 2014). This demonstrates the importance of this topic since failure to account for colloid-facilitated solute transport can severely underestimate the transport potential and risk assessment of these contaminants (Bradford et al. 2011). Pollutants can move faster and farther in the presence of mobile colloids than they do in their absence, and these colloids can inhibit the degradation or availability of pollutants, thereby contributing to their transport downstream onto distant sites and other environmental compartments away from their original source (Gavrilescu, 2014).

1.5 Colloids in Aquatic Environments

1.5.1 Colloidal Particles' Impact in Aquatic Environments

Generally, all particles affect natural water by forming associations with pollutants and affecting the transport process of these pollutants throughout the waterbody. Colloidal particles are available abundantly in aquatic environments and they have different compositions, shapes, structure, coatings and surface chemistry (Wilkinson and Lead, 2007). As a result of their small sizes, these particles typically possess a long residence time and considerably influence the transparency of water. Colloids originate commonly from natural processes, for instance weathering, volcanic explosions and bacterial activity, as well as anthropogenic practices such as effluent from wastewater treatment plants or direct sewage discharge and livestock wastewater from farms and ranches (Hartland et al., 2013). Their high reactivity, specific surface area and high organic content render them a strong adsorption capacity to countless substances including emerging pollutants, and consequently, affect their behaviour, chemical composition and environmental fate (Gibson et al., 2009). Ribeiro et al. (2017) stated that interactions between these pollutants and colloidal particles are to be expected. Accordingly, the toxicity, bioaccumulation, concentration and speciation of EDCs are ultimately controlled and co-regulated by the influence of colloids. Besides, persistency of colloids in river water affects these aquatic ecosystems and may as well lead to microorganism toxicity (Pokhrel et al., 2013). The major impact on the transport and fate of pollutants and also on ecosystem balance has garnered increased consideration lately regarding aquatic colloids.

1.5.2 Colloidal Particles' Association with Pollutants

Colloidal particles are considered a suitable medium for pollutants' adsorption, having a high surface area compared to all the natural components in water (below 10 m^2/g). Thus, these particles can be associated with different types of pollutants. This robust sorptive capacity of colloidal particles has been well established and it relies mainly on their size, concentration, solution pH and reaction time (Yan et al., 2016). Inorganic colloidal particles are particularly effective in pollutant adsorption due to their high surface reactant area. This association can take place through different mechanisms such as ion exchange or surface interactions. Moreover, colloidal particles associated with pollutants can undergo different transformation processes which can be stimulated by sunlight and presence of oxides or other compounds (Yan et al., 2015a). Several studies have illustrated that the transformation behaviour of pollutants is in part reliant on their adsorption tendencies and river hydrodynamics (Wang et al., 2016; Liu et al., 2017). Hydrophobicity was also stated as a significant factor that influences the sorption of phenolic xenoestrogens, such as BPA, onto particulate and dissolved organic carbon (Gong et al., 2016). Recently, studies have started to account for EDCs' occurrence in other phases such as the colloidal phase, which is formed by the interaction of organic or inorganic particles with EDCs. Huang et al. (2019a) stated that colloidal-bound EDCs were notable and varied between 7.80 & 44.3% in municipal secondary effluents. Zhou et al. (2007b) reported that EDCs' adsorption onto aquatic colloids of diverse origins varied considerably as well. Furthermore, it has been suggested that EDCs' sorption onto colloidal humic acid particles has a major significance in aquatic environments as it reduces the bioavailable concentration and toxicity to aquatic creatures (Kim et al., 2016).

In addition, most studies look at the presence and distribution of pollutants in aquatic environments with a specific focus on the so-called "dissolved phase", which is defined though arbitrarily, but conveniently as any substances that pass through a membrane filter with pores from 0.22 µm to 0.70 µm (Duan et al. 2013). However, it has become well known that small colloids, which are abundant in aquatic environments, actually pass through these membrane filters and are thus often neglected for their role in pollutant behaviour and distribution. Therefore, this conventional dissolved phase can be divided further into a spectrum of colloidal particles, known as the "colloidal phase", as well as a further filtrate that is termed as the "soluble phase" (Lead & Wilkinson, 2009). Prior studies though limited proposed that colloidal particles can be a significant sink for certain pharmaceuticals (Yang et al., 2011), and lately, they have attracted more consideration due to their high pollutant reactivity and mobility implications (Graham et al., 2014). For instance, polycyclic aromatic hydrocarbons (PAHs) were found to be predominantly attached to smaller colloids rather than larger particulate matter in stormwaters and landfill leachates (Kalmykova et al., 2013). Hofmann and von der Kammer (2009) reported that the engineered carbonaceous nanoparticles could increase pollutants' transport relying mainly on their size, sorption kinetics and residence time. Colloidal particles have different origins, chemical compositions, atomic structures and physicochemical properties. These characteristics highlight and underline their significance and impact in pollutants' adsorption transport (Lead et al., 2018).

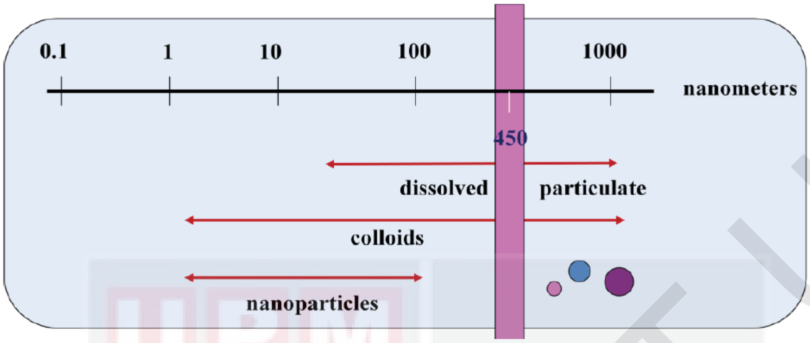


Figure 1.1 : Particle size domains of component phases in waters (Ketkoom, 2011)

1.5.3 Colloidal Particles' Transport in Water

Currently, several researchers view colloidal particles as a principal agent and transport carrier of organic pollutants, heavy metal and radioisotopes in water bodies. As mentioned above, the high surface area and low settling velocity of colloidal particles and the state of near continuous suspension in water columns make colloidal particles very efficient mediums that adsorb dissolved pollutants readily (Miller & Karathanasis, 2014). Ideally, pollutants can be sorbed onto an immobile surface and remain bound to this medium, thus posing little danger in water. However, with the presence of colloidal particles these pollutants can get attached to these particles which in turn increase their mobility. Besides, the partition of EDCs onto colloidal particles has been shown to be relatively independent from the physio-chemical characteristics of these compounds, especially their octanol water partition coefficient (K_{ow}) (Liu et al., 2005). Also, the sorption of polar EDCs by colloidal particles might be credited to the physio-chemical characteristics of these colloids, and BPA is considered a moderate hydrophobic chemical that attaches readily to colloidal particles and dissolved organic matter as well (Petrie et al., 2019). The binding interactions between colloidal particles and BPA considerably affect the transformation and migration of BPA in aquatic environments (Bhatnagar and Anastopoulos, 2017).

Field studies and lap tests have showed that colloidal particles have the ability to carry pollutants to long distances and increase their distribution in aquatic systems (Kalmykova et al., 2013). Colloidal facilitated transport, a well-known concept especially in soil porous medias and land fractures, can increase the extent and range of pathogens and pollutants travelling through waterways. The presence of metals associated with colloidal particles has been widely reported, these reports emphasised the role of colloidal particles as carriers and their influence in enhancing the mobility of contaminants. One of the decisive factors in colloidal facilitated transport of pollutants in water is that colloids must be present amply in order to adsorb a substantial amount of pollutants, a fact that is underestimated in laboratory tests (Ketkoom, 2011).

Temperature, pressure, pH and ionic strength are among the key factors dictating colloidal particles' mobility and distribution. The movement of colloidal particles increases with high flow rates and with increasing pH values (Casanova et al., 2018). On the other hand, ionic strength has an inverse relation with colloids' mobility, since solutions with high ionic strength enhance aggregation kinetics and hinder colloidal mobility (Gan et al., 2019). Studies on colloidal particles' mobility and facilitated transport have focused on particles and contaminants' movement through soil zones and their effect on groundwater.

To this date, there are insufficient studies that display and illustrate the role of colloids in natural waters. Therefore, characterizing colloidal particles and understanding their behaviour in water is essential in trying to predict their movement and influence on pollutants' reactions and transport in water. This work includes an investigation on the relationship between colloidal particles and BPA, as a predominant emerging pollutant and endocrine disruptor. The effects of colloidal particles' behaviour and characteristics (size distribution, concentration, zeta-potential & molecular weight) on BPA's fate and behaviour in river water are analysed and illustrated both analytically and experimentally, and are further described and depicted using computational modelling. The aim is to gain a better insight on the role of natural and man-made colloidal particles in pollutant transport in the aquatic environment.

1.6 River and Water Quality Modelling

Mathematical models are governed by conservation laws of mass and momentum. These models are used to illustrate the spatial and temporal variation of different physical processes in the hydrologic system along with extra information regarding land use and cover, climate and hydrology. Modelling the hydrologic response to natural or anthropogenic induced changes offers a unique understanding of these physical processes such as stream flow, fluid and pollutant transport in the surface and subsurface and the interaction between the atmosphere and land surface (Yu, 2015). Models are robust instruments for illustrating the behaviour of pollutants in complex natural systems, and thus are indispensable for scientific and regulatory intentions. Models can offer a framework that is essential for a clear and well-defined scientific understanding of environmental fate and behaviour of pollutants in water. The fate and transport of pollutants in rivers are very intricate processes that involve physical transport along with chemical and biological kinetics. Thus, it is vital that mathematical models employed to estimate and elucidate pollutants' fate and related processes are flexible, reliable and accurate (Ji, 2012). These fate and behaviour models can add value to scientific efforts by exploring the relative effects of the processes contributing to pollutants and particles' fate and behaviour in complex ecosystems, thereby identifying dominant processes.

Additionally, a complete environmental fate assessment of any pollutant or variable involves four elements. The first element is the environmental fate model that outlines the environmental system undertaken and the processes occurring within this system that govern the fate of its components and pollutants. The second factor is pollutant's physicochemical properties, which include the rate of degradation and partition coefficients. The third item involves details about the emission levels of the pollutant in various regions and at different stages. The latter two elements constitute entry variables to the environmental model, and the model results are derived from the combination of these three elements. Lastly, the fourth element of the overall assessment is field data. This element is independent from the overall process and it shows levels of the pollutant measured in the environment. Model results should be compared to the field data whenever it is available, and any kind of disagreement needs to be analysed (Scheringer et al., 2014). The term "fate" contains three principal types of processes: (i) transformation and degradation, (ii) circulation or exchange of pollutants between various environmental compartments inside one spatial domain or region of the model, and (iii) transport, either from one section of the model system to an adjacent one or out of the model system with moving water, sediment, soil or air (Scheringer et al., 2014).

In order to illustrate pollutants' environmental fate, it is necessary to gain insight regarding what type of transformation and transportation processes these compounds undergo, what are the time scales for these processes, what happens when certain environmental conditions change and how will these different processes affect the mass balance of these pollutants in a connected environmental system such as water. Environmental fate models are useful and practical tools that offer solutions and explanations to these inquiries. They make it possible to predict the levels of pollutants in different environmental compartments, compare the effects of competing processes, and identify the processes and parameters that require more detailed investigation via laboratory experiments.

1.7 Environmental Fate and Modelling of Colloidal Particles and BPA

Generally, when organic pollutants enter aquatic environments, their adsorption and attachment onto suspended particles undoubtedly come to be one of the most critical factors determining their behaviour, transport and fate. Several studies have demonstrated that the transformation behaviour and transportation of pollutants rely on their adsorption characteristics in the aquatic environment. Therefore, it is important to understand the adsorption mechanism of organic pollutants on natural colloids. Hence, a considerable amount of information has been gathered in the last decade or so regarding the environmental fate of nano chemicals in association with colloidal and nano particles in aquatic systems. However, some difficulties remain unmitigated (Scheringer et al., 2014). Klaine et al. (2008) even stated that colloidal particles are a critical factor that affect the behaviour and fate of nano pollutants in general. Therefore, a better characterization of colloids and how they interact with nano pollutants is essential to further understand their fate in the environment. Moreover, these interactions and relationships must be incorporated into environmental fate models for nano pollutants.

As mentioned earlier, colloidal particles have been shown to play a significant role in determining the behaviour, distribution and fate of BPA in aquatic environments, which is commonly attributed to their unique characteristics such as high surface area, concentrated organic carbon content and their high affinity toward pollutants (Kalmykova et al., 2013). Additionally, BPA's fate in water has been shown to be highly affected by the physicochemical characteristics of colloids such as particle size, zeta-potential and fluorescence intensity (Yan et al., 2015a), thus their colloidal adsorption should significantly affect their reactivity, bioavailability and mobility (Yan et al., 2015b). However, the precise influence of colloids on BPA and their relationships are still inadequately described. Furthermore, recent studies have run into some bottlenecks in exposure assessment due to the immense complexity of nanomaterial fate processes. An aspect of this complexity is the enormous variety of types and forms of nano particles and natural colloids, interacting chemical substances and reactive particle surfaces (Praetorius et al., 2013). Another dimension of uncertainty derives from the environmental transformations of nano chemicals and particles, which are known to take place at unpredictable spatial and temporal scales (Nowack et al., 2012).

In aquatic environments, determining the transformation, fate and transport of emerging pollutants, especially pollutants bound to colloidal and nano particles is a challenging process and depends greatly on a wide spectrum of intrinsic and extrinsic parameters (Sani-Kast et al., 2015; Clavier et al., 2019). Water characteristics such as temperature, pH, ionic composition and water hardness strongly affect the transport mechanisms and chemical reactivity of organic pollutants and nanoparticles (Koelmans et al., 2015; Ellis et al., 2016). Presence of living organisms (Carnal et al., 2015) and suspended particulate matter can also influence these processes a great deal (Praetorius et al., 2014). Often, slight changes in water chemistry can profoundly affect pollutants' characteristics as well as interactions and bindings with other substances surrounding them. Furthermore, these changes impact key processes such as degradation and dissolution of pollutants as well as settling, aggregation and sedimentation of particles, and consequently the distribution and transport of both within river sections (Ellis et al., 2016). The presence of natural organic matter specifically, even in modest concentrations is expected to have an important impact on pollutants' behaviour in water (Praetorius et al., 2014).

Consequently, modelling colloidal facilitated transport of pollutants becomes even more complex. Due to their particular size and nature, colloidal particles' behaviour in riverine systems is investigated using concepts that have been recently used in nanoparticle transport models (Sani-Kast et al., 2015; Quik et al., 2015). Most of environmental colloids and nanoparticles consist of the low molecular weight breakdown products of biological decay (humic matter), numerous fibrillar and mesh-like organic compounds, and minerals generated during the chemical weathering of rocks, mainly the oxides and oxyhydroxides of manganese, iron, aluminium and aluminosilicates. These types of compounds have the effect of generally increasing the size of colloids and nanoparticles via aggregation (Hartland et al., 2013). Both colloidal and nano particles have certain similarities besides the overlap in size range, and the fundamental concepts of surfaces and interfaces are essential for these particles (Wang et al., 2015). Furthermore, the DLVO theory is applicable to both colloidal and nano particles, this theory describes the balance between van der Waals attractions and electrostatic repulsions in a liquid medium (Hornyak & Rao, 2016). Therefore, it is feasible to assume that colloidal particles associated with BPA will aggregate with other colloidal or nanoparticles, or that colloidal aggregates will directly adsorb BPA forming complexes with varying hydrodynamic diameters, as it was implied in several research articles (Baalousha et al., 2011; Wang et al., 2015; Besseling et al., 2017; Lead et al, 2018).

The potential hazards of this association and linkage between emerging pollutants such as BPA with colloidal particles to humans and ecosystems, even in minute concentrations, are highly dependable on the transport pathways of colloids in natural systems, and on the concentration of both components in different environmental compartments. Furthermore, the risk magnitude may as well hinge on whether the particle is freely dispersed or aggregated. The behaviour of colloidal particles in aquatic environments is controlled by their stability and aggregation tendencies, which in turn are significantly reliant on the surrounding environmental conditions (Xu et al., 2018). Akin to organic pollutants, several parameters have also been stated to have an impact on the stability of colloidal particles, for instance natural organic matter, pH, electrolytes and ionic strength (Philippe & Schaumann, 2014).

Therefore, to have a comprehensive insight on the effects of colloidal particles on emerging pollutants and their concentration in aquatic systems, it is imperative to illustrate their transport and fate in water in order to properly assess any risks or hazards posed by any pollutant associated with these particles. Subsequently, new environmental fate models have emerged as a valuable tool in examining the relationships between colloidal particles and emerging pollutants such as BPA.

One of the most popular environmental programs is The Water Quality Analysis Simulation Program (WASP). WASP is a dynamic, mass-balance, transport and fate model that enables users to simulate pollutant concentration and movement in surface water and sediment (Knightes et al., 2019). WASP allows for time-varying processes of advection, dispersion, point and diffuse mass loading, boundary conditions and exchange. Moreover, it can be linked to other hydrodynamic models and import data from different databases. It is one of the most widely used water quality models in the US and throughout the world.

WASP includes two specific modules: The Eutrophication Module and The Toxicant Module. WASP developers in the past years have concentrated on the eutrophication module (Wool et al., 2013), and many modelling studies have focused on conventional pollutants and eutrophication (Sadeghian et al., 2018; Nguyen et al., 2018; Gargallo et al., 2018). The Toxicant module, on the other hand, received less work over the years with limited development since the 1990s. Most recently, WASP8 (version 8.2) was released (April 2, 2019), which included an overhaul of WASP's framework and made evident restructures and upgrades on the toxicant module. One of the clear motives for this effort was the explicit interest in emerging pollutants and their potential exposure concentrations, particularly nanomaterials, once released into the environment (Knightes et al., 2019). Subsequently, WASP8 Advanced Toxicant module received a total makeover that redesigned its state variables in distinctive arrays to allow simulations of any number of solid particles, chemical solutes and nano particles. Furthermore, Additional functionality options have been added to incorporate nano and colloidal particles' specific processes, distinctively particle attachment kinetics and aggregation. This new version of the Advanced Toxicant Module has been applied in limited research works. Bouchard et al. (2017) estimated the fate and transport of multi-walled carbon nanotubes in Brier Creek, Georgia (US), while Camacho et al. (2018) employed it as part of an uncertainty and risk analysis for a margin of safety of a nutrient TMDL in Sawgrass Lake, Florida (US). WASP's Advanced Toxicant Module resembles other novel environmental programs employed in this field of study (NanoDUFLOW & SimpleBox4Nano) in having similar elements, functions and flexibility.

Regarding this research, after estimating and characterizing colloidal particles and BPA concentrations in water samples and illustrating their interactions and related variables, WASP will be applied to simulate these interactions and relationships between BPA and colloidal particles in water as well as their transport and fate along the river course. The purpose is to make a prediction on where these colloidal-bound BPA molecules will end up in the river and their level in different sections, since several processes influence the form in which they exist and how they are transported in surface water.

1.8 Landscape Patterns & Land Use Impact on River Water

Ecological integrity of rivers and streams has been shown to be fundamentally connected with the surrounding landscapes and land applications (Staponites et al., 2019). River water composition is significantly influenced by numerous natural & anthropogenic activities and with the growth of world population and the incessant urbanization, landscape alterations have had an evident impact in recent times. Landscape ecologists define spatial patterns as the structure, arrangement and placement of objects within any particular landscape. This can range from river banks, to patches of forestry, to man-made settlement landscapes such as cities and towns. Each one of these environmental settings are organized and planned in a discernible pattern that can offer a lot of data pertaining to its history, composition, and ecosystem (Turner & Gardner, 2015). Meanwhile, land use is regarded as one of the components of landscape that has a significant role in generating and spreading pollution. Land use is defined in general terms as a series of practices on land, implemented by individuals in order to obtain benefits from its amassed resources. Moreover, Land use is related to the service that the land provides, it is not concerned with the surface cover on the ground. In short, land use indicates how people are managing the terrain and utilizing the land (Giri & Qiu, 2016).

Land use patterns have a significant impact on aquatic environments and river water quality (Bu et al., 2014), and they influence water systems through non-point pollutants which pose a significant risk to water quality, particularly in residential areas (Jia et al., 2013). Furthermore, natural landscapes have long been fragmented and transformed into impervious areas, which often lead to a surge in surface runoff. This continuous transformation alters the hydrological processes and makes it easier for pollutants to reach river networks, leading to additional strain and degradation in water quality (Barbosa et al., 2012). Typically, land use types dictate the sort of pollutants generated on land and carried via runoff into waterbodies. Water quality variables have been strongly related with the configuration and proportion of land uses inside multiple areas. In fact, US EPA (2015) mentioned that the significant increase in urban population since establishing the Clean Water Act has made stormwater contaminants become the chief reason of impairment in Colorado's urban surface waters. Furthermore, recent studies offered a strong evidence that urbanization generates excessive amounts of nutrients, sediments and metals that affect ecological characteristics and stability of surface waters (Son et al., 2015; Park & Park, 2015).

Similarly, the relationship between landscape patterns and water quality deterioration is considered to be a pattern-process relationship, in which energy, nutrients and material in a landscape are influenced by permeability, connectivity, aggregation and configuration (Mitchell et al., 2013). Several studies have highlighted the impact of landscape patterns on water chemistry and quality variation, and particularly the significant bearing of landscape features on stream health (Beckert et al., 2011; Zhang et al., 2013; Zhang et al., 2019a). Griffith (2002) even mentioned that the association between landscapes and water quality is area specific and non-stationary and above all complex. The spatial structure of landscapes in particular is essential for assessing the connection between landscapes and water quality at different scales (Zhang et al., 2019a). Furthermore, landscape composition and configuration may well be a key factor that have some bearing on hydrological processes, chemical cycles, energy flows and natural habitats (Mitchell et al., 2013). Hence, it has become extremely imperative to identify and illustrate the impacts stemming from the surrounding environment on water quality parameters with specific emphasis on land use patterns and landscape metrics in order to implement sustainable water management strategies.

Landscape metrics are defined as quantitative indices that illustrate the spatial and structural aspects of landscapes based on information gathered from remotely sensed images, maps and GIS coverages. Landscape elements, as entities, are defined as discrete units or patches. Landscape patterns are described or interpreted via metrics developed to quantify a patch in terms of size, isolation and shape, and also regarding its makeup and mosaic characteristics such as patch richness, connectivity, diversity and contagion. Typically, the aim of landscape analysis is to link measures and configuration of landscape structure to specific effects on ecological processes, instead of treating these numerical descriptions of spatial pattern as independent estimations (Kupfer, 2012). Lately, studies have dedicated extra focus on spatial configuration of land uses and on different landscape metrics in an attempt to further understand the association and interconnection between them and water characteristics in watersheds. Incorporating advanced statistical analysis and spatial analysis techniques have made a notable progress in these studies (Bu et al., 2014).

Rivers in Malaysia are extremely turbid. Deforestation and land use changes, particularly the conversion of forests into oil palm plantations, have adversely impacted the water quality in rivers and contributed to the high volume of suspended solids in rivers. Also, the distinct rainy season in Malaysia usually generates considerable runoff volumes that travel across different land surfaces and carry various substances into waterbodies. Therefore, it is essential to examine the relationships of land use and landscape with water quality in any river basin from potential point and non-point pollution sources. Although it is still challenging to relate the numerous water quality variables to landscapes and land uses in urbanized areas with high heterogeneity.

In addition to illustrating colloidal-bound BPA fate and transport in Bentong River, we tried to illustrate the significance of spatial land uses and landscape patterns on water quality. Normally, it is somewhat difficult to assert and clarify these relationships in rivers, due to the continuous input from upstream. Nevertheless, we operated on the basic concept that surface settings and spatial composition will categorically change or have some bearing on water quality in adjacent water systems.

1.9 Problem Statement

Endocrine disrupting chemicals (EDCs) have received growing attention recently due to the endocrine disrupting effects on humans, fish and other aquatic vertebrate species, even at concentrations as low as the ng L-1 level. Bisphenol A, one of the most prominent EDCs, is widely utilized in industrial production and is found in daily used products. This compound is only partially removed in wastewater treatment plants and ultimately ends up in ambient waters via effluent discharge, runoff and landfill leachate. Upon entering the water system, this compound can be absorbed by aquatic organisms, or adsorb onto solids or colloidal particles which inhibit its degradation in water and may aid in its transport to distant places posing a severe threat to the ecosystem.

The impacts of current land development and effluent discharge from sewerage systems within the Bentong catchment area are a major concern to the waterbody, especially with the Semantan water intake situated downstream of Bentong River. This water intake is being tapped by many people in the Bentong region and around it, which may ultimately expose them to BPA among other chemical compounds and cause some health concerns in the future due to the accumulative characteristic of these compounds. Moreover, with the projected development plan for Bentong area, the pollution load will definitely escalate with population surge. Consequently, the damage is bound to get much worse and emergence of different pollutants will ultimately cause novel problems.

1.10 Objectives of the Study

The overall objective of this study is to gain insight on the potential release of Bisphenol A, its transport and fate in the aquatic environment in association with colloidal particles. The emphasis is directed toward the possibility of these pollutants reaching the Pahang-Selangor Raw Water Transfer (PSRWT) intake downstream of Bentong River.

Therefore, the focus will be on both natural and synthetic colloidal and nano particles and on BPA compounds in Bentong River with the following specific aims:

1. Illustrate the relationship and correlation of water quality variables, land use types and landscape spatial metrics, also identify pollution factors and most polluted sites.
2. Illustrate the interactions of BPA in association with colloidal and nano particles.

3. Quantify BPA's concentration in the colloidal and soluble fractions in Bentong River.

4. Characterize the relevant colloidal and nano particles' properties in Bentong River

5. Simulate the influence of different environmental & hydrological parameters on the colloidal-bound BPA's fate & transport in the river using WASP simulation program.

1.11 Scope and Limitations of the Study

The study will take place in Bentong District, samples will be circumscribed to Bentong River only starting just before it enters the main district and ending after it departs just before it reaches the water intake. The study will be limited to only colloidal particles excluding the particulate suspended solids bed sediments. Moreover, the focus will be mainly on Bisphenol A, a widely spread endocrine disrupting compound, and its association with colloidal particles in Bentong River. This relationship will be illustrated with experimental work as well as simulation processes using an environmental model to demonstrate further the extent of this association and possible risks along the river course. BPA will be measured in both colloidal and soluble phases without expanding to other phases such as the total, particulate or the traditional dissolved phase. Sediment samples were left out of the study which would have given an insight into BPA's accumulation in the river bed. BPA samples were also limited to seven samples instead of the 22 sites planned for other water variables.

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He holds a Bachelor degree in Environmental Science and Technology from the University of Mosul, Iraq, where he also obtained his Master's degree in Environmental Science. His Master's thesis dealt with river water quality and the impact of point and non-point pollution as well as the influence land use and cover.

LIST OF PUBLICATIONS

Shehab, Z.N., Jamil, N.R. and Aris, A.Z., 2020. Modelling the fate and transport of colloidal particles in association with BPA in river water. Journal of Environmental Management, 274, p.111141.

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