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OCCURRENCE, RISK ASSESSMENT AND PUBLIC RISK PERCEPTION OF MULTICLASS ENDOCRINE DISRUPTING COMPOUNDS IN DRINKING WATER SUPPLY SYSTEM

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By

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Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the degree of Doctor of Philosophy

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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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Presence of endocrine disrupting compounds (EDCs) in drinking water concerns the security and sustainability of the supply system. The problem is not only limited to the pollution level of EDCs in raw and treated water, but also exposure of organisms to EDCs especially human via daily drinking water consumption. The trace level in tap water challenges the identification of the multiclass EDCs. With the wide ranging nature and characteristics of the pollutants themselves (if known at all), the analytical methods were made available only to single EDC group and/or its metabolites. The limited EDC monitoring in tap water and evidence of human exposure risk caused the increase of associated risks as they may have been underestimated and even unknown. The present study describes an analytical method based on solid phase extraction followed by liquid chromatography-tandem mass spectrometry (SPE-LC-MS/MS) for analysis of multiclass EDCs (hormones, pharmaceuticals, plasticizers and pesticides) in tap water in a single extraction step. The method was validated with recovery between 85 to 119% for most of the EDCs and method detection limit ranging from 0.01 to 2.56 ng/L. Method precision was achieved with linearity > 0.9 and relative standard deviation less than 15% for the targeted compounds. A total of 14 EDCs i.e., five hormones, seven pharmaceuticals, one plasticizer and one pesticide was detected in Langat River, a drinking water source treated for Malaysian drinking water supply. Chloramphenicol and 4-nonylphenol were below method detection limit in both raw and treated water. Prevalent occurrence of EDCs was observed in Malaysian tap water up to 66.40 ng/L (bisphenol A). Triclosan and 4-octylphenol were only detected in tap water at concentration up to 9.74 and 0.44 ng/L, respectively. Variation also observed in different housing types. For local exposure, human health risk assessment was based on human morphological, drinking water consumption patterns and household practices collected through a survey using a newly developed and verified questionnaire. This study captured the complex dynamic of the public-perceived risks on safe drinking water quality in regards to EDCs and the influencing factors, providing a comprehensive conceptualization of the predictors of environmental risk perception, trust, attitude and knowledge. The public tend to perceive the risks through nonrational processing system that highly subjected to cognitive and affective influences. Also, the perceived EDCs contamination in tap water was found to have no association with measured values. Monitoring and risk assessment are the initial processes in multibarrier approach in drinking water supply system for safe water resources. Fulfilling the gap of risk perception and identification of the role of risk perception through development of risk perception model were useful in formulating the efficient preventive and intervention measures with known target groups and materials. Higher trust towards stakeholders and perceived risk on human exposure to environmental risk, as well as positive attitude towards drinking water quality improvements can increase the public perceived risk on drinking water. The subsequent public improvement in terms of risk behavior development was also essential for effective risk governance and communication, supporting the aforementioned multibarrier approach.

Keywords: Endocrine disrupting compound (EDC); SPE-LC-MS/MS; Tap water; Risk assessment; Risk perception; Drinking water safety

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

KEWUJUDAN, PENILAIAN RISIKO DAN PERSEPSI ORANG AWAM TERHADAP RISIKO KEPELBAGAIAN KELAS SEBATIAN MENGENDALA ENDOKRIN DALAM SISTEM BEKALAN AIR MINUMAN

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Kehadiran sebatian mengendala endokrin (EDC) dalam air minuman membimbangkan keselamatan dan kemampanan sistem bekalan. Masalah ini bukan hanya terhad kepada tahap pencemaran EDC dalam air mentah dan terawat, tetapi juga pendedahan organisma terhadap EDC terutamanya kepada manusia melalui penggunaan air minuman harian. Tahap pencemaran EDC yang rendah dalam air paip memberi cabaran dalam identifikasi kepelbagaian kelas EDC. Dengan sifat dan ciri-ciri pencemar yang pelbagai (jika diketahui), kaedah analitikal hanya disediakan untuk kumpulan tunggal EDC dan/atau metabolitnya sahaja. Pemantauan EDC dalam air paip serta bukti risiko pendedahan manusia yang terhad menyebabkan risiko yang berkaitan mungkin telah dipandang ringan malah tidak diketahui. Kajian ini menerangkan kaedah analisis berdasarkan pengekstrakan fasa pepejal diikuti dengan kromatografi cecair spektrometri jisim (SPE-LC-MS/MS) untuk analisis kepelbagaian kelas EDC (hormon, farmaseutikal, bahan pemplastik dan racun perosak) dalam air paip dengan langkah pengekstrakan tunggal. Kaedah ini telah disahkan dengan peratus perolehan semula antara 85 to 119% bagi kebanyakan EDC dan had pengesanan kaedah antara 0.01 hingga 2.56 ng/L. Ketepatan kaedah dicapai dengan kelinearan > 0.9 dan sisihan piawai relatif kurang daripada 15% untuk sebatian yang disasarkan. Sejumlah 14 EDCs iaitu lima hormon, tujuh farmaseutikal, satu bahan pemplastik dan satu racun perosak telah dikesan di Sungai Langat iaitu sumber air minuman yang dirawat untuk bekalan air minuman di Malaysia. Kloramfenikol dan 4-nonilfenol berada di bawah had pengesanan kaedah dalam air mentah dan terawat. Kehadiran EDC lazim diperhatikan di dalam air paip Malaysia mencapai 66.40 ng/L (bisfenol A). Triclosan dan 4-oktilfenol masing-masing dikesan dalam air paip di kepekatan sehingga 9.74 dan 0.44 ng/L. Variasi pencemaran juga diperhatikan dalam jenis perumahan yang berbeza. Untuk pendedahan tempatan, penilaian risiko kesihatan manusia adalah berdasarkan morfologi manusia, corak penggunaan air minum dan amalan isi rumah yang dikumpulkan melalui kaji selidik menggunakan borang soal selidik yang baru dan disahkan. Disamping itu, kajian ini mengungkapkan dinamik kompleks risiko orang ramai terhadap kualiti air minuman yang selamat berkaitan dengan EDC serta faktor-faktor yang mempengaruhinya. Seterusnya, ia memberi konseptualisasi komprehensif mengenai ramalan persepsi risiko persekitaran, amanah, sikap dan pengetahuan. Orang awam cenderung untuk menilai risiko melalui sistem pemprosesan yang tidak berasas yang tertakluk kepada pengaruh kognitif dan afektif. Persepsi pencemaran EDC dalam air paip didapati tidak bersekutu dengan nilai yang diukur. Pemantauan mutu air dan penilaian risiko adalah proses awal dalam pendekatan multibarrier untuk memastikan sumber air yang selamat dalam sistem bekalan air minuman. Pembangunan model persepsi risiko dapat memenuhi jurang persepsi risiko serta mengenalpasti peranan persepsi risiko dalam merumuskan langkah-langkah pencegahan dan intervensi yang cekap dengan kumpulan sasaran dan bahan yang diketahui. Kepercayaan kepada pihak berkepentingan dan persepsi risiko alam sekitar terhadap pendedahan manusia yang lebih tinggi, serta sikap positif dalam peningkatan kualiti air minum dapat meningkatkan persepsi risiko masyarakat terhadap air minum. Berikutnya, perbaikan dari segi perkembangan tingkah laku risiko orang awam adalah juga penting dalam mengurus tadbir dan komunikasi risiko yang berkesan, sekaligus menyokong pendekatan multibarrier yang disebutkan di atas.

Kata kunci: Sebatian mengendala endokrin (EDC); SPE-LC-MS/MS; Air paip; Penilaian risiko; Persepsi risiko; Keselamatan air minuman

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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AB	Gastrointestinal absorption rate
AChE	Acetylcholinesterase
ACN	Acetone
ADI	Acceptable daily intake
AF	Assessment factor
AhR	Aryl hydrocarbon receptor
ANOVA	One-way analysis of variance
AR	Androgen receptor
B40	Bottom 40% income group
BW	Body weight
CAT	Catalase
CCL	Contaminant Candidate List
CH_3COOH	Acetic acid
ChV	Chronic value
CNS	Central nervous system
C_s	Maximum detected concentration
CV	Coefficient of variation
DDE	Dichlorodiphenyldichloroethylene
DDT	Dichlorodiphenyltrichloroethane
DWEL	Drinking water equivalent level
DWI	Daily drinking water intake
DWI/BW	Daily water intake per body weight
DWTPs	Drinking water treatment plants
EC50	Effective concentration of 50%
EDCs	Endocrine disrupting compounds
EDSP	Endocrine Disruptor Screening Program
ER	Estrogen receptor
ESI	Electrospray ionization
EU	European Union
FAI	Free androgen index
FOAD	Fetal origins of adult disease
FOE	Frequency of exposure
GAC	Granular activated carbon
GnRH	Gonadotropin-releasing hormone
GR	Glucocorticoid receptor
GSI	Gonadosomatic index
GST	Glutathione S-transferase
HAAs	Halogenated acetic acids
HCl	Hydrochloric acid
HMW	High molecular weight
HPLC	High performance liquid chromatography
HQ	Hazard quotient
IBR	Integrated biomarker response
IGFBP1	Insulin growth factor binding protein 1
IUGR	Intrauterine growth retardation
K_{OC}	Soil sorption coefficient
LC50	Lethal concentration of 50%

LC-MS	Liquid chromatography mass spectrometry
LC-MS/MS	Liquid chromatography-tandem mass spectrometry
LIF	Lipofuscin
LMS	Lysosomal membrane stability
LMW	Low molecular weight
log K_{OW}	Octanol-water partition coefficients
M	Mean
M40	Middle 40% income group
MBRs	Membrane bioreactors
MDL	Method detection limit
MEC	Maximum measured environmental concentration
MeCN	Acetonitrile
MeOH	Methanol
Min	Minimum
Max	Maximum
MLR	Multiple regression analysis
MOFs	Multiple oocyte follicles
MQL	Method quantification limit
MRM	Multiple reaction monitoring
NA	Not available
Na_2EDTA	Tetrasodium ethylenediamine-tetraacetate dehydrate
NABC	Needs, approaches, benefits and challenges analysis
ND	Not detected
NF	Nanofiltration
NH_4OH	Ammonium hydroxide
NIS	Sodium/iodide symporter
NR	Nuclear receptor
NRRA	Neutral red retention assay
NSAID	Nonsteroidal anti-inflammatory drug
OCPs	Organochlorine pesticides
OPPs	Organophosphorus pesticides
PAHs	Polycyclic aromatic hydrocarbons
Pax8	Paired-box gene 8
PBBs	Polybrominated biphenyls
PBDEs	Polybrominated diphenyl ethers
PCA	Principal component analysis
PCBs	Polychlorinated biphenyls
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctanesulfonic acid
PNEC	Predicted no-effect concentration
POPs	Persistent organic pollutants
PPAR	Peroxisome proliferator-activated receptor
PPCP	Pharmaceuticals and personal care product
PR	Progesterone receptor
QA	Quality assurance
QC	Quality control
RO	Reverse osmosis
ROS	Reactive oxygen species
RQ	Risk quotient
RQ_{mix}	Risk quotient of the mixture

RSD	Relative standard deviation
S/N	Signal-to-noise
SD	Standard deviation
SDGs	Sustainable Development Goals
SPE	Solid phase extraction
STP	Sewage treatment plant
SWOT	Strengths, weaknesses, opportunities and threats analysis
T20	Top 20% income group
TG	Thyroglobulin
THMs	Trihalomethanes
TR	Thyroid hormone receptor
TRC	Toronto Research Chemicals
TSHβ	Thyroid stimulating hormone beta-subunit
TTF1	Thyroid transcription factor 1
TTR	Thyroid transport though reduction of transthyretin
UF	Ultrafiltration
UPW	Ultrapure water
US EPA	United States Environmental Protection Agency
US FDA	United States Food and Drug Administration
VTG	Vitellogenin
WHO	World Health Organization
WWTP	Wastewater treatment plant

CHAPTER 1

INTRODUCTION

1.1 Background of the study

Drinking water has ambivalent effect on the public health, as drinking water is a potential source of human exposure to pollutants. However, the focus has predominantly been on the concerns related to the occurrence of contaminants in the environment and these contaminants have primarily arisen from industrial origin since the beginning of the Industrial Revolution in 1784. Water sources, such as rivers, are a favorable way to dispose of industrial waste and groundwater is often contaminated via leaching from dumping sites, contributing to public health diseases via ingestion, inhalation and dermal absorption.

The occurrence of emerging organic contaminants, particularly endocrine disrupting compounds (EDCs) in drinking water, was resurrected as a major concern in the mid-1970s after the first mention in 1965, yet there were few studies after the mid-1970s (Daughton et al., 2016). Scientific efforts were focused on other environmental contaminants such as heavy metals (e.g., lead, which is regarded as the oldest global contaminant) before the potential environmental impacts of EDCs as contaminants emerged as a widespread concern after some were found to have potential environmental impacts with similar health risks i.e., endocrine dysfunction in living things. With the limited epidemiological studies and experimental toxicology studies, most of the studies that have been reported thus far are case studies on the presence and risk of EDCs in aquatic systems and treatment plants (wastewater and water) (Barber et al., 2015; Liu et al., 2017; Omar et al., 2018; Padhye et al., 2014).

Currently, occasional legislative and policy enactments in countries throughout the world have demonstrated the challenges related to achieving water security to ensure better quality and healthier lives, regardless of their development level. The number of water and health-related issues is expected to increase due to ongoing natural disasters (extreme events triggered by global warming phenomena) and anthropogenic hazards (oil spills and discharge under industrial as well as domestic activities) (Figure 1.1). The subsequent potential outbreaks of waterborne diseases and the increasing number and occurrence of emerging organic contaminants challenge the regulatory agencies responsible for the creation of legislation and policies. With the adoption of Sustainable Development Goals (SDGs) by the United Nations, most countries have become committed to providing a safe drinking water supply, thereby safeguarding public health for all by 2030. In the contexts of safely managed drinking water and sanitation services, the quality of a drinking water supply with minimized risks of priority chemical contamination forms part of the concern. Additionally, the United States Environmental Protection Agency (US EPA) has published a list of priority drinking water contaminants for regulatory consideration. The Contaminant Candidate List (CCL) includes endocrine disruptors such as pesticides and pharmaceuticals (veterinary and human).

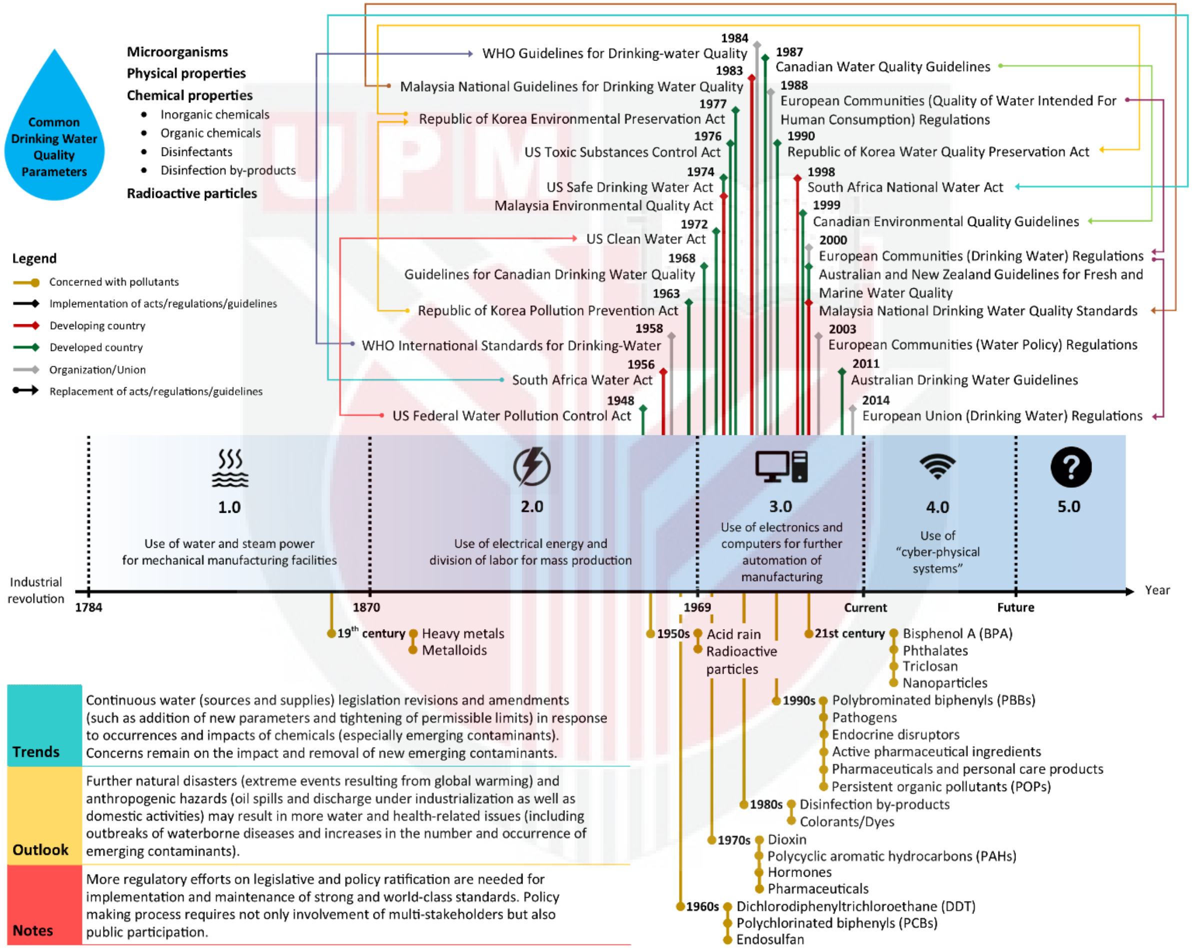

Figure 1.1: Chronology of (a) major emerging concerns related to water pollution and (b) new and changes in water legislation in relation to industrialization.

Unlike tap water, bottled water was commonly regulated as a food such as European Communities (Hygiene of foodstuffs) Regulations and United States Federal Food, Drug and Cosmetic Act. A review of the existing drinking water regulations revealed that the commonly regulated drinking water quality parameters include microorganisms, radioactive particles, physical properties and chemical contents (e.g., inorganic chemicals, organic chemicals, disinfectants and disinfection byproducts) (Figure 1.1). Organic pollutants, such as pesticides, petroleum products and solvents, have been greatly explored and developed. Nonetheless, the World Health Organization (WHO) has revealed that the current WHO Guidelines for Drinking Water Quality are inadequate based on the current exposure levels to EDCs in drinking water (WHO, 2011a). Thus, the public is inadvertently exposed to EDCs via drinking water consumption; moreover, the associated risks may have been underestimated and unknown and remain to be investigated.

Due to the profound health risks of EDCs, preventive and intervention actions are required to regulate the occurrence of EDCs in the drinking water supply to ensure access to safe drinking water. The adverse impacts and knowledge gaps that are pertinent to the involvement of the public and government in regulating emerging EDCs. In the context of improving risk governance and communication, public improvement is the most effective measure to create awareness of risk behavior (Wachinger et al., 2013). Moreover, risk perception has a potential mediating influence on the development of risk behavior (e.g., preparedness, reduction, prevention and mitigation) (Martin et al., 2009). Based on the unknown public-perceived risks on EDCs exposure via drinking water consumption, the role of the public risk perception in regulating EDC contents in the drinking water supply remains unknown.

1.2 Problem statement

Currently, EDCs had been observed in human fluid such as urine, blood, sweat and breast milk (Azzouz et al., 2016; Faniband et al., 2014; Shekhar et al., 2017). EDCs are persisted and dispersed in the environment through (i) wastewater treatment plant (WWTP) and sewage treatment plant (STP) effluents, (ii) human, livestock and animal excretion, (iii) manufacturing and application (e.g., medicine formulation and pest control) and (iv) environmental processes such as runoff and infiltration (Aris et al., 2014; Simazaki et al., 2015). Figure 1.2 depicts the causes and effects of EDCs in drinking water supply system. EDCs are highly persistent, bioaccumulative, toxic and long-range transported due to the high resistance to chemical, physical and biological transformation. Apparently, the elevated concentrations of EDCs in most drinking water sources contribute to relatively incomplete removal of EDCs in drinking water treatment plants (DWTPs) (Gou et al., 2016; Simazaki et al., 2015). Also, conventional drinking water treatments, which commonly used in developing countries, were largely known for not being efficient in removing EDCs, whereas advanced treatments, with increased removal rates, were not able to obtain complete removal, particularly owing to the biological persistency and hydrophilicity of EDCs (Boleda et al., 2011; Kim et al., 2007). Climate change was also identified as the cause of the occurrence and distribution of EDCs such as pharmaceuticals and pesticides with the associated health risks (Chiu et al., 2017; Coppens et al., 2015). Leaching of plasticizers from food packaging materials especially plastic bottles alerts the potential human daily intake and health issues of endocrine disruptors, plasticizers (Li et al., 2010). Recently, water purifiers were identified as a source of contamination organophosphate flame retardants in the drinking water supply (i.e., tap water, purified water and bottled water) of Koreans population (Lee et al., 2016).

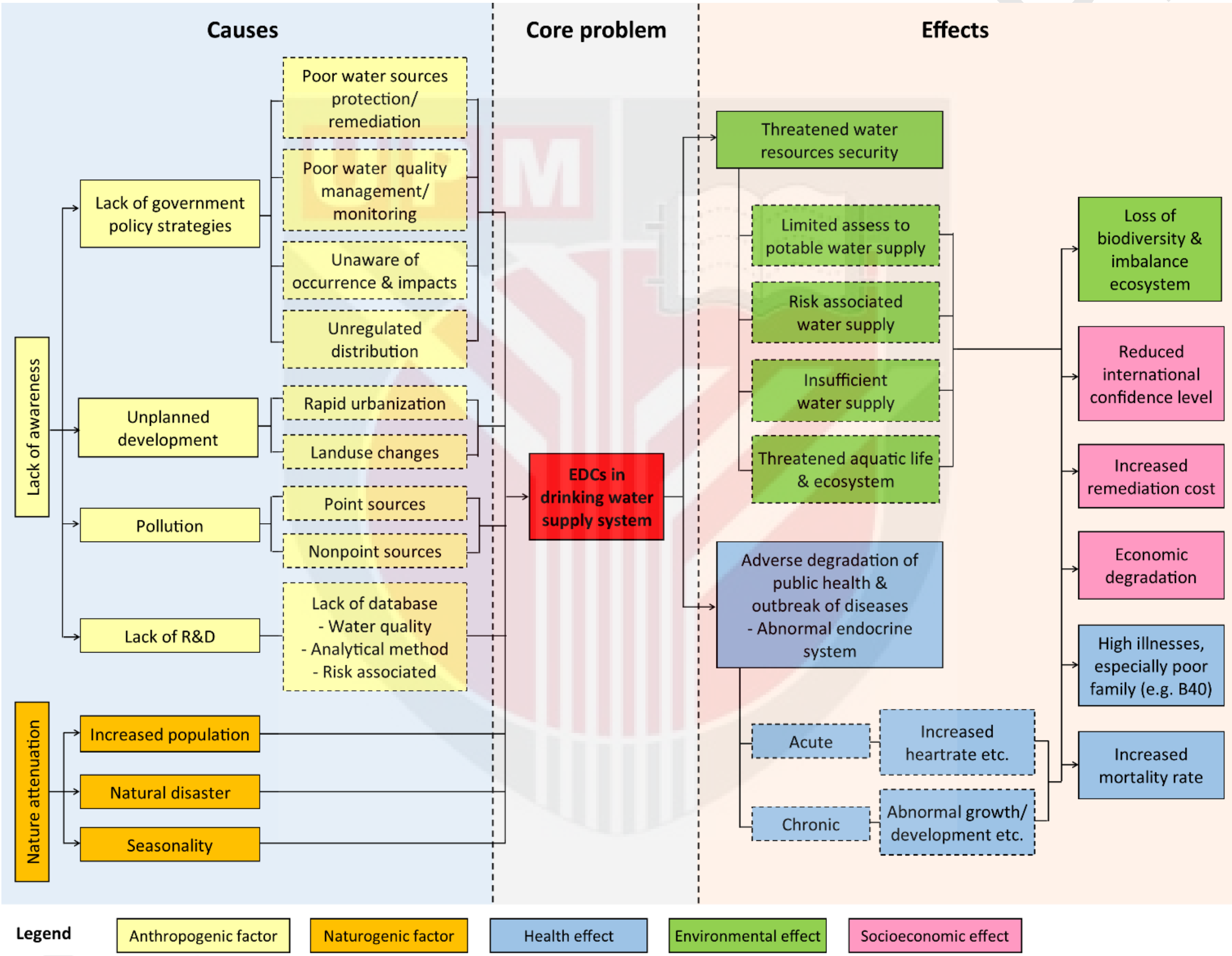

Figure 1.2: Causes and effects of endocrine disrupting compounds in drinking water supply system.

Lifelong water consumption has led to a pharmaceutical exposure level (< 10% of a daily medical dose), even after the treatment of drinking water reduced the negligible risk by up to 80% (Houtman et al., 2014). The potential health issues of EDCs are not fully understood yet due to there being relatively scarce research on their exposure and associated risks via drinking water consumption. Nonetheless, the actual risks, i.e., knowledge gaps, urgently need to be evaluated to confirm or disprove the effects of EDCs via drinking water consumption. In general, EDCs interfere with the endocrine system, particularly hormone signals, by antagonizing the modes of action and mechanisms of endogenous hormones, especially through nuclear receptors. The endocrine dysfunction effects in exposed individuals and populations ranging from acute to chronic diseases, namely, epigenetic deregulation, immune effects, metabolic syndromes, reproductive abnormalities, behavioral changes, disrupted fetal development and growth, neurological disorders and abnormal cell proliferation. The consequences depend on several factors, including the age at exposure, exposure duration (both external and internal exposure), exposure magnitude (dosage) and presence of other pollutants (Kabir et al., 2015). In particular, the potential hormesis in pollutant exposure, which presents a nonlinear dose-response (low doses stimulate extremely adverse effects, while higher doses have no effect), remains largely unknown (Jiang et al., 2016).

The risk of exposure to inorganic contaminants, such as heavy metals and arsenic in drinking water, is predominantly given high priority, especially in many developing countries (Chappells et al., 2014; Chowdhury et al., 2016; Flanagan et al., 2015). Nevertheless, it is not a common international practice to regulate or provide guidelines for EDCs in drinking water, indicating that there are knowledge gaps and undervalued perspectives on the potential occurrence of EDCs in drinking water and the health risks associated with EDCs via drinking water consumption. The growing concern regarding EDCs is limited to their environmental risks and occupational health and safety risks, whereas the perceived risks of EDC exposure via drinking water consumption remain unknown.

Given that the trace level of EDCs and the various matrix interferences, detection and quantification of the broad scope of EDCs in environmental matrices are formidable challenges. EDC monitoring has high cost for simultaneous development and optimization of analytical methods, followed by the need of highly sensitive instruments for method validation and sensitivity improvement. To date, the development of extraction method was circumscribed to single EDC group and its metabolites in tap water analysis (Gaffney et al., 2015; Gros et al., 2012; Leung et al., 2013). The wide diversity of complex EDCs also challenges risk assessment and risk management, especially in term of the associated risks of combined exposures such as EDCs mixtures. Moreover, inadequate information on degradation and transformation of the complex EDCs during treatment processes and the potential risks derived from the EDC mixtures, metabolites and by-products challenge the risk characterization. Treatment and remediation processes are subjected to numerous ongoing studies because greenhouse effect and cost effectiveness are also the factors of the removal efficiency in this time of growing energy crisis, considering sustainable development for the whole system. Therefore, the current databases (i.e., monitoring data, treatment processes, metabolism and associated risks) and regulations (usage and/or manufacturing controls, discharge and/or disposal practices, environmental quality standards and drinking water regulatory compliance) of emerging EDCs may be inadequate for governing and mitigating EDCs to protect the environment and ensure access to safe drinking water.

Therefore, EDC occurrence in the drinking water supply and the potential health risks associated with EDCs via drinking water consumption should not be neglected. Notably, one cannot say there are no potential risks of EDC consumption via daily drinking water intake; thus, the potential risks and knowledge gaps should be urgently evaluated to confirm or disprove this. Apparently, scientific efforts are solely required to fill this knowledge gap to ensure safe access to drinking water. These efforts concern not only the scientific community but also the public, thus challenging the adoption of preventive and mitigation measures in regulating EDCs in drinking water. This aspect is a challenge for the scientific community that works on evidence/data in nature and the extent of contact with EDCs; thus, the public is not expected to demonstrate good risk behavior (e.g., awareness and concern) and participation.

Meanwhile, the role of the perceived risks in regulating EDC contents in the drinking water supply remains unknown. Currently, the relatively low public awareness of and political responsibility for (i) water source protection, (ii) water supply security, (iii) risk assessment (environmental and health), (iii) treatment efficiency and (iv) future prediction are challenges for the creation of legislation and policies related to a safe drinking water supply. This challenge is clearly observable when the public risk perception and actual drinking water quality are inconsistent and the association is instead skewed (Ochoo et al., 2017). Existing legal regulations are incompetent to regulate the occurrence and distribution of EDCs, while politically unprioritized and/or unregulated EDC usage and discharge lead to continuous EDC contamination. Therefore, this study aims to address the following research questions:

1. How to determine and quantify multiclass EDCs in drinking water sources i.e., river water and drinking water supply i.e., tap water?
2. What is the concentration of multiclass EDCs in drinking water supply system in Malaysia?
3. How are multiclass EDCs distributed in drinking water supply system (between river water and tap water and within each matrix) in Malaysia?
4. What is the local exposure to multiclass EDCs?
5. Do the multiclass EDCs pose potential ecological risks in riverine ecosystem and/or human health risks among different life stages through drinking water consumption?
6. How does the public perceive risks of multiclass EDCs in drinking water supply system?
7. What are the influencing factors of public in perceiving multiclass EDCs in drinking water supply system?
8. Does association exist between actual and perceived drinking water quality of the public in regard to EDCs?

1.3 Objectives of the study

This study aims to determine the occurrence of multiclass EDCs (pharmaceuticals, hormones, plasticizers and pesticides) in drinking water supply system using the newly established and validated analytical protocol based on solid phase extraction followed by liquid chromatography-tandem mass spectrometry (SPE-LC-MS/MS). Potential ecological and human health risk of multiclass EDCs in drinking water supply system will be further appraised. Nevertheless, qualitative analysis, questionnaire survey on the derivatives of human risk assessment will be done for better reflection on local exposures to EDCs based on the particular human morphology, drinking water consumption pattern and household practices. Also, questionnaire survey involves evaluation of the public-perceived risks of drinking water quality with potential EDCs contamination in tap water, the influencing factors and the association between actual and perceived quality that remain unknown to be investigated.

Specific objectives of this study are:

1. To establish a valid and reliable protocol for simultaneous analysis of trace level of multiclass EDCs in tap water in a single extraction step.
2. To determine and compare the spatial level and distribution of multiclass EDCs in urban drinking water supply system.
3. To appraise the potential ecological risk of EDCs in drinking water sources i.e., river water and human health risk of EDCs in drinking water supplies among different life stages via drinking water consumption.
4. To elucidate the public-perceived risks of EDCs in multiclass EDCs in urban drinking water supply system, association between actual and perceived drinking water quality and influencing factors.

1.4 Scope of the study

This study covers the following:-

1. Establishment of a valid analytical method for simultaneous analysis of multiclass EDCs in tap water in a single extraction step.
2. Introduction of a tool in multibarrier approach for safe drinking water which serves as an analytical procedure for determination of multiclass EDCs contamination in tap water elsewhere.
3. Update on current status of multiclass EDCs in term of the occurrence, variation, distribution in the drinking water supply system (from source to supply).
4. Risk characterization, profiling and prioritization of EDCs in Malaysian drinking water supply system (human health and ecological impacts).
5. Questionnaire development, validation and survey for local exposure study.
6. Generating database of EDCs level, the associated risks, human morphology, drinking water consumption patterns and household practices.
7. Identification of vulnerable age group in EDCs exposure via daily water intake.

8. Identification of public risk perception, influencing factors and association between actual and perceived risk for public improvement in terms of risk behavior (e.g., preparedness, reduction, prevention and mitigation).
9. Regulating the gap of public risk perception for effective risk communication and governance.
10. Proposing a risk management and monitoring framework for multiclass EDCs in the drinking water supply system to support multibarrier approach in drinking water supply system monitoring and management for safe water resources (raw water and treated water).

1.5 Significance of the study

This study can contribute to comprehensive exposure studies in determining the nature and extent of contact with EDCs. In respect to safeguarding human health via safe drinking water, this study represents the solution for detection of the broad scope of EDCs in tap water at trace concentration. This is expected to be useful in supporting multibarrier approach in drinking water supply system monitoring and management for safe water resources (from source to tap), especially drinking water supply security that remain unknown to be investigated. Meanwhile, the validated and verified method serves as an analytical procedure for determination of trace level of multiclass EDCs contamination in tap water elsewhere. Thereby, EDCs monitoring would not only limit to the environmental compartments (e.g., surface water, sediments and biota) but also drinking water supply, concerning the human health risks via daily water intake aside from the ecological risks that were largely known. Thus, this study is expected to be useful for water resources monitoring and management purposes, especially in regulating water supply contamination and human health risk implication.

Consequently, this study can be used as a principal basis for legislative and policy making, especially regarding the integration of risk mitigation into existing legislative and policy frameworks to prescribe more stringent drinking water regulatory compliance. In the context of improving risk governance and communication, revisiting drinking water quality with preventive and intervention measures ensure safe drinking water access and reduce the extent of EDC exposure and risk to the environment and humans. Overall, an in-depth focus on the public-perceived risks of EDCs in drinking water and the potential factors influencing the risk perceptions of a population are substantially efficient to understand the public risk perceptions of human exposure and the associated health risks and accelerate the public awareness.

Specifically, these are applicable in developing countries such as Malaysia where numbers of catchment are highly urbanized and anthropogenically impacted; while drinking water treatment plants are commonly equipped only with conventional technology. Revealing the distribution level and updating the current status of EDCs in Malaysian drinking water supply system with the detailed human health risk assessment, the toxicity level and human health risk of EDCs exposure through consumption of drinking water supply among adults and children can be known. Ascertaining the unknown risk perceptions of EDCs in drinking water, the unknown role of perceived risks in ensuring access to safe drinking water could be appraised for controlling the occurrence of such potential risks in the global water system.

1.6 Research Framework of the study

The research framework developed for the study on multiclass EDCs in drinking water supply system, risk assessment and public risk perception is depicted in Figure 1.3. The research framework is detailed out in the form of research matrix (Table 1.1). Figure 1.4 explains the methodology of the study from preparation to measurement, followed by exposure and risk assessment, as well as the risk perception analysis.

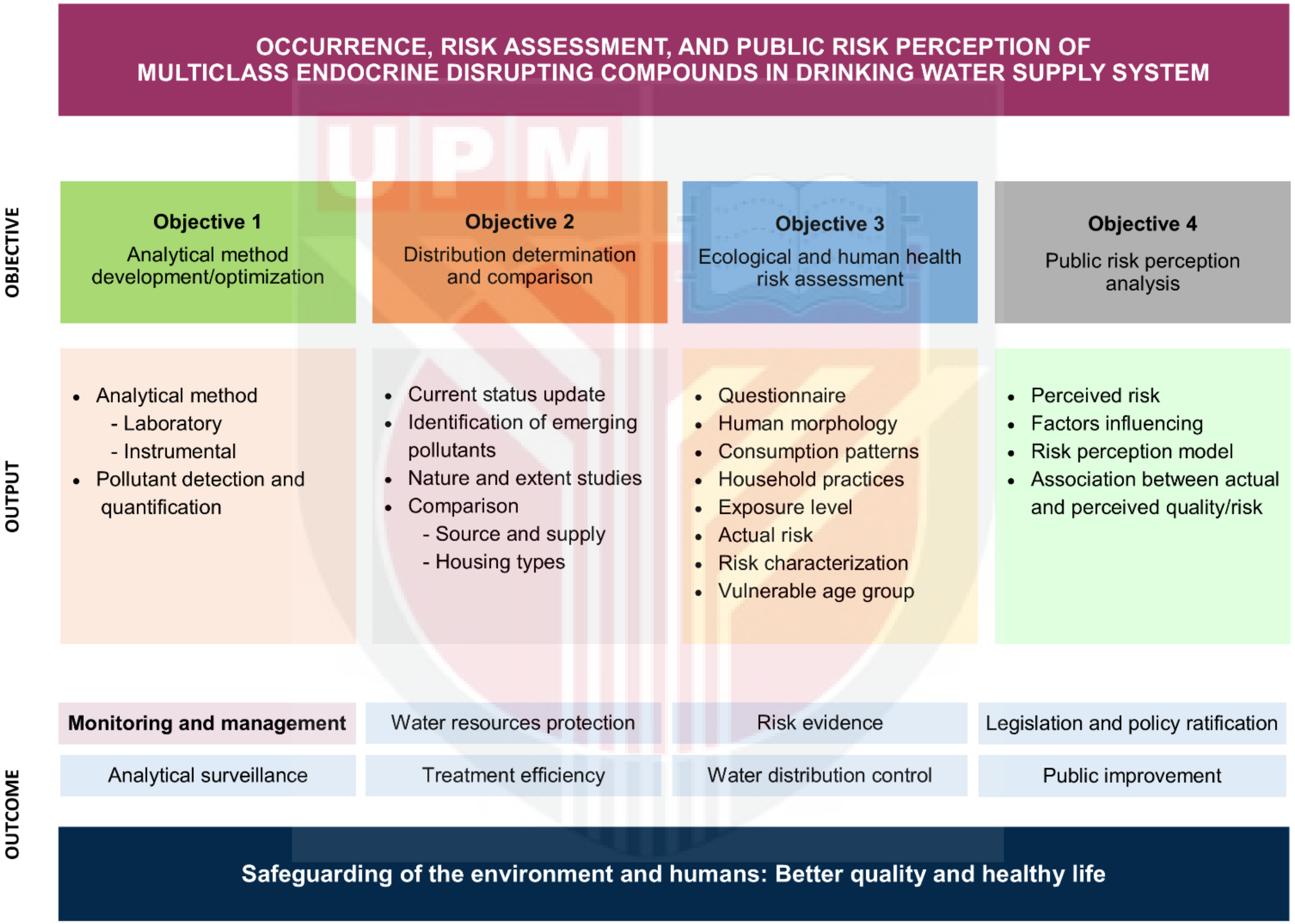

Figure 1.3: Research framework for the study on multiclass endocrine disrupting compounds in drinking water supply system, risk assessment and public risk perception.

Table 1.1: Research matrix for the study on multiclass endocrine disrupting compounds in drinking water supply system, risk assessment and public risk perception.

Justification

- EDCs contribute to wide ranging diseases due to abnormal endocrine system, particularly disruption of nuclear receptors in the exposed individuals and populations.
- EDCs are persisted and dispersed in the environment through: (i) wastewater treatment plant (WWTP) and sewage treatment plant (STP) effluents, (ii) human, livestock and animal excretion and (iii) agricultural practices (e.g., pest control and fertilizer application).
- EDCs are highly persistent, bioaccumulative, toxic and long-range transported due to the high resistance to chemical, physical and biological transformation.
- The subsequent elevated concentration of EDCs in the most drinking water sources contribute to the relatively poor removal of EDCs in drinking water treatment plants (DWTPs).
- The extraneous contaminations (e.g., pipe leaching and leaking) in drinking water distribution network also contribute to EDCs contamination in drinking water supply, typically tap water, posing possible risks to public health.
- Drinking water supply, such as tap water, is an additional and crucial route of human exposure to the health risks associated with EDCs.

Research problems

- Lack of research and development (i.e., low quality and robustness of the database on water quality monitoring and risk assessment).
- Trace level contamination of the wide diversity of complex EDCs challenges: (i) detection and quantification of the broad scope of EDCs in the interference-rich environmental matrices and (ii) simultaneous development and optimization of analytical methods, followed by method validation and sensitivity improvement.
- Unwarranted control of drinking water source quality and treatment efficiency in drinking water supply systems.
- EDC exposure to humans via drinking water consumption.
- Interruption of human health risk assessment for risk mitigation because of inadequate databases, for example, (i) degradation and transformation of the complex EDCs during treatment processes and (ii) potential risks derived from the EDC mixtures, metabolites and by-products.
- Lack of public awareness in water supply security and water source protection from EDCs.
- Unknown public risk perception of EDCs contamination in water supply and their role in risk governance and communication.
- Existing legal regulations are incompetent to regulate the occurrence and distribution of EDCs.
- Politically unprioritized and/or unregulated EDC usage and discharge lead to continuous EDC contamination.

Objectives	Activities	Findings/Outputs	Publications/Intellectual properties
1. To establish a valid and reliable protocol for simultaneous analysis of trace level of multiclass EDCs in tap water in	✓ EDCs (18) extraction method (laboratory) development and optimization for tap water and river water analysis. ✓ EDCs (18) chromatographic condition (instrumental) development and optimization for tap water and river water analysis.	✓ A validated method for detection and quantification of EDCs (18) in tap water and river water employing SPE-LCMS/MS at lower method detection limit.	1. Wee, S.Y., Tuan Omar, T.F., Aris, A.Z. and Lee, Y. 2016. Surface water organophosphorus pesticides concentration and distribution in the Langat River, Selangor, Malaysia. *Expo. Health* 8: 497-511. [Q1, IF: 4.762, Top 5%] 7. Wee, S.Y., Ismail, N.A.H., Haron, D.E.M., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. Analysis

a single extraction step.		✓ Quality assurance (QA) and quality control (QC) e.g., precision, accuracy, linearity, recovery etc. ✓ Method detection and quantification limit.	of pharmaceuticals, hormones, plasticizers, and pesticides in drinking water. [*Environ. Pollut.* - Under review, Q1, IF: 6.792, Top 10%]
2. To determine and compare the spatial distribution of multiclass EDCs in urban drinking water supply system.	✓ Detailed review on EDCs occurrence and distribution in drinking water supply system, especially tap water. ✓ Tap water and river sampling and analysis for preliminary study. ✓ Tap water (drinking water supply) sampling (*n* = 155). ✓ River water (drinking water source) sampling (*n* = 10). ✓ Physicochemical analysis for descriptive analysis. ✓ EDCs (18) extraction and quantification in tap water and river water. ✓ Statistical analysis. - One-way analysis of variance (ANOVA) for significance of the difference between EDCs. - Bivariate analysis for linear relationship between physicochemical properties and the occurrence of EDCs. - Independent *t*-test for comparison between different water and housing types.	✓ Preliminary study on EDCs content in tap water. ✓ Determination of the occurrence and distribution of EDCs (18) in tap water and river water. ✓ Expected factors contributing to the occurrence of EDCs (18) in drinking water supply system. ✓ Human exposure to EDCs (18) via drinking water consumption. ✓ Proposed solution for reducing EDCs in drinking water supply system.	1. Wee, S.Y., Tuan Omar, T.F., Aris, A.Z. and Lee, Y. 2016. Surface water organophosphorus pesticides concentration and distribution in the Langat River, Selangor, Malaysia. *Expo. Health* 8: 497-511. [Q1, IF: 4.762, Top 5%] 2. Wee, S.Y. and Aris, A.Z. 2017. Endocrine disrupting compounds in drinking water supply system and human health risk implication. *Environ. Int.* 106: 207-233. [Q1, IF: 7.577, Top 5%] 5. Wee, S.Y., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. 2019. Occurrence and risk assessment of multiclass endocrine disrupting compounds in an urban tropical river and a proposed risk management and monitoring framework. *Sci. Total Environ.* 671: 431-442. [Q1, IF: 6.551, Top 10%] 6. Wee, S.Y., Haron, D.E.M., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. 2020. Active pharmaceutical ingredients in Malaysian drinking water: consumption, exposure, and human health risk. *Environ. Geochem. Health* 1-15 [Q1, IF: 3.472, Top 15%]. 10. Wee, S.Y., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. Multiclass endocrine disrupting compounds in tap water of different housing types [*Chemosphere* - Under review, Q1, IF: 5.778, Top 10%].

				11. Wee, S.Y., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. Comparison of the target multiclass endocrine disrupting compounds in drinking water supply system [*Sci. Rep.* - Under review, Q1, IF: 3.998, Top 20%]
3. To appraise the potential ecological risk of EDCs in drinking water sources i.e., river water and human health risk of EDCs in drinking water supplies among different life stages via drinking water consumption.	✓ Questionnaire designing for local studies. ✓ Questionnaire validation. ✓ Pilot study. ✓ Data collection. ✓ Data input. ✓ Data analysis.	✓ Risk calculation and interpretation for ecological and human health risk ranking. ✓ Data analysis. ✓ Descriptive analysis. ✓ Statistical analysis.	✓ Potential ecological risk of EDCs in drinking water sources. ✓ Potential human health risk of EDCs among age groups through consumption of drinking water. ✓ Database creating on human morphology, drinking water consumption patterns and household practices. ✓ Risk characterization and risk prioritization for risk management and risk mitigation. ✓ Identification of vulnerable groups.	3. Wee, S.Y. and Aris, A.Z. 2017. Ecological risk estimation of organophosphorus pesticides in riverine ecosystems. *Chemosphere* 188: 575-581. [Q1, IF: 5.778, Top 10%] 5. Wee, S.Y., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. 2019. Occurrence and risk assessment of multiclass endocrine disrupting compounds in an urban tropical river and a proposed risk management and monitoring framework. *Sci. Total Environ.* 671: 431-442. [Q1, IF: 6.551, Top 10%] 6. Wee, S.Y., Haron, D.E.M., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. 2020. Active pharmaceutical ingredients in Malaysian drinking water: consumption, exposure, and human health risk. *Environ. Geochem. Health* 1-15 [Q1, IF: 3.472, Top 15%]. 8. Wee, S.Y., Aris, A.Z., Yusoff, F.M., Praveena, S.M. and Harun, R. Drinking water consumption patterns, household practices, and risk perception on endocrine disrupting compounds [*Environ. Sci. Policy* - Under review, Q1, IF: 4.767, Top 20%] 10. Wee, S.Y., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. Multiclass endocrine disrupting compounds in tap water of different housing types [*Chemosphere* - Under review, Q1, IF: 5.778, Top 10%]. 11. Wee, S.Y., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. Comparison of the target multiclass endocrine disrupting compounds in

			drinking water supply system [*Sci. Rep.* - Under review, Q1, IF: 3.998, Top 20%]. 12. Wee, S.Y., Aris, A.Z., Harun, R. and Praveena, S.M. 2018. Questionnaire - Exposure and risk perception on endocrine disrupting compounds (EDCs) in Malaysian tap water. Copyright, LY2018000940.
4. To elucidate the public-perceived risks of EDCs in multiclass EDCs in urban drinking water supply system, association between actual and perceived drinking water quality and influencing factors.	✓ Detailed review on possible influencing factors. ✓ Questionnaire designing for local exposure analysis. ✓ Questionnaire validation. ✓ Pilot study. ✓ Data collection. ✓ Data input. ✓ Data analysis. ✓ Descriptive analysis. ✓ Statistical analysis: - - One-way ANOVA for significance of the difference between public-perceived risks of EDCs within drinking water quality and variables. - Independent *t*-test between low and high risk perception groups. - Bivariate analysis for correlation between variables. - Principle component analysis (PCA) to identify components and variance explained. - Multiple linear regression (MLR) analysis to identify the influencing factors.	✓ Public risk perception on (i) environmental risks to human exposure and (ii) drinking water quality. ✓ Association between actual and perceived drinking water quality. ✓ Influencing factors. ✓ Risk perception model. ✓ Public engagement and awareness improvement. ✓ Risk communication and governance.	4. Wee, S.Y. and Aris, A.Z. 2019. Occurrence and public-perceived risk of endocrine disrupting compounds in drinking water. *npj Clean Water* 2: 4. [Q1, IF: 4.870, Top 5%] 9. Wee, S.Y., Aris, A.Z., Yusoff, F.M., Praveena, S.M. and Harun, R. Public risk processing on human exposure to environmental endocrine disrupting compounds [*Environ. Pollut.* - Under review, Q1, IF: 6.792, Top 10%]. 12. Wee, S.Y., Aris, A.Z., Harun, R. and Praveena, S.M. 2018. Questionnaire - Exposure and risk perception on endocrine disrupting compounds (EDCs) in Malaysian tap water. Copyright, LY2018000940.

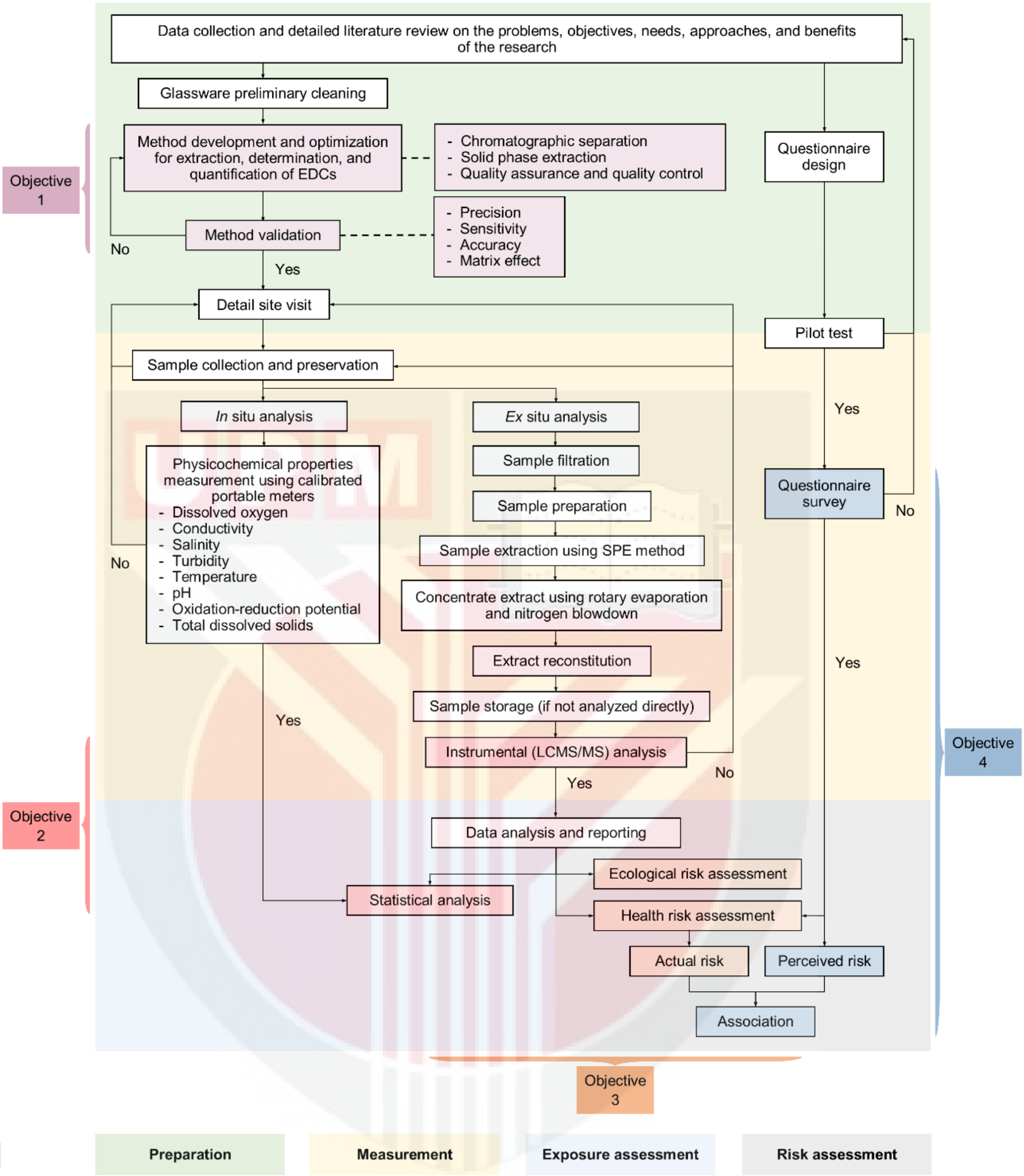

Figure 1.4: Methodology for the study on multiclass endocrine disrupting compounds in drinking water supply system, risk assessment and public risk perception.

1.7 Thesis outline

The body of this thesis consists of seven chapters that cover multiclass EDCs in drinking water supply system, the associated ecological and human health risks, public-perceived risk, association between actual and perceived quality and the influencing factors, concerning safe water for human health and well-being. The chapters, from the introduction to the conclusion are as follows:-

i. Chapter 1 is an introduction with a study background, problem statements, research questions, objectives, scope and significance of the study.
ii. Chapter 2 provides a comprehensive review of literature related to presence of multiclass EDCs, the sources, exposure routes and fate of EDCs in the environment and drinking water, effects and risks of EDCs on exposed individuals and populations, current public-perceived risks of EDCs and the potential influencing factors and proposed solutions for safe water resources.
iii. Chapter 3 demonstrates simultaneous analysis of multiclass EDCs in tap water at trace level, to answer Objective 1.
iv. Chapter 4 comprises water quality (raw and treated) and comparison of multiclass EDCs in drinking water supply system in Malaysia, to answer Objective 2.
v. Chapter 5 elaborates on local human morphology, drinking water consumption patterns, household practices and health risks, to answer Objective 3.
vi. Chapter 6 ascertains public-perceived risks of EDCs in drinking water supply system, association between actual and perceived drinking water quality and the influencing factors, to answer Objective 4.
vii. Chapter 7 summarizes and concludes on the findings and recommendations made on appropriate measures for monitoring and managing water resources.

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BIODATA OF STUDENT

Wee Sze Yee was born on November 25, 1992, in Kuching, Sarawak. She received her early education in SJK (C) Chung Hua No. 4. She completed her secondary education in SMK Green Road for both her Sijil Pelajaran Malaysia (SPM) and Sijil Pengajian Tinggi Malaysia (STPM). She has been staying in Kuching, Sarawak until she continues her Bachelor Degree in the field of environment in the Faculty of Environmental Studies, Universiti Putra Malaysia. In year 2016, she graduated with First Class Honour of Bachelor of Science (Environmental Science and Technology). During her final year project, she had focused in environmental monitoring, involving analytical method optimization and validation for environmental organophosphorus pesticides detection and quantification. Right after, she enrolled for the Master of Science program at Faculty of Environmental Studies, Universiti Putra Malaysia. She further her research in environmental monitoring (organic chemistry and analysis), risk assessment (ecological and human health), and public risk perception, concerning water security and safety in safeguarding the environment and human health for better quality and healthy life. Praise to God, she managed to publish her first hard work in scholarly journal *Exposure and Health* (Q1, IF: 4.532, Top 3%). In year 2017, she managed to covert her study to Doctor of Philosophy (Environmental quality and conservation) based on her research scope, progress, outputs and journal publication. Throughout her study, several scientific papers have been published subsequently (Q1, Top 15%). Not only that, she has been taking part actively in numerous research activities, seminars, workshops and conferences as either a speaker or a participant.

LIST OF PUBLICATIONS

Journals

Wee, S.Y., Haron, D.E.M., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. 2020. Active pharmaceutical ingredients in Malaysian drinking water: consumption, exposure, and human health risk. *Environ. Geochem. Health* 1-15. [Q1, IF: 3.472, Top 15%].

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Wee, S.Y., Aris, A.Z., Yusoff, F.M., Praveena, S.M. and Harun, R. Drinking water consumption patterns, household practices and risk perception on endocrine disrupting compounds in tap water [*Environ. Pollut.* - Under review, Q1, IF: 6.792, Top 10%].

Wee, S.Y., Aris, A.Z., Yusoff, F.M., Praveena, S.M. and Harun, R. Public risk processing on human exposure to environmental endocrine disrupting compounds [*Environ. Pollut.* - Under review, Q1, IF: 6.792, Top 10%].

Wee, S.Y., Aris, A.Z., Yusoff, F.M. and Praveena, S.M. Multiclass endocrine disrupting compounds in tap water of different housing types [*Chemosphere* - Under review, Q1, IF: 5.778, Top 10%].

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Intellectual property

Wee, S.Y., Aris, A.Z., Harun, R. and Praveena, S.M. 2018. Questionnaire - Exposure and risk perception on endocrine disrupting compounds (EDCs) In Malaysian tap water. Copyright, LY2018000940.

Speaker/Presented papers

1. Speaker on “Multiclass endocrine disrupting compounds in tap water of different housing types” at 11th Micropol & Ecohazard Conference 2019 (Micropol 2019), Seoul National University, Seoul, Korea, 20 - 24 October, 2019.
2. Speaker on “Organophosphorus Pesticides in Riverine Ecosystem and Ecological Risk Assessment” at International Conference on Environmental Forensics (iENFORCE2018), Universiti Putra Malaysia, 18 - 19 September, 2018.
3. Speaker on “Organophosphorus Pesticides Contamination in Surface Water of the Langat River, Selangor, Malaysia” at 3rd National Environment and Health Action Plan (NEHAP) Conference 2017, Putrajaya, 25 September 2017.
4. Speaker on “Determination of Multiresidues Organophosphorus Pesticides in River Water” at Regional Professorial Chair Public Talk, organized by Southeast Asian Regional Center for Graduate Study and Research in Agriculture (SEARCA), 21 February 2017.

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