University of Southampton

Faculty of Engineering and Physical Sciences

Water and Environmental Engineering Group

Investigation on Life Cycle Assessment of Centralised Wastewater Treatment, and its Upgrading using LCA

by

Siti Safirah Rashid

Thesis for the degree of Doctor of Philosophy

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Abstract

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Wastewater treatment plants (WWTPs) are designed and operated to prevent pollution to the environment by removing a variety of contaminants from wastewater before discharge. However, the pollutants in wastewater could be transferred to air such as greenhouse gases (GHGs) emission, to water by pollutants in the effluent, and to soil such as disposal of sludge due to wastewater treatment which could lead to negative effects on human health and the environment. The holistic environmental impact from WWTPs is very challenging to evaluate, and thus, a cradle-to-grave approach such as by life cycle assessment (LCA) is needed to analyse the consequences of WWTP's operation to the environment. However, due to the limitation in the life cycle steps, (e.g. lacking of complete local databases of WWTPs), continuous research involving LCA application in wastewater treatment is essential for the improvement of the existing LCA methodology. Thus, it is important to investigate if the potential factors such as rainfall, local toxic pollutants, and the integration of technologies to upgrade existing WWTPs could affect the operation of municipal WWTP and the environmental impacts. The aim of this work is to assess the life cycle impact of large centralised WWTPs based on the extended and comprehensive local databases for the improvement of LCA methodology and towards sustainable operation of wastewater treatment. The complete life cycle inventories based on the existing operation of the selected municipal WWTPs in Malaysia and the UK, were primarily established. Environmental impacts were assessed using LCA to understand the environmental burdens based on three different objectives in the three chapters which are Chapter 3, Chapter 4 and Chapter 5.

Chapter 3 presents a critical assessment on the environmental impacts of large centralised WWTPs, which is Millbrook wastewater treatment work (MWTW) in the United Kingdom (UK) and the Malaysian sewage treatment plant (MSTP) in Malaysia, with combined and separate sewer systems

in wet/dry season respectively, by using LCA. Both wastewater treatment plants (WWTPs) show a lower environmental burden in the wet season than in the dry season partially due to the dilution of wastewater by using FU1 (per 1m^3 treated wastewater). However, the WWTP receiving high strength wastewater (MWTW), demonstrates higher environmental impacts in the wet season by using FU2 (per 1 kg PO_4^{3-}eq. removed), due to the less efficient treatment caused by heavy rainfall in the wet season. Meanwhile, it is found that environmental impacts from the WWTP receiving low strength wastewater (MSTP) have no difference when using either FU1 or FU2. The results indicate that the environmental burdens particularly eutrophication and global warming caused by WWTPs are dependent on the correlations of rainfall intensity with wastewater quantity and quality instead of the combined or separate sewer system.

In the life cycle inventory of WWTPs, the consideration of local toxic pollutants in effluent and sludge which was not included in Chapter 3 could produce different environmental impacts particularly in human and ecotoxicity potentials. In addition, the consideration of local toxic pollutants in a developing country with low strength wastewater could lead to a varying result. Consequently, Chapter 4 presents additional site-specific data involving metals and pharmaceuticals and personal care products (PPCPs) to investigate toxicity impact from a large centralised wastewater treatment plant in Malaysia with low strength wastewater, to provide useful information for LCA practice by identifying the importance and contribution of PPCPs and metals, as well as the difference from model comparison. The result from this study indicates the importance of considering direct toxic pollutants to the environment even in low strength municipal wastewater. PPCPs contributed 11% to FEP using the CML-IA method but only 2.5% using the USEtox method. The reason for the different outcomes between LCIA methods is due to different calculations in the emission background (e.g. air, soil, or freshwater) and variation in the references for toxicity parameters used to calculate CF value of each pollutant. This work identifies the importance to consider toxic pollutants in the environmental assessment from WWTPs due to no apparent patterns of toxic pollutant's concentration and removal in high strength and low strength wastewater in developed and developing countries.

In many developing countries such as in Malaysia, wastewater treatment is still focused on chemical oxygen demand (COD) and suspended solid (SS) removal without considering nutrients. This has caused serious eutrophication problems, especially in nutrient sensitive areas. There is a possibility for large centralised wastewater treatment plants (WWTPs) in developing countries to look into the available nutrient removal technologies for upgrading to alleviate the problems caused by nutrient discharge into natural waters which were not considered in Chapter 3 and Chapter 4. Chapter 5 presents the design of three upgraded processes with nutrient removal and resource recovery for a typical Malaysian centralised WWTP and its comparative assessment of environmental impacts and economic cost. Process A is based on EBPR for P removal, nitrification and denitrification for N removal and AirPrex for P recovery. Process B is to use ferric precipitation to remove P, nitrification and denitrification to remove N, and Gifhorn to recover P from sludge. Process C is to adopt aerobic granular sludge (AGS) technology to do simultaneous N and P removal, and Gifhorn for P recovery. In terms of environmental impact, Process B shows the worst environmental impact in terms of FU1 (per m^3 treated wastewater), while Process A had the highest environmental burden in terms of FU2 (per kg struvite recovered). Process C has the least environmental impact with either of FUs. The total life cycle cost of Processes A, B and C are averagely 24% higher than the existing process. Overall results suggest that Process A is the optimum option if low financial impact are considered. But, in terms of environmental and technical benefits, Process C is the best option. The quantitative information from this study could provide guidance in decision making on upgrading the existing WWTPs especially in regions with diluted wastewater, which will underpin the transition towards a sustainable wastewater treatment.

Overall, this research work identifies the importance to consider local factors such as rainfall, site-specific inventory data, and the existing technology used, when selecting technology and designing process to upgrade WWTPs, in the environmental impact assessment of WWTPs. This research reports for the first time that the correlation of rainfall on wastewater inflow to the WWTP with a separate sewer system is similar to that WWTP with a combined sewer system, which will cause a paradigm shift for assessment municipal of WWTPs. Nutrient, heavy metals and PPCPs in influent and effluent, and electricity consumption are the major factors to affect environmental impacts. This research also identified wastewater strength, functional units and selection of LCIA method as critical factors affecting life cycle assessment and result interpretation for the assessment of WWTPs. Furthermore, the newly design work for nutrient removal and resource recovery from this study have the potential to guide future upgrading process in conventional wastewater treatment especially in the regions with diluted wastewater. The results in this study benefit to the wastewater management as there is increased pressure to reduce eutrophication impact in the water body, could guide for new policy making, and for future sustainable and efficient operation of municipal WWTPs in both developing and developed countries. Finally, the work in this study provide valuable knowledge and guidance for LCA methodology improvement when considering comprehensive and detailed local inventories in the analysis.

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Research Thesis: Declaration of Authorship

Print name: Siti Safirah Rashid

Title of thesis: Investigation on Life Cycle Assessment of Centralised Wastewater Treatment, and its Upgrading using LCA

I declare that this thesis and the work presented in it are my own and has been generated by me as the result of my own original research.

I confirm that:

1. This work was done wholly or mainly while in candidature for a research degree at this University;
2. Where any part of this thesis has previously been submitted for a degree or any other qualification at this University or any other institution, this has been clearly stated;
3. Where I have consulted the published work of others, this is always clearly attributed;
4. Where I have quoted from the work of others, the source is always given. With the exception of such quotations, this thesis is entirely my own work;
5. I have acknowledged all main sources of help;
6. Where the thesis is based on work done by myself jointly with others, I have made clear exactly what was done by others and what I have contributed myself;
7. Parts of this work have been published as:

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Definitions and Abbreviations

A-B	Adsorption-biological
AD	Anaerobic digestion
ADFP	Abiotic depletion (fossil fuel) potential
AGS	Aerobic granular sludge
AOA	Anaerobic/aerobic/anoxic
AP	Acidification potential
AS	Activated sludge
ASP	Activated sludge process
BOD_5	5-day biochemical oxygen demand
CAS	Conventional activated sludge
CBA	Cost-benefit analysis
CF	Characterisation factor
CHP	Combined heat and power
COD	Chemical oxygen demand
CSO	Combined sewer overflow
CTU	Comparative toxic unit
DOE	Department of environment
EBPR	Enhanced biological phosphorus removal
EEA	Economic efficiency analysis
EEC	European Economic Community
EIA	Environmental impact assessment
ELCD	European life cycle database
EP	Eutrophication potential
EPA	Environmental protection agency
Eq.	Equivalent
EU	European Union
FEP	Freshwater ecotoxicity potential
FiT	Feed-in-tariff
FU	Functional unit
GHG	Greenhouse gases

GWP	Global warming potential
HTP	Human toxicity potential
IPCC	Intergovernmental panel on climate change
ISO	International organisation for standardisation
IWK	Indah water konsortium (Malaysia)
kWh	kiloWatt hour
LCA	Life cycle assessment
LCC	Life cycle cost
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
MSTP	Malaysian sewage treatment plant/Malaysian STP
MWTW	Millbrook wastewater treatment work
N	Nitrogen
NEBA	Net environmental benefit analysis
ODP	Ozone layer depletion potential
O&G	Oil and grease
O&M	Operation and maintenance
P	Phosphorus
PAC	Polyaluminum chloride
PE	Population equivalent
PPCPs	Pharmaceuticals and personal care products
RAS	Return activated sludge
RBC	Rotating biological contactor
SBR	Sequencing batch reactor
SETAC	Society of environmental toxicology and chemistry
STP	Sewage treatment plant
TEA	Techno-economic assessment
TEP	Terrestrial ecotoxicity potential
TN	Total nitrogen
TP	Total phosphorus
TSS	Total suspended solids
UKM	University Kebangsaan Malaysia / National University of Malaysia
UNEP	United Nations environment program

USD	United states dollar
WAS	Waste activated sludge
WHO	World health organisation
WLC	Whole life cost
WWTP	Wastewater treatment plant

Chemical/Organic compounds

CO	Carbon Monoxide
CO_2	Carbon dioxide
CH_3OH	Methanol
CH_4	Methane
Cl	Chloride
DCB 1, 4	1, 4-Dichlorobenzene
$FeCl_3$	Iron (III) Chloride
HCl	Hydrochloric acid
H_2SO_4	Sulfuric acid
$MgCL_2$	Magnesium chloride
$Mg(OH)_2$	Magnesium hydroxide
$MgNH_4PO_4$	Struvite
N_2O	Dinitrogen Monoxide / Nitrous oxide
NaOH	Sodium Hydroxide
Na_2S	Sodium sulfide
NH_4^+	Ammonium
NO_{2-}	Nitrite
NO_{3-}	Nitrate
O_2	Oxygen
PO_4^{3-}	Phosphate

Chapter 1 Introduction

1.1 Research context

The anthropogenic activities for economic development cause disasters such as frequent drought and flood, in effect to global warming (World Health Organization, 2007). Without proper planning in the development, damage to the environment is inevitable and as such, the world requires efficient and sustainable development practice to reduce the environmental and climate change impacts. Currently, there are various environmental issues, such as global warming potential, eutrophication potential, acidification potential, ozone layer depletion potential and toxicity potential that are too serious to be ignored. It is related to various impacts through the air, water and soil, which could contribute in many negative effects on the ecosystem. Nevertheless, system activities and their associated impacts are very challenging and difficult to be evaluated, and thus needs an appropriate assessment concept that could analyse the consequences of human activities to the environment. One of them is the operation of wastewater treatment. The European Commission Council Directive (1991/271/EEC) concerning urban wastewater treatment highlighted their aim to protect the environment from adverse effects of discharges from urban and industrial wastewater. It is because wastewater treatment plant is known to be one of an important contributor to many different environmental impacts such as emission of greenhouse gases (GHG), eutrophication by nutrients enrichment in water bodies and ecosystem damage from the emission of heavy metals. In addition, the pressing by more stringent emission limit in the future requires the upgrading treatment of WWTPs for more efficient and sustainable operation. Thus, conducting an environmental impact assessment for particular technologies, products or processes are very important in order to identify their environmental impacts and potential mitigation strategies.

The conventional wastewater treatment could reduce pollutants such as organic matters, solids or nutrients up to 95% (Lee et al., 2010). However, the operational of WWTP requires an investment of resources in the form of energy and chemicals (Heimersson, 2016), and has the potential effect to the environment from the emission to air, water and soil. Due to the various environmental impact risk such as global warming, eutrophication, acidification and ecotoxicity potentials, there is a need for holistic assessment for the operational of WWTP. There are several environmental impact assessment tools that are related to the wastewater treatment such as life cycle assessment (LCA), environmental impact assessment (EIA) and techno-economic assessment (TEA). Among these environmental assessment tools, LCA is known as a tool for the application of holistic/complete environmental assessment of a product or system. In LCA for WWTP, goal and scope, and detail calculations involving inventories and characterisation factors were used to determine the related resource requirements and the environmental emissions. While, interpretation of the LCA results highly depends on the objective, boundary conditions, the quality of data, indicators used and the assumptions made. Even though researches involving LCA for wastewater management is actively ongoing but it is still lacking in the transparency of the database and efficient methodological application (Heimersson, 2016). Hence, more research is required to identify factors and scenarios that affecting environmental impact result from the operation of WWTP, as well as to improve the LCA methodology. Thus, a detail literature review was conducted, focusing on the previous and existing research of different LCA inventories and methodology, and various technologies assessment of the wastewater treatment.

This research focuses on the improvement of life cycle assessment methodology for large centralised wastewater treatment, while investigating the impact by including the comprehensive data inventories related to the wet and dry season, variation of toxic pollutants (e.g. metals and micropollutants), and upgrading of WWTPs to nutrient removal and resource recovery. Micropollutants are antrophogenic origin/chemicals that occur in the aquatic environment with concentrations up to 1 microgram per litre (Stamm et al., 2016). In this study, pharmaceuticals and personal care products (PPCPs) were considered as the assessed micropollutants. The assessment is mainly focusing on the Malaysian context due to the relatively limited works in LCA for wastewater treatment in the developing countries with the tropical weather condition and diluted wastewater, as well as towards the improvement of wastewater management. Millbrook wastewater treatment work in the UK was used for the comparison with Malaysian STP in the seasonal study which representing developed countries. In overall, the research presented in this thesis demonstrates the coverage in the identification of three main areas of concern for the LCA methodology improvement and sustainable operation of WWTPs by conducting comprehensive site-specific life cycle data inventory. The main assessment is including; **1) Assessing environmental impacts of large centralised wastewater treatment plants with combined or separate sewer systems in wet/dry seasons by using LCA; 2) Evaluation of life cycle toxicity assessment methods of municipal wastewater treatment plants with the inclusion of direct emissions of heavy metals and PPCPs and; 3) Upgrading a large and centralised municipal wastewater treatment plant with sequencing batch reactor technology for integrated nutrient removal and phosphorus recovery: environmental and economic life cycle performance.** The scope of works include Malaysian and UK government guidance, international environmental policy, local climate, local wastewater characteristics and industrial practices to see how this context could affect the environmental impact from WWTP.

1.2 Research aims and objectives

The main aim of this research is to assess the life cycle impact of large centralised WWTPs based on the extended and comprehensive local databases for the improvement of LCA methodology and sustainable operation of wastewater treatment. Therefore, this research brings wastewater operation data gathered mainly from the Malaysian context (tropical region), from the United Kingdom (temperate climate), as well as comparison to other developed and developing countries. To the best of my knowledge, it will be the first model in Malaysia focuses on detail life cycle environmental impact, which is including critical analysis involving the seasonal effect, detection of emerging micropollutants from wastewater treatment and upgrading existing WWTP for nutrient removal and resource recovery. The combination of these aspects can provide useful guidance to the local and global management in wastewater treatment involving life cycle impact assessment. In order to achieve the aim, this research focuses on three objectives as described below:

a. To investigate the influence of rainfall on the environmental impacts of WWTPs by using LCA in two scenarios, i.e. large centralised WWTPs with high strength wastewater and low strength wastewater, respectively, but with similar rainfall effects on influent flow rate. Meanwhile, different functional units were studied to evaluate their influence on LCA results in the scenarios with/without rainfall.
b. To assess toxicity impact of PPCPs and metal emissions from a large centralised wastewater treatment plant in Malaysia with low wastewater strength, and to provide useful information for LCA toxicity assessment practice by identifying the importance and contribution of PPCPs and metals and comparing LCIA models.
c. To design upgrading processes based on an existing Malaysian centralised wastewater treatment plant with SBR technology for nutrient removal and resource recovery, and to assess economic burdens and environmental benefits or burdens of upgraded processes with life cycle assessment

1.3 Linkages between the three research chapters

This thesis comprises three result chapters (i.e. Chapters 3, 4 and 5) that each contributes towards the overall aim of this thesis. The linkages between the three chapters and the overall aim of the research are illustrated in **Table 1**. The outline of the three result chapters is presented further.

Table 1. The linkages between the three chapters

Research aim: To assess the life cycle impact of large centralised WWTPs based on the extended and comprehensive local databases for the improvement of LCA methodology and towards sustainable operation of wastewater treatment.

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Chapter 3

Objective - To evaluate the effect of dry and wet season on the environmental impacts from two large WWTPs based on two different functional units, by using local and conventional monthly databases.

Outcome - Electricity consumption, nutrient in effluent, wastewater strength and functional units are the major factors to affect environmental impacts by LCA.

↓

Chapter 4

Objective - To assess toxicity impact of PPCPs and metal emissions from a large centralised wastewater treatment plant in Malaysia with low wastewater strength; and to provide useful information for LCA toxicity assessment practice by identifying the importance and contribution of PPCPs and metals and comparing LCIA models. This study use existing databases in Chapter 3, as well as heavy metals and pharmaceutical and personal care products (PPCPs) in effluent.

Outcome - Electricity consumption, heavy metals, PPCPs and LCIA methods selection are the major factors to affect environmental impacts by LCA.

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Chapter 5

Objective - Life cycle environmental and economic assessment of upgraded processes with nutrient removal and resource recovery in Malaysian STP using existing databases in Chapter 3 and 4, as well as the databases of upgrading of WWTP.

Outcome - Upgrading technology (i.e. three new scenarios/processes A, B and C) build in this chapter were based on the factors affecting environmental impact that were identified in Chapter 3 and 4 such as electricity consumption, nutrient in effluent, heavy metals, and pharmaceutical and personal care products. The upgrading design of Malaysian STP also consider other local factors and constraint such as low strength wastewater and current SBR technology that doesn't include nutrient removal and resource recovery. Thus this chapter is focusing on the improvement analysis of LCA that links with the outcome in Chapter 3 and 4. Since Chapter 3 and 4 identified that electricity consumption is the main contributor to most of the impact categories considered, the upgrading design considers electricity recovery that could reduce the emission of toxic pollutants from fossil fuels electricity production. Process contribution analysis has been provided to identify what processes can be improved to reduce energy consumption. Chapter 3 and 4 also identified that nutrient in effluent as the main contributor to eutrophication, thus upgrading design in Chapter 5 considers nutrient removal in the secondary treatment and phosphorus recovery technology. All three chapters mainly use CML-IA method and two similar functional units for consistent comparison. Chapter 5 also consider life cycle cost comparison between the upgrading WWTP and the existing operation of Malaysian STP (i.e. encompass of MSTP operation in Chapter 3, 4 and 5).

Chapter 3: 'Assessing environmental impacts of large centralised wastewater treatment plants with combined or separate sewer systems in dry/wet seasons by using LCA' was published by Springer Nature in the *Environmental Science and Pollution Research*. This chapter critically assesses the influence of wet and dry season on the environmental impacts of two large centralised wastewater treatment plants located in Malaysia and the UK with low strength and high strength wastewater respectively. Furthermore, this chapter evaluates the influence of two different functional units on LCA result in the scenarios with/without rainfall. The assessment is including various environmental impact issues such as eutrophication, global warming and acidification potentials. Whilst the focus of this chapter is on the seasonal effect with monthly inventory databases (e.g. electricity, transportation, chemicals, direct GHGs and nutrients in effluent), the extended inventory data concerning local toxic pollutants in effluent and sludge are discussed and investigated further in Chapter 4.

Chapter 4: 'Evaluation of life cycle toxicity assessment methods of municipal wastewater treatment plants with the inclusion of direct emissions of heavy metals and PPCPs' has been submitted to *Science of the Total Environment* (currently under review). As asserted in Chapter 3, toxicity impacts were due to indirect emissions such as from electricity and chemical production. To identify the accuracy of toxicity impacts such as to human toxicity and ecotoxicity particularly in low strength wastewater, direct emission from toxic pollutants in effluent and sludge were considered by identifying their impact from Malaysian STP and the result were compared to the toxic pollutants in high strength wastewater in previous studies. Thus, Chapter 4 investigates the toxicity impact of PPCPs and metal emissions from a large centralised wastewater treatment plant in Malaysia, to provide useful information for LCA practice by identifying the importance and contribution of PPCPs and metals when included in the data inventory. In addition, this chapter applied model comparison between different LCIA methods (e.g. CML-IA, Recipe Midpoint, IMPACT2002+, EDIP 2003 and USEtox) to compare results of human toxicity and ecotoxicity impacts when including direct toxic pollutants. Chapter 3 and Chapter 4 present environmental impact from the existing operation of the wastewater treatment plant. Nevertheless, with the consideration of more stringent regulation in the future in Malaysian situation as a developing country, upgrading the existing plant to nutrient removal and resource recovery could be the optimal option for the efficiency of the existing local WWTP's operation, which is considered in Chapter 5.

Chapter 5: 'Upgrading a large and centralised municipal wastewater treatment plant with sequencing batch reactor technology for integrated nutrient removal and phosphorus recovery: environmental and economic life cycle performance' was published by Elsevier in the *Science of the Total Environment*. Considering future stringent regulations in the developing countries, this chapter focuses on the design of three upgrade processes with nutrient removal and resource recovery based on the existing Malaysian STP, as well as the application of LCA to assess the environmental impact from the new upgraded processes. With the data of MSTP used in Chapter 3 and Chapter 4, wastewater treatment with nutrient removal and resource recovery were designed as process A, B and C to further assess their environmental impact and benefit towards sustainable application. In addition, the economic assessment of the newly designed treatment is presented to identify the economical and beneficial option. This research is also focusing on the effect of energy consumption to the hotspot identification, and the selection of three different functional units (i.e. per m^3 of treated wastewater, per 1kg PO_4^{3}-eq. and per 1 kg struvite recovered).

Besides presenting methodology improvement based on the comprehensive databases in various scenarios, the results presented in this thesis is useful to stakeholders for the efficient and sustainable strategies of wastewater management both in developing and developed countries.

Chapter 2 Literature review

Chapter 2 presents the literature review for this research. First, Section 2.1 discusses in detail about wastewater treatment system, the life cycle assessment method, and LCA application in wastewater treatment including benefits and problems. Next, Sections 2.2, 2.3, and 2.4 review research regarding the seasonal effect of WWTPs on LCA, the toxicity impacts from WWTPs by LCA, and the application of LCA to select/design sustainable WWTPs, respectively. These sections are related to the three main result chapters in this thesis. Then, Section 2.5 discusses the situation in Malaysian context regarding wastewater treatment. Finally, Section 2.6 explains the knowledge gaps identified from the overall review.

2.1 Life cycle assessment and its application in wastewater treatment

2.1.1 Introduction of municipal wastewater treatment system

Municipal wastewater treatment system is encompassed of sewer system and wastewater treatment plant (WWTP). There are two types of sewer system connected to WWTP: i) separate sewer system that has separate flow/network for rainwater runoff and domestic/industrial wastewater, ii) combined sewer system of the same sewer pipe for both rainwater runoff and domestic/industrial wastewater. A WWTP is consist of different processes/operating units (e.g. pre-treatment, primary treatment, secondary treatment, sludge treatment and tertiary treatment). Pre-treatment and primary treatment are mainly focused on removal of particulate pollutants such as solids, grit and greases. Secondary treatment treats organic matter, nitrogen and phosphorus that contained in the sewage, through biological and chemical processes (Hao et al., 2019). Tertiary treatment is applied to remove remaining small particles and pathogens in some of WWTPs. Finally, sludge treatment treats the excess sludge for stabilisation and volume reduction through sludge thickening and dewatering. The sludge is sent to landfill, used in agriculture, incinerated or transported to composting plant. Meanwhile, constructing a WWTP is a challenging process that implicates various types of materials such as concrete, timber, steel and plastics, and involves detail operational design/equipment. The operation of WWTP requires: i) large amount of electricity for pumping/aeration, ii) chemicals for sludge treatment and phosphorus removal and, iii) transportation of waste, sludge and chemicals (Morera et al., 2016). Consequently, WWTP has substantial environmental impacts during its life cycle (i.e. construction, operation and demolition) due to the energy consumption, chemical usage, sludge generation, effluent discharge and gas emissions (Piao et al., 2015).

2.1.2 The state-of-the-art methods for applying LCA in wastewater treatment plants

Over the last 50 years, an increased awareness has developed in global society about protecting the environment particularly water resources. In relation to that, the European Commission Council Directive (1991/271/EEC) concerning urban wastewater treatment stated that the objective of wastewater treatment is to protect the environment from adverse effects of discharging urban and industrial wastewater. A large number of wastewater treatment plants (WWTPs) are designed and operated to prevent pollution to the environment by removing a variety of contaminants from wastewater before discharge. However, to some extent, the pollutants in wastewater could be transferred to air such as greenhouse gas (GHG) emission and soil such as disposal of sludge due to wastewater treatment, which could lead to negative effects on human health and the environment in other forms. This holistic environmental impact from WWTPs is very challenging to evaluate, and thus, a cradle-to-grave approach is needed to analyse the consequences of these plants to the environment. Some environmental impacts from the operation of WWTPs include climate change from the emission of GHGs, eutrophication from the emission of nutrients to the water body, and ecosystem damage from the emission of heavy metals. Therefore, conducting an environmental impact assessment for particular technologies, products, or processes is very important to identify their environmental impacts and potential mitigation strategies.

The application of environmental assessment tools provides reliable environmental impact information that assists in decision-making toward sustainable operation of a system/process. At present, the impact of a wastewater treatment system can be assessed through different evaluation tools such as the life cycle assessment (LCA) method, economic and exergy analysis (Muga & Mihelcic, 2008), the environmental impact assessment (EIA) method, and net environmental benefit analysis (NEBA). LCA is an approach or method in assessing the environmental impacts that associated with all the stages in life cycle of commercial products, processes or services. In LCA, environmental impacts are assessed from raw material extraction of the product/process to the final disposal of the materials, i.e cradle to grave (https://en.wikipedia.org/wiki/Life-cycle_assessment). The use of LCA has started to draw great attention by many researchers or industrial practice in identifying the environmental impacts and to evaluate the sustainability of wastewater treatment/technology selection (Pasqualino et al., 2009; Meneses et al., 2015; Piao et al., 2015). This is because LCA provides a complete framework of assessment starting from the goal and scope (objective), life cycle inventory, life cycle impact assessment and interpretation. Meanwhile, an economic analysis could be assessed using cost-benefit analysis (CBA) (Lavee, 2011; Liang & van Dijk., 2012), life cycle costing (LCC) (Rawal & Duggal., 2016), and techno-economic analysis (TEA). Usually, this economic evaluation can be combined with LCA to produce a robust evaluation for a system-level analysis towards the sustainable operation of WWTPs. Due to the suitability and wide used of LCA for WWTPs, this literature review focuses on the detailed application of LCA for WWTPs to identify the potential of the LCA method from wider aspects.

Compared with LCA analysis for the manufacturing sector, an LCA of wastewater treatment is relatively new with about 20 years of practice. Since 1995, more than 60 international peer-reviewed articles dealing with WWT and LCA have been published with different inventories, boundary conditions, functional units, and impact assessment methods. The various research have shown that LCA has evolved in these past two decades to include more improvement and systematic assessment. An extensive review of existing LCA studies was conducted for this research to assess the state-of-the-art knowledge on the environmental impact and benefit of LCA to identify the knowledge gap. Based on the selection of related journals, 50 published articles related to WWT and LCA from the international scientific journals were reviewed, and 65% of the papers from 2006 to 2019 were summarised, the results of which are shown in a table in **Table 5**. A life cycle assessment is an approach that considers environmental, economic, and social impacts that a product or service will produce throughout its life cycle. It can be used as a technical tool to identify opportunities to reduce the environmental effects associated with a specific product, system, material, or activity. LCA has been applied in various research settings to analyse the environmental impacts of different WWTPs including industrial and municipal facilities. However, the scope of assessment is rather challenging due to the variation in defining the system boundaries and the difficulty in considering wastewater composition and the type of pollutants. Different options of wastewater treatment technology have different performance and different impacts on the environment, which may take place from different phases in a WWTP's life cycle. In the following overview, relevant studies within this field of research are briefly described mainly to generate a benchmark of LCA methodology application in the wastewater treatment field.

Based on the detailed review (as in **Table 5**), published research on the LCA of wastewater treatment can be classified into two types. One type is focused on using LCA to facilitate technology comparison and selection from the environmental impact point of view. The other type is focused on working on different steps of the LCA method itself (i.e, goal and scope, inventory, impact assessment and interpretation) to improve the reliability of the LCA results. Some researchers have even developed new models for the calculation of new characterisation factors or new impact categories, such as a new characterisation factor for pollutants or substances to provide more representative and reliable analysis. For instance, one study conducted an environmental evaluation of common technical options for urban wastewater treatment (Hospido et al., 2008), whereas another identified the overall environmental impact of WWTPs (for both water and sludge treatment) using LCA methodology (Pasqualino et al., 2009). Some studies have also conducted specific evaluations of GHG emissions from WWTPs (Gupta & Singh., 2012; Daelman et al., 2012; Corominas et al., 2012), including one environmental-economic evaluation of the sludge treatments process in Korea that used life cycle analysis (Piao et al., 2015). In more detail, LCA methodology was also used by Ontiveros & Campanella (2013) to evaluate the environmental performance of three different advanced biological nutrient removal processes in Argentina: modified UCT, five stages bardenpho, and modified bardenpho from WWTPs. This evaluation can guide the selection of the biological nutrient removal process in the Argentina context from both technological and environmental points of views. In a different aspect, Yoshida et al. (2014) conducted a study on the improvement of life cycle inventory and methodology involving the consideration of onsite GHG emissions and long-term emission data in the land application of sewage sludge. In addition, Morera et al. (2016) worked on the improvement of LCA methodology in the urban wastewater treatment system and emphasized the improvement of construction detail inventory including sewer system and inventory improvement with scale assessment. Recent research identified that most studies using LCA for WWTPs aim to evaluate the environmental impact of different technologies including identifying advanced and conventional emission parameters (Lorenzo-Toja et al., 2016; Alfonsín et al., 2016), analysing control strategies of WWTP performance (Meneses et al., 2015), and identifying the environmental trade-off of different process alternatives. These findings showed that LCA can assess various aspects of identifying environmental impact from wastewater treatment but that the methodology from the provided framework by the International Standard Organisation (ISO) could be further improved. The social factor is more complicated and is not included in this review.

Review of 50 published LCA studies of WWTPs (from 2006 to 2019) identified that most of the published studies have been concentrated in Europe, the USA, and Australia with little application in developing countries such as India, Thailand, and Malaysia. In terms of technology coverage, only a few studies have applied LCA to resource recovery, especially in developing countries. The analysis in these 50 papers also revealed that, there is variability in the definition of the functional unit and the system boundaries, the selection of the life cycle inventory and impact assessment methodology, and the procedure results interpretation. As supported by Hauschild et al. (2013), the LCA standard of ISO 14040 is still general and unspecific in its requirement. Therefore, there is a need to investigate and develop standardized guidelines for the wastewater treatment operation by evaluating the key steps in the LCA methodology to improve the quality of LCA-WWTP.

2.1.3 Key steps for LCA assessment

LCA is a standardised methodology to evaluate the environmental impacts associated with a product or process during its complete life cycle as described in two ISO norms (ISO 14040 and 14044). The concept of life cycle assessment first emerged in the late 1960s. In the 1970s, LCA only focused on energy and raw materials, but later the analysis system expanded to emissions, water, air, and soil. Starting in the 1990s, LCA was applied in wastewater treatment after being identified as suitable for related environmental assessments. In 1994, the ISO began developing standards for the LCA method as part of its 14000 series on environmental management; however, the method was not yet designed in detail for all fields of assessment (Corominas et al., 2013). Nevertheless, since then more studies on LCA have been undertaken and published in various disciplines, which included a variety of boundary conditions, databases, impact assessment methods, and interpretations.

Several software have been developed including free and commercial software to assist in the analysis of LCA. At present, various types of commercial LCA software are available such as SimaPro (El-Sayed et al., 2010), Gabi7 (Tomei et al., 2016), and Umberto. SimaPro was developed by Pre-Consultants in the Netherlands and has been used for more than 20 years in various studies and projects. It is a user-friendly tool that helps to model and analyse complex products or systems such as water and wastewater treatment. It can also calculate environmental impacts and detect environmental hotspots in a systematic way (Morera et al., 2016). In addition, OpenLCA that was developed in Germany is another free software for LCA user (Ontiveros & Campanella, 2013). All of these softwares are professional life cycle modelling tool and available with various embedded databases such as Ecoinvent, European Life Cycle Database (ELCD) and U.S. Life Cycle Inventory (USLCI). However, one of the challenges of LCA is that it requires detailed inventory information for each system assessed (Balkema et al., 2002). Previous studies have also identified inconsistencies in the selection of pollutant coverage and the quantification of emission pathway because LCA only provides framework methodology, which is mainly for production and not for process treatment. Thus, a detailed review on LCA methodology steps was conducted to understand more about the application of LCA. The structured methodology in LCA as stated in ISO starts with defining the goal and scope followed by life cycle inventory (LCI), life cycle impact assessment (LCIA) and ends with a results interpretation as shown in **Table 2**. This methodology highlights the general steps of a LCA with general characteristics that have been identified within each step.

Table 2. LCA methodology steps for environmental impact assessment from WWTPs

Goal and Scope	Life Cycle Inventory	Life Cycle Impact Assessment	Interpretation
Objective	Input data (e.g. influent, energy and chemical consumption)	Classification (e.g. eutrophication, global warming, acidification, ozone depletion, human toxicity, freshwater ecotoxicity and resource depletion potentials in midpoint impacts	Comparison of impact analysis
System boundary	Output data (e.g. emission to air, water and soil)	Methodology selection (e.g. CML-IA, EDIP,IMPACT 2002+, eco-indicator99, Recipe and USEtox)	LCA method evaluation
Functional Unit (e.g. 1 m^3 of wastewater)			Data quality/sensitivity analysis
			Normalisation and weighting (optional)

2.1.3.1 Goal and scope

In detail, the goal and scope of LCA consist of the objectives, system boundary, and functional unit. The objectives consist of the environmental analysis, the technology comparison, and their effect on the environment or the analysis of life cycle inventory and methodology to various impact categories. The system boundary determines which unit process shall be included in LCA analysis (ISO, 2006) such as construction stage, operation, sludge treatment and disposal, and demolition phase. As shown in **Table 5**, 85% of the studies merely covered the operational phase because it contributes to the highest total environmental impact (El-Sayed et al., 2010; Zang et al., 2015; Meneses et al., 2015). Lorenzo-Toja et al. (2016) reported that the environmental impact from the construction phase is almost negligible for many impact categories compared with the operational phase. Similarly, Pasqualino et al. (2009) stated that the environmental impact from the construction and demolition phases also could be considered negligible. In the operational phase, approximately 80% of the studies included sludge treatment and disposal in the system boundary due to the importance of this stage to the overall impact (Corominas et al., 2013). Finally, the functional unit is usually defined as the treatment of a volume of wastewater in 1 m^3; however, some studies have used population equivalents (PE/year). In addition, several other options are available for the functional unit in LCA-WWTP such as the quantity of sludge produced and the quantity of removed pollutants (Hospido et al., 2008). However, no strong justification appears to exist between its selection and technologies used in a specific system of WWT. To analyse this issue, Rodriguez-Garcia et al. (2011) studied the effect of a functional unit based on wastewater volume (m^3) to identify the different effluent quality of six typologies of WWTPs. They found that global warming and economic cost decrease following better eutrophication. By contrast, studies with similar FU found that a trade-off exists between lower eutrophication and higher environmental and economic impact when involving more demanding/upgrading treatment such as water reuse. These conflicting results show that discrepancies still exist when using single FU to identify the effect of different treatment technologies. Therefore, Rodriguez-Garcia et al. (2011) have suggested that a second FU should be introduced in specific studies such as those on eutrophication reduction (kg PO_4^{3-}) to overcome this limitation and strengthen the system under study. This suggestion was supported by Corominas et al. (2013) who determined that a FU of a system could influence the final result, especially when comparing WWTPs with different influent qualities or different removal rates.

2.1.3.2 Life cycle inventory

After the goal and scope are determined, the second step in a LCA is data collection and inventory build-up, a crucial stage when performing an LCA study. In general, LCI aims to identify the inputs (resources), the outputs (effluent and waste), and the respective amount of emissions over the entire life cycle of the specific process. Generally, it is given in physical units such as kilogram (kg), cubic metre (m^3) and kilowatt-hour (kWh). Wastewater treatment data inventory includes the foreground as the primary data (operation), which is usually compiled from the operational record, detailed design document, sampling works, and vendor-supplied information. By contrast, background data (secondary input) such as energy production and chemical production, are normally provided by the LCI database (e.g. the Ecoinvent (Corominas et al., 2013) and the ELCD). Ecoinvent, which was developed by the Ecoinvent Centre in Switzerland, is one of the major data inventory providers used in various sectors. In the LCI phase, identified inventories are collected for all processes of the boundary and calculated to the same functional unit.

2.1.3.3 Life cycle impact assessment

Prior to the calculation of the environmental impacts, the assessment methodology must be selected to give direction to the category of impact required, such as the midpoint level or the endpoint level. Several different methodologies are available to identify related impact categories in the LCA such as Eco-Indicator99 (El-Sayed et al., 2010), Recipe (Zang et al., 2015), EDIP 2003, USEtox, IMPACT2002+, and CML2001 (Kalbar et al., 2013). CML2001 is found to be the highest number in the methodology used by researchers due to its extensive impact categories, high relevance to wastewater treatment at the midpoint level, and accurate results as shown from a previous study (Pasqualino et al., 2009). For the life cycle impact assessment (LCIA) phase in every methods, the data in a specific FU from the LCI are multiplied with their characterisation factor (CF) to convert to environmental impacts in various categories. Characterisation factors (CFs) of pollutants are provided to practitioners either in the literature or by the software used (Muñoz et al., 2008). CF models are built based on the mechanism of the cause-effect chain starting from the emission of pollutants until the receiving compartments. CF values are the total results of environmental fate, exposure and the resulting effect on the exposed section such as human (Huijbregts et al., 2005). CFs were calculated by multiplying fate factor (FF) to exposure factor (XF) and effect factor (EF). Fate factor (FF) denotes the residence time of the substances/pollutants in the receiving compartments. Exposure factor (XF) relates to the actual concentration of substances taken by receiving compartment, e.g. human. Effect factor (EF) is correlated to the route of exposure, e.g. ingestion and inhalation effect to human toxicity. Exposure factor and fate factor are combined to form intake factor (IF) of a substance (Rosenbaum et al., 2008).

Nevertheless, various discrepancies still exist between these methods provided in LCA. To address this issue, Pizzol et al. (2011) compared nine different methodologies focusing on the impact of metals on human health. The results showed a poor agreement between the methods. For example, the contribution of metal to total human health changed greatly between the methods. This poor agreement is due to the different types of metal considered and the different techniques used to calculate the characterisation factor. This indicates that there is no unified LCIA method, especially for the human health impact category. **Table 3** lists the origin or provider of each methodology provided in LCA.

Table 3. List of environmental impact assessment methods

Method	Developer
CML 1992/CML-IA	Centre for Environmental Studies, University of Leiden
Eco-indicator 95/99	Pre Consultant B.V
Eco-points 97	Swiss ministry of the environment
EDIP 2003	Institute for product development (IPU) at the Technical University of Denmark
IMPACT 2002+	Swiss Federal Institute of Technology Lausanne (EPFL), Switzerland
Recipe	Pre Consultants, Radbound Universitteit Nijmegen and CE Delft

Midpoint environmental impact categories are provided in each method. For example, in CML-IA, the midpoint categories involving wastewater treatment normally include abiotic depletion (fossil fuel), eutrophication, global warming, acidification, ozone depletion, and human toxicity potentials. However, water, land, and energy use have been increasingly gaining attention in this research area as new impact categories, depending on the objective of the study. In contrast to midpoint categories, the endpoint damage category is always considered in the LCA assessment as an endpoint area of protection. The categories include damages to human health, ecosystems, and resource availability.

2.1.3.4 Interpretation

The final stage of LCA methodology is interpretation. This final stage can identify and evaluate information from the result of the life cycle impact assessment because it can determine the level of confidence in the final results. It starts with an understanding of the accuracy of the result and how it meets the goal of the study. According to Corominas et al. (2013) and based on the ISO 14040:2006, the interpretation part in the LCA includes; (a) identification of important issues based on the results of the LCI and LCIA; (b) evaluation of the study considering completeness, sensitivity, and consistency checks; and (c) conclusion, limitations, and recommendations.

2.1.4 Geographical relevance of LCA for WWTPs

Before the 2000s, the majority of the traditional LCA approach was based on site-independence where no consideration was given to geographical and temporal factors. The reviewed study showed that some published papers used secondary data (e.g., from literature) or simulated data to conduct LCA analysis due to the lack of the available primary data, leading to much less reliable results. However, the results still could provide some guidance to a certain degree. For the inventory practice, approximately 55% of studies for LCA-WWTP were based on site operation while others still depended on the estimations, existing simulation data, previous reports, and literature due to the limited availability of reliable databases. The other reason was that performing onsite measurements that obviously can reduce the data uncertainty is often not feasible as it is expensive and time-consuming.

The analysis of the geographical distribution in LCA found that only a few studies were conducted in developing countries such as India, Egypt, Thailand and China. As a consequence, the distributions of studies with regard to the assessed wastewater management systems by LCA on environmental concerns are specific to a few regions only. The drawback of this analysis system is that another country in a different region with a different temperature or economic value is not suitable to refer to the existing available data and impact results. This situation shows that the fairly distributed databases around the world are still lacking in LCA analysis studies for WWTPs, especially in developing countries. This idea was supported by Renou et al. (2007) who reported that location-specific factors are critical especially for eutrophication and terrestrial ecotoxicity impact category due to the transportation effect by pollutants. Therefore, the selection of inventory data is critical to LCA analysis especially when local factors are accounted for to provide reliable results.

To overcome this limitation, there was a trend after 2000 towards making LCA more site-dependent that considered more site-specific characterization factors such as eutrophication, toxicity impact, and acidification potentials. This is because the point of emission may have a strong impact on these regional and local impact categories. For global warming and ozone layer depletion, characterization factors are justifiable because the emission location has no influence on the transportation effect (Gallego et al., 2010). Therefore, it is important to identify specific characterisation factors that impact the countries that have different geographical, climatic, and economic factors, which are significantly lacking in developing countries. This brings into question how the importance of regionalisation criteria and the database will influence the LCA results. Therefore, there is still some possibility that the LCA method for wastewater treatment impact assessments can be improved, especially outside of Europe with consideration for the variability of treatment technology. For example, Yoshida et al. (2014) studied the effects of three different inventory databases to the LCA results that are from the European Pollution Release and Transfer Registry (EPRTR), the Denmark national discharge limit data and data collection scheme conducted at the WWTP in Copenhagen, Denmark. They found that the LCA results depend heavily on the onsite data input. For instance, the EPRTR did not capture impact for particulate matter and terrestrial eutrophication. They found that primary data (i.e. site data collection scheme) from WWTPs gave the most reliable LCA results but still needed some improvements, such as the expansion of substance coverage and additional detail collection of energy and chemical usage. On the other hand, for the temporal effect, even though (Lorenzo-Toja et al., 2016; Alfonsín et al., 2016) identified no clear difference in environmental performance between WWTPs from the Atlantic and Mediterranean regions of Spain, the effectiveness of using the existing secondary databases to different region especially in a different climate of developing country is uncertain. Therefore, it is well proved that site-specific inventory data is the key to obtain reliable LCA results.

The above review shows that research focused on specific local conditions and inventory effects to the LCA results have been rarely assessed in LCA-WWTP related studies. Most of the studies also did not stress the importance of geographically different impacts in terms of data inventory and local factors (e.g. temperature and rainfall). In fact, some of the research outside of Europe uses European datasets to its region without adjusting for uncertain information such as the local impact factors of electricity. One of these factors is the availability of generic database, which decreases the need for the importance of local primary data. Furthermore, most of the characterisation and normalisation factors are also based on European conditions, where these factors are currently used globally only due to its availability. However, very few studies have been conducted using LCA in developing countries. The lack of primary data and underrepresentation of the life cycle thinking concepts in developing countries are possibly the main causes for the restricted number of studies published. In the wastewater sector, besides energy and chemical production data, the most important aspects are the effects of temperature, rainfall intensity, local pollutants, and design criteria (e.g. combined or separate sewer systems), all of which could be included and analysed. Moreover, the impact of treatment technology is greatly dependent on the local situation/factors such as geographic location, wastewater characteristic, energy type and source, choices of sludge and waste disposal options, and size of markets for products derived from WWT system such as fertiliser.

Therefore, it is important to have inputs based on a localised primary and secondary database with regard to a local characteristic that is representative specific region such as tropical developing regions or Europe. In other words, the new localised database can keep the commercial data inventory as a benchmark. **Figure 1** shows that the difference in climate is clear between continents.

For example, Europe has a temperate climate (i.e. warm in summer and cold in winter) while tropical zones having warm weather year-round. Indeed, regionalisation is recognised as an important step towards improving the accuracy, precision, and confidence in LCA results.

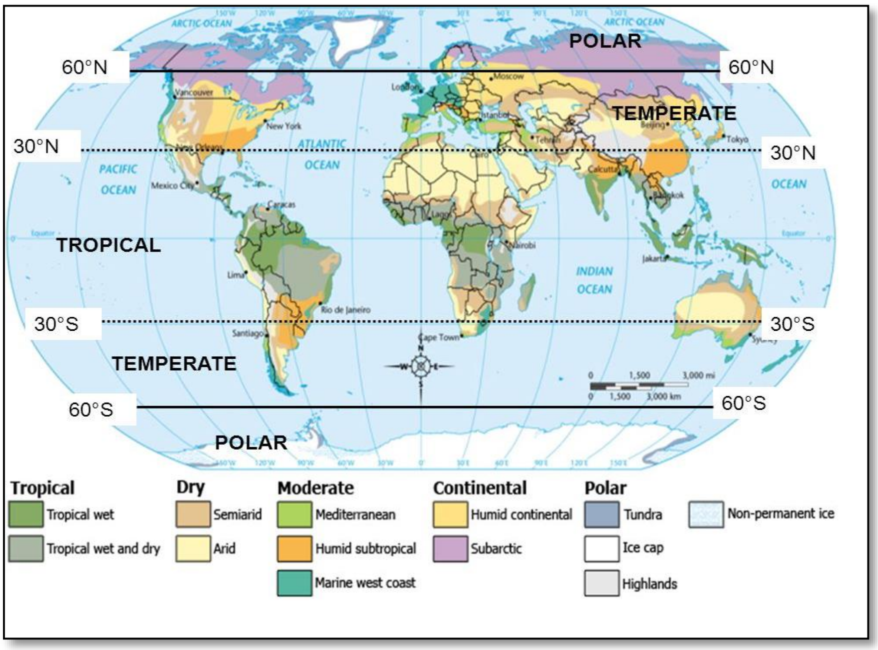


Figure 1. Worldwide climate classification, from (https://simple.wikipedia.org/wiki/Climate)

2.1.5 Benefits of LCA for environmental impact evaluation of WWTPs

The current LCA is well described in terms of the framework and can be applied to a wide range of products including waste and water cycles. Therefore, in this situation, LCA could be a tool to identify environmental factors and assess impacts from the wastewater treatment operation (Ontiveros & Campanella, 2013; Meneses et al., 2015). Furthermore, Foley et al. (2010) pointed out that while their research had provided new inventory data needed about WWTPs, without life cycle impact assessment modelling they could only identify a limited comparison for the impacts by the newly provided data. Besides identifying the environmental impacts from WWTPs, LCA can assess the trade-off of the new integration of existing technologies in terms of cost and environmental impact (Mayer et al., 2016). For example, Meneses et al. (2015) concluded that the technologies adopted for more stringent effluent standards from WWTPs (i.e. 10-15 mgN/L and 1-2mgP/L by EU Urban Waste Water Directive) could improve effluent quality but, at the same time, may require additional energy consumption, use chemical reagents, and produce more sludge. Hauck et al. (2016) found the trade-off between different environmental impacts by conducting a life cycle assessment. They reported a 16% reduction in marine eutrophication, but the climate change impacts increased with 9% from the traditional operation of the Dokhaven Wastewater Treatment Plant in the Netherlands. This increase was due to the increasing use of electricity and shows that the trade-off between effluent quality and other environmental impacts should not be neglected when applying advanced technology. In nutrient recovery, a similar phenomenon was observed. For example, struvite precipitation for phosphorus recovery improved the effluent quality from WWT while recovering nutrient resources. However, the chemical addition for pH control accounted for up to 97% of total struvite cost (Doyle & Parsons, 2002). Thus, by applying an established methodology, LCA can identify the trade-off of different technologies adopted in WWTPs.

Besides its benefits, LCA still has a series of shortcomings and limitations, especially related to the data quality and methodology choice. Therefore, research is needed to provide recommendations to future LCA practitioners on the suitable data requirement and impact assessment methodology for WWT. To evaluate these limitations, rigorous assessments should be considered especially from various aspects of the life cycle assessment (LCA) to wastewater treatment to identify the most significant environmental issues, including the economic effects. Thus, a detailed literature review regarding several issues such as seasonal effect, toxicity impact, and sustainable wastewater treatment to LCA was conducted to further guide the research of this study.

2.2 Seasonal effects of WWTPs on LCA

2.2.1 Inventory data in WWTPs for seasonal study

It is a normal practice worldwide that storm runoff is combined with domestic wastewater through a combined sewer system for treatment. The combination of untreated wastewater and the storm runoff (i.e from roadside or run-off from agricultural land) could overload WWTPs during storm weather. Thus, the extra volume of water overflows directly into the receiving waters without treatment, influencing the environment negatively. Combined sewer overflow (CSO) containing untreated wastewater due to a certain design limit (e.g. >6 DWF discharge directly to nearby stream) results in major water pollution. Risch et al. (2018) stated that loads from untreated storm water are important contributors to eutrophication and ecotoxicity in freshwater. Eutrophication impact by CSO are depending on the concentration value of organic matters, nitrogen and phosphorus in the stormwater and the untreated wastewater. Water flowrates and wastewater characteristics are closely related to rainfall (Mines et al., 2007a) and whether the sewer collection system is separated or combined with stormwater. Infiltration is another source of water flow into the collection system. The amount of flow that can enter a collection system from groundwater or infiltration may range from 0.01 to 1.0 m^3/d.mm.km (Metcalf & Eddy, 2004). This was supported by a study by Pasqualino et al. (2009) who found that the quantity and quality of influent wastewater are varied according to the number of people served, differences in lifestyle, and seasonal variations in the weather. In fact, Renou et al. (2007) compared the results of different wastewater characteristic to various impact categories using LCA. They found that approximately 20% variation in wastewater flow characteristics is due to rain events and the type of wastewater collection, both of which could generate variation in the characteristics of substance flow and subsequently affect the difference in environmental impact. Research by Mines et al. (2007b) demonstrated that flows in WWTPs increase as rainfall intensity increases in the combined sewer system. On the other hand, a separate sewer system has a lower influence of rainfall on the volume of WWTPs than the combined sewer system.

In addition, the concentration and load of wastewater in WWTPs that change with the variation of rainfall intensity could affect wastewater treatment performance, as well as the quality of discharged effluent to the environment. For example, biological nutrient removal is usually better in summer than the wet/winter season due to higher microbial activity at a higher temperature. The characteristics of the influent that are affected by rainfall and temperature can be the driving factors affecting the efficiency of WWTPs and consequently affects pollutants level in the effluent (Lorenzo-Toja et al., 2015). Moderate to strong correlations were identified concerning rainfall intensity and pollutant concentrations in the influent, and rainfall intensity and volumetric flow rate to WWTPs, at 24 WWTPs in Georgia, USA with combined sewer systems (Mines et al., 2007a). The squared correlation coefficient (R^2) between flow rate and the average monthly rainfall ranging from 0.21 to 0.85 indicates that different WWTPs in different catchment areas with combined sewer systems were affected by rainfall intensity to different extents (Mines et al., 2007b). It is believed that strength wastewater in the dry season usually achieve satisfactory levels of pollutant removal, while diluted influent by stormwater during wet season is likely to cause operational issues (Lorenzo-Toja et al., 2015; Risch et al., 2018), leading to lower treatment efficiency. Nevertheless, lower pollutant concentrations in effluent of WWTP were reported during wet weather due to the dilution which could reduce the eutrophication level (Joel, 2017; Li et al., 2018).

Since the early 1990s, the application of LCA methodology in wastewater treatment has still been in progress especially with associated environmental impact categories such as toxicity-related impacts, temporal assessment (Shimako et al., 2018), energy balance, and water and land use (Zang et al., 2015). In a study on the effect of wastewater treatment (WWT) on the environment, many variables simultaneously change with time and location. By measuring and analysing the significance of each parameter related to time and seasonal change, it may be possible to understand their relationship and assess the sustainability of the system. Currently, many LCA studies evaluate the environmental impact of WWTPs using one dataset/static LCA analysis (Halleux et al., 2006; Foley et al., 2010; Rodriguez-Garcia et al., 2014). However, municipal wastewater treatment is sensitive to time-related processes because wastewater flowrates to WWTPs, wastewater characteristics, and biological removal efficiency especially for nutrient removals such as nitrogen and phosphorus are time dependent (Skoczko et al., 2017). Many of LCA studies of WWTPs are based on dry weather conditions or one set database without considering rainfall effects. This results to less holistic assessment for the whole year with temporal variability in identifying environmental burdens from WWTPs. This temporal assessment is especially significant to the vulnerable receiving waters because environmental impact assessment during dry season only may overestimate or underestimate the environmental burdens such as eutrophication and freshwater ecotoxicity impacts. This literature review (as in **Table 5**) found that more than 90% of the LCA-WWTP studies were based on one set of influent data and one set of effluent data (Halleux et al., 2006; Foley et al., 2010 ; Rodriguez-Garcia et al., 2014). This conventional approach of LCA for WWTPs was focused on the removal efficiency of organic load and nutrients (e.g., biochemical oxygen demand, chemical oxygen demand, nitrogen, and phosphorus) and the effects from energy and chemical consumption (Lorenzo-Toja et al., 2016). The aim was to provide only one set of LCI that included input, output, and emissions to identify the differences of impact categories in LCA such as eutrophication potential, global warming potential, and acidification potentials. Most of the studies investigated about the one-time impact of sewage pollutants on the ecosystem (Halleux et al., 2006; Foley et al., 2010; Rodriguez Garcia et al., 2014) but did not consider other factors such as the difference in wastewater composition between various seasons or times, which could affect the variance of environmental impacts in LCA. According to Mines et al. (2007), wastewater flows are seasonally variable especially to precipitation, which could result in different flow conditions. This aspect is still poorly quantified and considered in LCA methodology for WWTPs. To be more specific, existing practice has ignored the fluctuation of influent characterisation over the season (Joel et al., 2017), a variation of treatment operation due to a different temperature and the corresponding effluent quality. For example, it is very sensible to expect that the eutrophication category is more serious in cold season at a low temperature due to lower nitrification rates and lessened in warm season at a higher temperature when a midpoint level assessment is conducted. These factors indicate that it is important to conduct an LCA based on the time relevant cases. However, LCA studies to wastewater treatment have had problems with data quality related to life cycle inventory (Flores-alsina et al., 2013) because the life cycle evaluation is not straightforward due to the spatial and time-related issues. For instance, the quantity and quality of wastewater treated vary according to the plant capacity, cultural and economic factors, environmental regulation requirements, technology implementation, and seasonal variations in the weather (Meneses et al., 2015).

Since 2010, some researchers have tried to develop a dynamic LCA method with consideration to time (Li et al., 2017). However, many parameters can influence the temporal profile of a dynamic LCA result, resulting in difficulty to apply LCA for wastewater treatment. In addition, no consensus exists about the dynamic LCA method to use. In some situations, temporal change is relatively much easier. Thus, it could be possible to pursue a season-based LCA method instead of daily-based dynamic LCA analysis to overcome constraints of both dynamic and static methods. For example, in some tropical areas such as Malaysia and Singapore, annual temperature is almost constant. Due to high rainfall precipitation in these areas, separate sewer systems are mainly adopted to mitigate flooding. In seasonal assessment (e.g. summer and winter; dry and wet), temperature and rainfall are inevitably intertwined. A few seasonal studies have been conducted for combined sewer systems such as winter and summer by Lorenzo-Toja et al., 2016; Alfonsín et al., 2016; humid and dry by Moreira et al., 2004; and dry and wet by Li et al., 2017. Moreira et al. (2004) highlighted that environmental analysis for a Spanish municipal WWTP between wet(rainfall) and dry was not essential because the data variability in every seasons in Spain is more significant than the variation affected by rainfall. Lorenzo-Toja et al. (2016), however, found that Atlantic region with the highest rainfall intensity achieved low life cycle environmental impact in the assessment for several regions in Spain with different rainfall intensity (i.e. from 300mm to >1000mm). A recent study by Li et al. (2017) conducted research on the influence of rainfall on the effect of WWTPs with combined sewer system in China. All environmental impact categories studied (e.g. abiotic depletion potential (ADP), acidification potential (AP), eutrophication potential (EP), global warming potential (GWP) and photochemical ozone creation potential (POCP) show a rising trend when monthly rainfall intensity below 193.2 mm, and then achieved a stable state. One of the reason for this result is, higher mass load of BOD and total phosphorus in wastewater during the rainy season leading to higher energy consumption. In addition, the sewer system affects wastewater characteristics and treatment. However, these factors were not considered in the literature. A combined sewer system carries the stormwater from the roadside and, in the worst case, carries pollution from agricultural land, resulting in higher nutrient levels in wastewater. Additionally, it involves higher energy consumption due to the increased volume of wastewater to be treated, resulting in increased environmental burden. Risch et al. (2017) identified that wet weather contributes up to 62% of the total impact on freshwater ecotoxicity using a combined sewer system. In winter, WWTPs have a higher power consumption and produce less energy if an energy recovery system provided. Thus, to identify the difference in dry and wet seasonal impacts that could improve the LCA methodology, it is important to provide a critical analysis involving flowrate variation due to rainfall events and the associated life cycle inventories. Nevertheless, most LCA studies about the influence of rainfall on WWTPs have focused more on the combined sewer system and less on the separate sewer system. The disadvantage of the combined sewer system is that during heavy rainfall, the overflow containing various pollutants can impair the quality of the water body, causing a seasonality effect. In the worst situation during a period of heavy rainfall, the wastewater volume in combined sewer system can surpass the capacity of the wastewater treatment plant. The real burden of the combined sewer overflow can be assessed and compared using LCA but is rarely considered.

2.2.2 Functional units in seasonal studies

The selection of a functional unit is important in LCA. Choosing different functional units may lead to different LCA results when studying rainfall because influent wastewater quality is changed by rain. Due to the effect of the variation of wastewater volume between seasons, there is a potential that inaccurate results will be obtained when using only one functional unit (Rodriguez-Garcia et al., 2011). In the application of LCA for WWTPs, besides the most commonly used functional unit (1 m^3 of treated wastewater), several other options are available such as the volume of sludge produced and the quantity of removed pollutants (Hospido et al., 2008). To date, there is no strong justification involving FU selection and technologies used in a specific system of WWT. There is argument that per m^3 treated wastewater as FU could not reflect the variation of wastewater flowrate, or wastewater treatment efficiency in WWTPs (Corominas et al., 2013). This restriction resulting to problems in assessing two different systems with different wastewater volume and different treatment efficiency. As an alternative, per kg pollutant removed such as per kg of COD-eq. removed (Wang et al., 2018) or per kg of PO_4^{3-}eq. removed (Rodriguez-Garcia et al., 2011) could be better FU when considering the influent difference and treatment efficiency for the comparative assessment. Per population equivalent could also be considered when considering the difference in wastewater flow rate and its associated pollutants load (Gallego et al., 2008; Kalbar et al., 2013). According to Rodriguez-Garcia et al. (2011), the comparison between two different functional units (e.g, per m^3 treated wastewater and per kg PO_4^{3-}eq removed) resulted in the opposite main environmental potential results, highlighting the importance of the selection of the functional unit. This result is also supported by Corominas et al. (2013) who found that determining a second FU of a system could influence the final result, especially when comparing WWTPs with different influent qualities or different removal rates. It is thus suggested that WWTP-related LCA studies should be conducted using more than one functional unit to strengthen a better understanding of the system under study and to avoid misleading conclusions (Zang et al., 2015). For the study of rainfall effects on the environmental burdens of WWTPs, the selection of functional unit is more important because the influent volume and quality of WWTPs changed by rainfall could affect the treatment performance due to the dilution of the influent and the disturbance to biological treatment. Therefore, a second FU should be introduced in any specific study such as a unit in eutrophication reduction (kg PO_4^{3}-eq) in order to achieve a more comparative result and better interpretation.

2.2.3 Seasonal LCA for WWTPs in developed countries

Due to the importance of rainfall effects on the wastewater inflow rate, treatment performance of WWTPs, and pollutant concentrations in the influent and effluent, the review highlights that a few studies assessed the effects of rainfall on the environmental impacts of WWTPs. The research was mainly in European countries and based on the humid and dry season (Moreira et al., 2004) and winter and summer (Lorenzo-Toja et al., 2016). Nonetheless, conclusions from these studies are not consistent. For instance, based on a Spanish municipal WWTP, Moreira et al. (2004) highlighted that the difference of wet and dry seasons for life cycle environmental assessment was not crucial because the data variability in each seasons in Spain is more significant than the variation caused by rainfall intensity. However, Lorenzo-Toja et al. (2016), identified that the Atlantic region with the highest rainfall resulted in the least LCA environmental impact from WWTPs. Rainfall intensity in different regions in Spain is from 300 mm to >1000 mm. Lorenzo-Toja et al. (2016) conducted a sampling campaign of wastewater treatment in January and February for the winter season and June and July for the summer. The results showed that winter has the highest environmental impact due to 35% higher electricity consumption (mainly for heating), higher waste production, and a higher concentration of pollutants (with no specific percentage reported). Other than the difference in winter and summer effects, they predicted that variability exists in annual precipitation that could affect environmental performances, which is related to the dry and rainy season. However, they found that a limited number of LCA studies are conducted related to this factor, which requires whole year databases from WWTPs.

2.2.4 Seasonal LCA for WWTPs in developing countries

As mentioned, only a few studies have conducted temporal assessments in other regions of different climates such as Latin America, the Middle East and Asia. As a result, less work has been conducted on dry and wet seasons or based on the rainfall effect. For example, in Malaysia, the temperature effect is almost negligible due to constant temperature all year round at a daily average of 27 ^{o}C; however, the rainfall intensity may have an important effect on wastewater characteristics and emissions. Risch et al. (2017) identified that wet weather contributes up to 62% of the total impact on freshwater ecotoxicity using the combined sewer system. On the other hand, a country with high precipitation such as Malaysia practises the separate sewer system. This type of sewer system could decrease the influence of rainfall on the volume of WWTPs compared with the combined sewer system, and it raises a question about which sewer systems are more environmental friendly when involving emissions from WWTPs. In another example, Joel et al. (2017) conducted a study on the dry and wet season in 2013 in Kenya using trickling filter and oxidation ponds, and the results showed lower figures recorded during the wet season for most pollutant parameters. Nevertheless, a seasonal effect study on an activated sludge system could apply LCA for more reliable environmental impact results, especially for the midpoint assessment categories. The results of three years of data from a WWTP in China with a subtropical monsoon climate revealed that five environmental impacts (i.e, abiotic depletion, acidification, eutrophication, global warming and photochemical ozone creation potentials) increased linearly with monthly precipitation below 200 mm/month (Li et al., 2017). This result that there is higher environmental problem during the wet season. These contradictory results about rainfall effects by LCA (i.e. no impact, positive impact, or negative impact) in developing countries indicate that some key issues/factors which influence the environmental impact are still not fully understood. The possible factors are: rainfall effects on WWTPs should be nearly related to how much they can cause changes in flow rate and pollutants concentrations, instead of the solely considers precipitation amount. In addition, one of the most important factors affecting the efficiency of WWTPs has been discovered to be the characteristics of the influent especially wastewater strength (Lorenzo-Toja et al., 2015). Wastewater strength during wet weather should affect treatment performance as it determines the dilution rate, treatment efficiency due to the dilution, and final effluent quality that affects the environmental profile of WWTPs.

In developing countries, numerous non-LCA studies have been conducted on the effect of rainfall on the flow rate of the combined sewer system in WWTPs (Mines et al., 2007b), but limited studies have assessed the rainfall event impact to the separate sewer system and wastewater treatment. Due to infiltration into the collection system, the wastewater flow and characteristics of WWTPs change even in the separate sewer system during the rainy season. Comparative LCA studies of separate sewer system's WWT during dry and wet season is still lagging and thus remains the limitation in the seasonal study of WWTPs. For this type of the collection system, rainfall affects the influent of stormwater via sewer manhole, the ageing sewer network and broken pipe. Thus, to overcome this limitation, it is important to provide a critical analysis involving rainfall events, flowrate variation of the separate sewer system, and the LCA results in seasonal effects. To obtain the expected outcome, a thorough study on time-specific LCA for wastewater treatment is needed to provide guidance on LCA inventory practices (i.e. in what period data should be collected and the technology selection, for relatively small, stable and predictable environmental impacts). As an indirect benefit, this seasonal analysis could provide guidance on future regulations about the environmental impacts from WWTPs. Thus, it is important to investigate LCA sensitivity to time specifically from wet and dry weather by carrying out time-specific LCA in a tropical country to understand how variations in rainfall intensity throughout the year could lead to changes in the environmental impact/profiles.

In overall, there are several differences of LCA-WWTPs between developed and developing countries. It is found that LCA-WWTPs in developed countries which are mostly concentrated in Europe and USA with temperate climate are more site specific and consist of advanced treatment technology including nutrient removal and resource recovery. The sewage characteristic in developed countries are mostly medium to high strength, and mainly practices combined sewer system. The inventory and methodology selection of LCA in developed countries are varies and much improved compared to the studies in developing countries. Meanwhile, LCA-WWTPs in developing countries such as in Africa, Middle East and South East Asia only involves conventional LCA method and many studies still lacking of the primary data (i.e site specific data). Furthermore, there is limited study of LCA-WWTP in developing countries considering; a) dry/wet climate; b) low to medium strength wastewater and; c) separate sewer system, which could be assessed in developing countries. In summary, there is a need to improve the data quality of the inventory practice, reduce uncertainty in LCA of wastewater treatment, and perform analysis beyond the conventional inventory practice such as including spatial and temporal variability and additional pollutant substances. In addition, detail seasonal analysis could provide guidance for future regulations about the environmental impact from WWTP.

2.3 The toxicity impact study using LCA

Toxicity impact from municipal wastewater treatment has attracted great attention in recent years especially when more and more emerging pollutants are detected from municipal wastewater. Although the toxic pollutants such as metals are present in low concentrations, their continued release from wastewater effluent to the environment is believed to have the potential to cause long-term hazards to humans and the environment (Bolong et al., 2009; Alfonsín et al., 2014). Therefore, assessing toxicity from sewage treatment plants started to gain attention increasingly in the last decade to determine the degree of hazards for micropollutants (or other priority pollutants that are not targeted for removal most sewage treatment plants) might cause and if measures need to be taken particularly in vulnerable and sensitive areas.

2.3.1 Importance of micropollutants

Population growth and the associated human activities, such as industry and householder activities, lead to an increase in the content of hazardous elements in wastewater, making the urbanised areas a key pathway for metals and other toxic pollutants to the environment. According to the European Economic Community 1991 (EEC, 1991), the sewage treatment process contributes a considerable amount of direct pollutants to the environment through sludge and effluent-containing toxic substances such as metals and micropollutants. Micropollutants are bioactive contaminants that cannot be fully eliminated with traditional wastewater treatment and are released daily in wastewater such as PPCPs, pesticides, and hormones. A wide range of factors influence the quantity and quality of toxic pollutants such as size of catchment, lifestyle, economic development, and local medical and farming practices, with the three largest sources of PPCPs being hospitals, housing area, and industry (Al-Odaini et al., 2010; Antonio et al., 2016). Bolong et al. (2009) pointed out that these toxicity substances are emitted back to the environment from effluents or adsorbed to the sludge at an average of 65%, depending on their lipophilic characteristics (i.e., the ability of compounds to dissolve). In 2000, the EU framework directive already identified 33 priority pollutants in the aquatic environment including cadmium, lead, mercury, and nickel. Meanwhile, emerging pollutants such as PPCPs have been described as the generation of new pollutants into the environment in significant amounts with harmful effects on organisms due to their abundant nature, persistence, and bioactive and toxic characteristics in the environment. For instance, potential pharmaceuticals such as carbamazepine, diclofenac, and ibuprofen are considered as priority PPCPs for environmental monitoring because of their persistence formation in the water body, and possible contribution towards adverse human health effects (Archer et al., 2017). It is because most of the PPCPs are not completely biodegradable and cannot be totally removed by the conventional wastewater treatment technologies. Furthermore, the continued release of micropollutants from wastewater effluent is believed to cause long-term hazards because the contaminants could form new toxic mixtures in the water body (Bolong et al., 2009; Alfonsín et al., 2014). Unfortunately, most of the current WWTPs, especially in developing countries, are not specifically designed to eliminate micropollutants (Gallego-Schmid et al., 2019). This is because monitoring action to these micropollutants/PPCPs have not been applied in most WWTPs due to discharge guidelines and standards do not yet exist for most micropollutants.

2.3.2 Inventory data from WWTP for toxicity study

2.3.2.1 Heavy metals

The emission of metals from WWTPs consists of direct and indirect pollutants from electricity consumption, chemical consumption, effluent, and sludge. The direct metals in sewage such as mercury, copper, nickel, lead, and zinc mostly come from industrial and domestic wastewater, as well as rainwater runoff that enters the sewer system and leads to WWTPs (Üstün, 2009). These metals will eventually reach the environment from the effluent and sludge. The other indirect source of heavy metals are from electricity production such as barium, hydrogen fluoride, and nickel, which cause toxicity in humans by air or water contamination. Besides electricity production, metals were also released from chemical production and transportation, but these sources normally have a small contribution compared with those released during electricity production.

2.3.2.2 Pharmaceuticals and personal care products

PPCPs are micropollutants that also enter the environment after passing through sewer lines and WWTPs. Classes of these pharmaceuticals include hormones, antibiotics, beta-blockers, and antidepressants. Four classes of personal care products are found: fragrances, preservatives, disinfectants, and sunscreen agents. Most of the PPCPs, such as triclosan, 17a-ethinylestadiol, 17b-estradiol, and bisphenol-A, have been found at different levels of concentrations. In recent years, a few studies have been conducted to determine the behaviour of these micropollutants in domestic and industrial wastewater, including surfactants, pharmaceuticals, personal care products, and endocrine disruptors, from the wastewater treatment process. Among these pollutants, pharmaceutical compounds have been identified as a great concern to surrounding communities as no legal standards have been set for their discharge into surface waters. For instance, recent investigations found that the concentration of pharmaceutical compounds in raw wastewaters (i.e., antibiotics, anti-inflammatories, hormones, and analgesics) vary greatly, resulting in inconsistencies in their behaviour during the treatment steps and their removal efficiencies. In addition, increasing focus has been given to micropollutant elimination through anaerobic digestion of wastewater sludge in the recent years. Due to the variability in treatment levels for micropollutant removal, the real environmental impacts of a WWTP through LCA may be underestimated (Lorenzo-Toja et al., 2016). Therefore, a comprehensive analysis of the facilities must not disregard these emissions despite not being typically monitored by the governing environmental agency.

2.3.3 Life cycle impact methods used for toxicity study

Another vital issue in toxicity impact categories is uncertainty in the selection of the LCIA method and its calculation tool. In LCA, midpoint toxicity impacts have been classified into human toxicity, terrestrial ecotoxicity, freshwater ecotoxicity, and marine ecotoxicity. The various models differ substantially in terms of scope and modelling principles and, most importantly, can fail to arrive at consistent characterisation factors. The selection of the most suitable LCIA method to toxicity impact is still uncertain (Renou et al., 2007). Various characterisation models (e.g. USES-LCA, EDIP and USEtox) have been used to calculate environmental impacts such as human toxicity but vary substantially in terms of scope and modelling principles. CML-IA is the most commonly used methodology for LCA analysis of WWTPs, followed by EDIP2003. The CML-IA method considers a multi-media fate, exposure, and effects model (Huijbregts et al., 2000). In CML-IA, human toxicity is considered, and ecotoxicity is separated into three impact categories: FEP, freshwater aquatic ecotoxicity; MEP, marine aquatic ecotoxicology; and TEP, terrestrial ecotoxicity. The impact in these categories is expressed to a reference substance namely 1,4-dichlorobenzene (DCB). By contrast, the characterisation of toxic effects in the EDIP2003 model is based on the independent fate, exposure, and effects model. EDIP2003 allows the practitioner to calculate toxicity potentials for human toxicity and ecotoxicity potentials, where human toxicity is divided into three different exposure routes: HTP via air, HTP via water, and HTP via soil. Ecotoxicity is divided into three impact categories: acute FEP, chronic FEP, and chronic TEP, wherein the impact is expressed as volume (m^3). IMPACT2002+ method provides human toxicity (carcinogens and non-carcinogen), freshwater ecotoxicity and terrestrial ecotoxicity. These impacts are expressed in different units (e.g, kg C_2H_3Cl eq. for human toxicity and kg TEG soil for terrestrial ecotoxicity). To synchronise modelling methods and characterisation factors for toxicity impact, a life cycle initiative was introduced in 2002 by the United Nations Environment Program and the Society of Environmental Toxicology and Chemistry (UNEP-SETAC) (Rosenbaum et al., 2008). In this programme, huge works were made to identify the sources of differences in toxicity related models/calculation (Hauschild et al., 2013). Based on a range of existing LCIA methods (e.g. Impact 2002 and CML-IA), USEtox was developed where infinite time is used as a sole time horizon (Rosenbaum et al., 2008). The USEtox model is based on the toxicity assessment of pollutants and comprises of six emission compartments: urban air, rural air, freshwater, seawater, natural soil, and agricultural soil. It assesses freshwater (aquatic) ecotoxicity and human toxicity with both cancer and noncancer effects (Rosenbaum et al., 2008) and is expressed in comparative toxic units (CTUh/kg). The United Nations Environment Program (UNEP) and the Society for Environmental Toxicology and Chemistry (SETAC) launched the Life Cycle Initiative to enable users around the world to put life cycle thinking into effective practice (Rosenbaum et al., 2008). As a result, USEtox was developed and recommended as a scientific consensus model after a comparison between several models such as CalTox, IMPACT 2002, USES-LCA, BETR, EDIP, WATSON, and EcoSenee for assessing toxicity-related effects in LCA (Rosenbaum et al., 2008; European commission, 2013). However, due to the complexity of computing characterisation factors, the CFs provided in USEtox are only interim instead of recommended for metals and dissociating and amphiphilic substances (Rosenbaum et al., 2008). In addition, available CFs for PPCPs in the existing USEtox model are very limited, and the modelling on fate, exposure, and impact pathways of chemicals is inaccurate (Emara et al., 2018). IMPACT 2002+ and USEtox are based on similar models, which representing chemical fate, exposure, effect, and optionally severity model. CML 2002/CML-IA is only differs by the calculation of effect and severity indicators. EDIP is a simplified approach that approximates some of these processes without fully describing them (JRC European commission, 2011).

Based on various LCIA methods, some studies use more than one method for the assessment in their research. Muñoz et al. (2008) quantified potential environmental impacts on 98 priority and emerging pollutants using EDIP97 and USES-LCA methodology in WWTPs in Spain. They found that nickel is the priority pollutant in marine ecotoxicity potential using USES-LCA, whereas EDIP did not include this impact category. For further clarification on the LCA methodology for toxicity, Renou et al. (2008) investigated the influence of method selection through a case study of a full-scale WWTP in France. They concluded that no obvious variation was observed within impact categories representing global environmental impact, but great variation was generated by various LCA methodologies associated with toxicity impact categories. In this situation, not only the inventory of toxicity substances but also the assessment methodology need to be improved in LCA. Thus, the toxicity assessment of WWTP was suggested to identify whether the choice of the LCIA methods could influence the final result, strengthen the understanding of the system under study, and avoid a misleading conclusion (Li et al., 2019). This review concluded that the comprehensive methodology evaluation about toxicity impact from WWTPs containing both toxicity substances such as heavy metals and PPCPs is still lagging where the variabilities of toxicity substances in the wastewater, CF availability, and methodology choice could be the main impact to final result.

2.3.4 Toxicity impact studies by LCA in developed and developing countries

Due to the importance of the toxicity effect from WWTPs, a few studies have started to evaluate the effect of toxicity substances such as heavy metals and PPCPs to the environment especially to human toxicity and freshwater ecotoxicity. Most studies related to WWTPs and toxicity impact originate from developed nations and to a lesser extent from developing countries (Lorenzo-Toja et al., 2016; Shimako et al., 2017; Emara et al., 2018). Nevertheless, conclusions from these studies are not consistent and comprehensive. Renou et al. (2007) found that one major issue in LCA toxicity impact research concerned large discrepancies between the life cycle impact assessment methods, mainly for human toxicity, but no detail comparison pertaining specific substances was made between the methods. Wenzel et al. (2008) conducted an LCA study of different wastewater treatment options. They considered the potential toxicity from heavy metals, endocrine disruptors, PAHs, phthalates, and detergents but only nine substances in total. Muñoz et al. (2008) demonstrated that PPCPs were relevant when assessing the release of the influent and effluent of WWTPs, but only PPCPs and direct heavy metals were considered without comparison with the indirect effect such as energy demand. Li et al. (2019) found ecotoxicity impact results using the USEtox model increased by 25% after involving 126 PPCPs in life cycle inventory (i.e. based on secondary data in literature) of advanced wastewater treatment. Lorenzo-Toja et al. (2016) conducted a life cycle assessment considering data of heavy metals and PPCPs in WWTPs in Spain. The results showed no significant impact was found in the effluent life cycle assessment when PPCPs were included in the toxicity assessment using the CML 2002 methodology. However, they identified a significant effect of PPCPs (at an average increase of 40%) in the influent life cycle assessment scenario, highlighting that the impact of these micropollutants in the untreated wastewater cannot be neglected. In addition, they mentioned the lack of a scientifically robust scheme on which PPCPs emissions can be modelled, especially during the end-of-life phase with limited coverage of active pharmaceutical ingredients in LCIA models. According to Muñoz et al. (2008), a problem exists for LCA practitioners due to the lack of relevant substances especially for non-regulated pollutants such as PPCPs. These inconsistent results indicate that some key factors that may influence the toxicity environmental impact potentials by LCA are still unclear. For example, the toxic substances in WWTPs should be closely related to how efficient these toxic substances can cause the change to the toxicity impacts instead of the absolute load of substances assessment only.

Previous research shows that most of the toxicity impact categories were evaluated based on toxicity emissions from electricity and chemical consumption in WWTPs such as the emission of sulphur dioxide and nitrogen oxides which contributes to human toxicity (Hospido et al. 2008; Piao & Kim., 2016). Besides this, impacts on terrestrial ecotoxicity are mainly due to the emission of heavy metals (mostly copper and zinc) into the soil during the end-of-life of sludge. Kalbar et al. (2013) reported an almost similar result for 4 categories—human, freshwater, marine and ecotoxicity impact for 4 different types of WWT, because they are not designed to remove heavy metals and other micropollutants. Thus, these previous studies considered conventional operational parameters about the composition of the influent and effluent with only a few studies considering heavy metals and organic pollutants such as mercury and COD. In LCA, the presence of emerging pollutants in sewage are rarely considered due to the lack of local characterization factors representing environmental fate, exposure to humans and aquatic organisms, and toxic effects caused (Alfonsín et al., 2014). As supported by Carballa et al. (2005) and Suárez et al. (2008), PPCPs should be selected based on their occurrence in wastewater and local characterisation factor. Therefore, further research needed to better characterise the implications of micropollutants in the aquatic environment for the LCA methodology (Morera et al., 2016).

To analyse in details of the effect of these emerging pollutants, Lorenzo-Toja et al. (2016) conducted an assessment of heavy metals and PPCP site measurement campaigns in Spain related to the winter and summer seasons along with the site-sampling of GHGs in two different units of WWTPs located in two different climatic regions, the Atlantic and the Mediterranean. The results for the toxicity impact–related categories indicated that similar performance was obtained in both regions, with winter is the most harmful season. However, a high concentration of heavy metals and PPCPs in the influents during summer (57% higher than in winter) explains that there is a seasonal variation effect. Nevertheless, despite being assessed at different temperatures during the winter and summer, other contrary aspects were found that need to be highlighted. For example, it is important to identify the micropollutant effect based on lifestyle, temperature, high precipitation, and wastewater strength. Moreover, there has not been sufficient assessment of pharmaceutical pollutants in the environment from the Southeast Asian region with low strength wastewater. Thus, evaluation of production and usage of pharmaceutical products in all countries of Southeast Asia has been considered to be essential. The volume of the pharmaceutical industry and human population in these countries has increased significantly in pharmaceutical contamination and its associated risk. For example, in Asia, the concentrations of antibiotics such as roxithromycin, trimethoprim, and sulfamethoxazole are high in both influent and effluent wastewater and surface water. Thus, the study of distribution and behaviour of PPCPs, as well as heavy metals, in the environment is crucial due to large quantities of its manufacturing; however, little is known about this topic, especially in a tropical country such as Malaysia. As a developing country, Malaysia has seen a rapid development of a better living conditions, leading to longer life expectancy and increased demand of pharmaceutical use at home or in the hospital. To date, a number of pharmaceuticals have been detected in the effluents samples from WWTPs in Malaysia, namely salbutamol, atenolol, metoprolol, mefenamic acid, salicylic acid, and furosemide. Moreover, most of the previous toxicity studies for WWTPs were from developed countries with high strength wastewater (e.g. COD value, 250-750 mg/L (Lorenzo-Toja et al., 2016). This highlights the lack of studies concerning both metals and PPCPs contents from WWTPs in developing countries with low strength wastewater, which could produce different environmental impacts. Furthermore, during wet weather periods, domestic wastewater with rainfall is a major component of urban wastewater influent to a WWTP. How the highly diluted water affects metals and PPCPs removal and the effluent concentration were barely discussed. This is especially true for tropical weather countries with high rainfall intensity, where sufficient data on this topic is not currently available.

In summary, although most WWTPs met the local authority's regulatory requirements, many PPCPs and heavy metal compounds are still incompletely removed and later are discharged to the water stream and enter the environment in unknown amounts, especially in developing countries. This contrasts with the level of information about the effect of micropollutants from wastewater in LCA aspects already published and well documented in European and other developed countries with mostly high strength wastewater. Therefore, further research is needed to investigate the occurrence of local organic pollutants, heavy metals, and PPCPs in WWTPs to identify their importance and contribution to provide useful information for LCA practice. Overall, there is a need to improve this gap of knowledge in LCA specifically in the Southeast Asian region by investigating the impact of inclusion metals and PPCPs from WWTPs, as well as identifying the results from different LCIA methods.

2.4 Evaluation of sustainable wastewater treatment by LCA

The world is moving towards sustainable development and a circular economy. In 2015, the United Nations set 17 sustainability-developing goals (https://www.un.org/sustainabledevelopment/). This global strategic platform included developing countries, even though developed countries generally have more resources for sustainable development. One of these 17 goals focuses on water and sanitation. The goal includes supporting developing countries in water and sanitation programmes including water efficiency, wastewater treatment, and recycling and reuse technologies. Some developing countries such as China have planned to build concept WWTPs to reconceptualise water, carbon, and energy systems from the systems level, which can help build a 'circular economy' that closes resource loops to achieve sustainable development. Thus, further studies on the sustainable application in WWTPs combined with LCA, especially in developing countries, are crucial for continuous guidance towards reaching a circular economy in the wastewater industry.

2.4.1 Sustainable application in wastewater treatment

A WWTP consist of various processes that are typically in series (e.g. pre-treatment, primary treatment, secondary treatment, and sludge treatment) (Metcalf & Eddy., 2014). Each unit has specific function designed to remove pollutants in wastewater. Pre-treatment largely removes large solids, grit, and oil, whereas primary treatment designed to remove suspended solids. Secondary treatment is usually based on a biological process that treats organic matter, nitrogen, and phosphorus. Finally, sludge treatment treats the excess sludge by a thickening and dewatering process, and the dewatered sludge is sent to landfills, agriculture land, or incineration plants. An operating WWTP normally uses a large amount of electricity (e.g. for pumping and aeration), as well as chemicals that enhance nutrients removal and improve sludge dewatering process. The operation of a conventional WWTP is not sustainable and generates various environmental impacts such as eutrophication, acidification, and global warming potentials. This chapter reviews and discusses in detail the environmental issues derived from a WWTP and its potential sustainable treatment by using the LCA application.

2.4.1.1 GHG emission

Wastewater treatment operation generates a significant amount of GHGs including carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) (Gupta & Singh., 2012; Chai et al., 2015). CO_2 is mainly produced from the process of fossil fuels to energy as indirect emissions, which involves with 14-36% of total emissions from a WWTP. Methane is formed in the sewer system and under anaerobic conditions, whereas N_2O contributes to 23-43% of emissions during the biological nitrogen removal process (Gupta & Singh, 2012; Daelman et al., 2012). Reducing these direct and indirect emissions from WWTPs could assist in tackling global warming wherein energy reduction and recovery through AD, nitritation-anammox, and A-B process (A stage for carbon capture to improve energy recovery by digestion, and B stage for biological treatment to improve effluent quality) in WWTPs could further reduce GHG emissions. For example, the combination of the anaerobic digestion with combined heat and power, and energy-optimising activated sludge could save over 102,000 tonnes CO_2/year, which equals 50% of energy optimization (Sadler et al., 2009). However, to quantify the correct emission, an established environmental tool is required because a complex calculation must be completed, including the range of electricity and chemical consumption, as well as site-specific factors. Currently, the quantification of direct GHG emissions are implemented by observing CH_4 and N_2O emission, which present global warming potentials (GWP) of 21 kg CO_2 eq. and 310 kg CO_2 eq. per kg of compound emitted, respectively (Listowski et al., 2011). The GHGs are produced within the WWTP in various locations and treatments. The main sources of CH_4 emission are in anaerobic conditions such as sludge thickeners and sludge storage tanks (Daelman et al., 2012). Nonetheless, another important source of CH_4 is the sewer system (Masuda et al., 2015). Thus, CH_4 is not only emitted from the anaerobic tanks but also in aerated areas via stripping. Meanwhile, N_2O is mainly reported to be released from anoxic zones of activated sludge configurations where nitrification and denitrification reactions lead to the production of N_2O (Kampschreur et al., 2009). Additionally, some studies have also pointed out that N_2O emissions occur in de-gritter units, sedimentation tanks, secondary clarifiers, and sludge treatment tanks (Harriss et al., 1995). Overall, a suitable methodology needs to be identified to calculate GHG emissions from WWTP and find suitable technologies to reduce these emissions.

2.4.1.2 More stringent discharge standards

The discharging of nitrogenous components of wastewater effluent to a water body can cause the deterioration of water quality and eutrophication to aquatic life (Sun et al., 2010). Therefore, the higher limit of effluent discharge from WWT has been introduced especially in urban areas and developed countries such as the USA, Europe, and Japan. For example, the EU Urban Waste Water Directive has set requirements at 10-15 mg N/L and 1-2 mg P/L, which require the improvement and upgrading of wastewater treatment technology such as applying enhanced biological phosphorus removal (EBPR), anaerobic ammonium oxidation (anammox), and aerobic granular sludge (AGS). Meanwhile, in most developing countries, the discharge requirement is lower because most of the technology is still at a lower efficiency for treatments than that used in developed countries. For example, most of the WWTPs in developing countries only consider nutrient and organic matter removal, whereas most treatment plants in Europe have already applied resource recovery technology such as anaerobic digestion and water reuse technology. Therefore, the scale of environmental impact varies depending on the local factor, as well as regulation and technology adopted mainly for eutrophication impact, which is regularly monitored. The monitoring of effluent data is normally compulsory for all WWTPs to identify the level of nutrients discharged into the water body where it could affect the quality of river or ocean. Thus, the assessment of site-specific discharge standards from a WWTP is crucial, especially in urban areas. These assessments can be potentially used by decision-makers to assess effluent quality regulation and consider for upgrading requirements in the future.

2.4.1.3 Sludge treatment and disposal

For the sewage sludge generated from WWT, approximately 10 million and 8 million tonnes of dry sludge were generated in the European Union and United States, respectively, in 2010 (Wan et al., 2016). This problem affects the environment where energy is consumed for the treatment and disposal process, polluting underground water and soil by heavy metals and GHG emissions; an estimated 32-39% of CH_4 is emitted from the sludge (Chai et al., 2015). Apart from 90% reduction of sludge volume after incineration (Kasina et al., 2019), integration technology of anaerobic digestion and struvite recovery could help to reduce the amount of sludge from WWTPs. For instance, Amersfoort WWTP in the Netherlands, which has commissioned three advanced technologies including struvite recovery by Ostara, could reduce 17% sludge volume while recovering 45% of phosphate and producing 60% more biogas to energy.

2.4.1.4 Nutrient removal and recovery

Nutrient removal from WWTPs consists of treatments to remove nitrogen and phosphorus before being discharged to water body and requires different processes. In nitrogen removal, nitrogen is oxidised from ammonia to nitrate through nitrification process, which takes place in aeration tanks/secondary treatment tanks. This process is followed by denitrification where nitrate is converted to nitrogen gas, which is released into the atmosphere and consequently removed from the wastewater. Denitrification process needs anoxic conditions to encourage proper biological reaction. Various technologies are increasingly available for nitrogen removal from wastewater that leads to a cleaner discharge to a water body and sustainable application. For instance, nitrification-denitrification is increasingly applied worldwide due to its technical maturity. Other nitrogen removal technologies such as aerobic granular sludge (AGS) and anammox are increasingly applied because they have the potential to reduce energy and chemical consumption. Apart from this, the A-stage from AB process removes about 55-65% of the organic load, and approximately 80% of nitrogen elimination is achieved in the B-stage (Nowak et al., 2011). Phosphorus removal can be achieved by chemical phosphorus precipitation such as using iron chloride. Phosphorus can also be biologically removed using polyphosphate-accumulating organisms (PAOs) in EBPR. PAO could accumulate great quantities of phosphorus within their cells, and separate the phosphorus from the treated water. Other phosphorus removal technologies include ion exchange chemical removal and the emerging aerobic granular sludge (AGS) process.

The regular application of nutrient reuse from WWTPs is applying sludge to agricultural lands such as composting due to the nitrogen and phosphorus content in the sludge, both of which can be nutrient sources for plants. However, not all the sludge can be directly applied to agricultural lands due to the pollutant contents such as heavy metals, which can harm the environment. This is the reason why more nutrient recovery research and application is increasingly conducted worldwide for a better or more sustainable consumption. Recently, the focus has been on chemical phosphorus product due to its scarce resource. For instance, various technologies have been developed to recover phosphorus from WWTPs such as Ostara from Canada and Gifhorn, Airprex, and Unitika from Japan where the struvite can be sold as fertiliser. For example, struvite crystallisation by Airprex was used to retrieve phosphorus following the anaerobic digestion process and EBPR. Struvite were produced by air stripping the reactor, while adding chemical product such as magnesium. In addition, P recovery process could improve the effluent quality and minimize sludge production while meeting the stringent P discharge limit (<2 mg/L). In terms of efficiency, the recovery of phosphorus from the side stream can achieve up to 50% of P recovery potential, whereas 90% can be recovered from sewage sludge ashes by incineration. However, the combination of EBPR systems for P removal with P recovery technology has received wide interest because EBPR could increase the potential for P recovery by more than 90% (Urdalen, 2013).

2.4.1.5 Energy recovery to achieve energy neutral or positive wastewater treatment

Basically, energy can be recovered from WWTPs by the process of anaerobic digestion of sludge. However, according to Stillwell et al. (2010), this type of technology can only recover approximately 30-40% of the total energy requirements in WWTPs. Therefore, single technology such as the AD of sludge is not enough to achieve energy-neutral or -positive WWTPs, requiring appropriate optimization and technology improvements. For example, technologies such as anammox or the A-B process have to be integrated into the traditional operation of a WWTP to achieve energy-neutral status. As a reference, the Strass WWTP in Austria, which was designed and operated with two-stage activated sludge plant (A-B process) integrated with side-stream anammox and sludge digestion, has significantly achieved an 8% energy surplus (Jonasson, 2007). This is possible due to achieving an average energy consumption of 0.3-0.5 kWh/m^3 while the source of carbonaceous materials in wastewater reached a recovery of 1.7 kWh/m^3 energy. Therefore, by combining the emerging technologies such as side stream anammox with the adsorption-biological (A-B) stage process followed by anaerobic digestion (AD), energy self-sufficient wastewater treatment could be achieved. The environmental assessment tool such as LCA can be used to evaluate this technology integration and identify the environmental benefits from the energy recovery. However, an intensive assessment methodology should be identified for a convincing result due to the complex technology integration, which requires every detailed aspect of the design and assessment.

2.4.1.6 Integrated technology to upgrade wastewater treatment plant

Conventional WWTPs remove organic matter for the protection of the aquatic environment. However, with an increasing population, municipal WWTPs are faced with the challenge of ensuring sustainable treatment, which includes nutrient removal and resource recovery. Sustainable wastewater treatment greatly relies on treatment technologies. Several new technologies have been developed to treat wastewater more efficiently with low energy and chemical consumption, great potential for resource recovery, higher effluent quality, reduced sludge production, and reduced GHG emission. However, the vast majority of these novel technologies are still in the early stages of research without foreseeable commercialisation. For the pressing task of achieving or moving towards sustainability, plants must rely on existing and mature technologies. In fact, it has been widely accepted that applying the existing technologies and integrating them effectively can achieve more sustainable wastewater treatment instead of waiting for the maturity of novel technologies (Wan et al., 2016). Furthermore, it has been found that some technologies can reduce energy consumption and GHG emissions, whereas others achieve resource recovery. Besides the environmental impact, the integration of various technologies also deal with technical and economic impact assessments to identify those technologies that are technically applicable and economical (Amann et al., 2018). However, research on the holistic analysis of integrated technologies in the wastewater treatment area is still a fairly new approach. The lack of a comprehensive analysis about the comparison of the environmental impacts and benefits from integration treatment technology to existing plants hinders the practical application of the proposed technologies (Mininni et al., 2015). In addition, most of this type of research so far has not considered local factors, which may cause great discrepancies.

Thus, there is a possibility to introduce a new configuration of WWT involving nutrient removal and resource recovery (e.g. water, energy, and nutrient) from the suitable existing technology that may retrofit current technology treatment for the future. WWTPs could generate electricity and heat from the methane produced by sewage sludge in anaerobic digestion. Besides the energy efficiency, nitrogen and phosphorus removal has been adopted in many WWTPs mainly to reduce eutrophication impact in the water body. The elimination of phosphorus by chemical precipitation could achieve low phosphate concentration in the effluent, making this technology widely used (Maurer et al., 1999). A few technologies have been developed towards more sustainable treatment, and among the technologies that have been practised on full-scale systems are EBPR and struvite recovery. EBPR is able to decrease the number of chemicals used for the phosphorus removal (Maurer et al., 1999), and P recovery technology produces a high grade of P minerals in the form of magnesium ammonium phosphate (e.g. struvite - $NH_4MgPO_4.6H_2O$) for use as fertiliser (Pradel & Aissani, 2019). EBPR can be a less expensive process to construct and operate; it also generates less sludge and does not use a chemical substance (Blackall et al., 2002). Solids generated in EBPR can significantly offset the demand for synthetic fertilisers through integration with P struvite recovery technology (Foley et al., 2010). However, this nutrient removal and resource recovery treatment schemes have some limitations such as increases in energy consumption, chemical consumption, and cost (Bashar et al., 2018), so holistic assessments are needed for the sustainable upgrading of wastewater treatment. Sena & Hicks. (2018) highlighted that environmental impacts associated with P recovery that involve infrastructure construction, energy and chemicals required could outweigh the benefits. Furthermore, hotspot analysis to upgraded WWTPs for nutrient removal and resource recovery is important for the identification and selection of efficient technology.

2.4.2 Application of LCA to select sustainable WWTPs

This review clearly shows that different technologies have been developed, integrated, and applied to achieve energy and phosphorus recovery, improve effluent quality, and reduce GHG emissions. In addition, the successful demonstration of STRASS is an aspiring example to show that utilising a combination of existing technologies can lead to energy-neutral wastewater treatment and gain environmental benefits. As seen, this plants achievements are a promising demonstration of the sustainable wastewater treatment system. However, the question how to apply this to a wider context still needs systematic level assessment in environmental and economic aspects, with consideration for local factors. As more technologies are being developed or applied to upgrade existing wastewater treatments for resource (e.g. water, energy, and nutrients) recovery and more stringent discharge standards are being implemented, environmental impact analysis from different aspects is imperative to achieve ‘real’ sustainable wastewater treatment. This situation shows that the selection of a matured and efficient environmental and economic assessment tool is important to achieve convincing results towards more efficient wastewater treatment with low impact to the environment. In essence, sustainable wastewater treatment should, over a long-term perspective, be able to treat wastewater while protecting human health and environment with minimal use of scarce resources. In addition, it should also produce beneficial recovery products and be socially, technically, and financially viable. This is because wastewater, which was previously considered as a disposal liability, can now become valuable resources. Water reuse, nutrient removal and recovery, and energy self-sufficiency are among the core parts of wastewater treatment operations working towards sustainability. Apart from this, other environmental factors such as eutrophication, GHG emissions, and pollution from residual sludge have to be considered at the same time to evaluate the sustainability of wastewater treatment. This is because the current global concern is to identify the trade-off between environmental issues such as eutrophication, global warming, toxicity, and electricity used, with more stringent effluent limits and the increased utilisation of some resource recovery technologies such as struvite precipitation of phosphorus.

Due to the significant effect of upgrading technology to the environment and economic, a few studies have evaluated the effect of upgrading plants compared with the existing treatment. Nevertheless, research regarding upgrading wastewater treatment using LCA are various and inconsistent in terms of technology integration and assessment methodology, and most studies have not included an economic assessment. Studies have been conducted to identify the environmental effects of upgraded processes in WWTPs (Moreira et al., 2004; Hao et al., 2019), but the complexity of these studies vary with different system boundaries and selected technologies and impact categories. The impact of phosphorus recovery from WWT is rarely considered where, for example, comprehensive and quantitative LCA studies involving the impact of phosphorus struvite recovery from WWT technology are still limited (Zang et al., 2015). In fact, only a few studies of LCA focused on energy recovery and, for these, important methodological issues in LCA still need to be addressed. Therefore, due to the lack of methodology consistency and transparency in the current practice for LCA-WWT, it is important to emphasis on the need for a robust, transparent, and standard method for sustainable technology assessment. Corominas et al. (2013) has also indicated in their research conclusion that standardised guidelines to ensure the quality of LCA methodology is needed. A holistic assessment is especially important to the matured technology that is increasingly applied worldwide including in developing countries. In a study by Coats et al. (2011), LCA was applied to evaluate the impact from WWTPs with upgrading technology of phosphorus removal. They concluded that biological P removal as a best practice should only be added with a chemical process if necessary, based on the life cycle environmental analysis of two P removal scenarios (e.g. biological versus chemical P removal). The results by Hao et al. (2019) who studied LCA of resource recovery technology (e.g., water reuse, electricity, thermal, and P recovery) of WWTP in China found that thermal energy recovery from sludge incineration significantly contributed to 40% of total resource recovery score, followed by 30% electricity recovery, and achieved net-zero impact from total environmental value. Meanwhile, P recovery only achieved 6% from the total resource recovery process. This review indicates the need to combine both nutrient removal and resource recovery using local data to further identify their impact and benefit to the environment while improving LCA methodology itself.

2.4.3 Life cycle economic assessment of wastewater treatment

Economic assessment is one of the most important criteria in identifying the feasibility and efficiency of integrated technology in WWTPs (Hernandez-Sancho et al., 2010; Mayer et al., 2016). Evaluation of the capital, operations and maintenance costs, and product revenue are important criteria for technology integration. Standard LCA practices encompass only environmental impacts, which excludes economic and social impacts. However, some researchers have increasingly conducted economic analysis for WWTPs, such as a life cycle costing assessment for the selection of wastewater treatment (Rawal & Duggal, 2016), an economic valuation of environmental benefits from the wastewater treatment process (Hernandez-Sancho et al., 2010), and an economic assessment for greywater recycling using whole life cost (WLC) (Makropoulos et al., 2005). Morera et al. (2016) developed a novel method integrating environmental and economic criteria for selecting the best process for WWTPs. On the other hand, (Lin et al., 2016) suggested exploring a weighting system to monetize the environmental issues and convert all the economic and environmental criteria into a single sustainability score. Lorenzo-Toja et al. (2016) proposed a system value assessment using LCA and LCC for WWTPs based on ecoefficiency concepts. A modelling approach is needed to have a holistic environmental and economic performance of a diverse process (Lin et al., 2016). Less than 10% of the reviewed studies included an economic efficiency analysis. Furthermore, none of the previous studies assessed the consequence of product value from the wastewater industry involving energy recovery and nutrient recovery to agriculture in specific countries that integrated with the nutrient removal process. This issue carries some questions about how LCA and LCC can support the creation of a circular economy concept in WWT and ensure a positive environmental impact. Additional questions include where should the substituted materials and products be accounted for and who can claim the benefit.

To answer the questions above, a complete economic evaluation for the integration of the technologies proposed should be included and thoroughly evaluated towards a circular economy and sustainable development. This is because some technologies have not been applied in the wastewater treatment industry, and the recovered products such as struvite have yet to be fully accepted by agricultural organisations especially in developing countries. Therefore, an in-depth evaluation needs to be conducted on the economic aspects of the proposed integrated technology identify its compatibility mainly for energy and nutrient recovery. For example, the market value of recovered product such as P fertiliser could influence the economic situation where the price can be different across the world, depending on the demand, regulations, and social acceptance. In addition, economic evaluations of the capital, operation and maintenance, and product revenue are other important criteria for the integration technology besides environmental factors.

In summary of the environmental and economic assessments, an increasing number of wastewater evaluation methods only focus on a limitation aspect of sustainability, while the roles and contributions of the whole system are difficult to understand and thus could exacerbate problems when planning for achieving sustainability. Therefore, although some work on environmental and economic assessments has been done as mentioned previously, a lack of systematic analysis exists for the sustainable development of integrated WWTPs with resource removal and recovery. Furthermore, even though LCA application in wastewater treatment has grown significantly in the last few years, LCA was not originally designed for wastewater treatment analysis, and thus, some issues exist that could be improved including refining the data inventory, impact methodology, and economic indicators for more reliable life cycle assessment. This is because to achieve true sustainability, an assessment from an integrated perspective is needed wherein the environmental impacts of WWTPs do not exceed its benefits (Zang et al., 2015). Further research should consider wider impact categories through system analysis that considers temporal, spatial, and local specific criteria of WWTPs. This is because it is important to acknowledge the barriers that may vary based on geographical and cultural contexts (Larsen et al., 2009), so a study should focus on a tropical region, such as Malaysia.

2.5 Malaysian context

2.5.1 Review for the sustainable strategies in Malaysia

Malaysia is selected as the main case study for this research followed by United Kingdom. As pointed out before, local factors such as government guidance, policy, wastewater characteristics, pollution of water bodies, climate, main fuel and local practices for wastewater treatment could affect the selection of technologies towards sustainable development and environmental impact of the integrated wastewater treatment. Therefore, a detail review of Malaysia information and related characteristic is further discussed in this section.

Malaysia is located in Southeast Asia with a current population of 30.1 million people, producing the total volume of wastewater of 7.53 million m^3/day. As a developing country, the wastewater collection and treatment coverage is very low. Until the year 2013, only 50% of the wastewaters treated by mechanized plants while others still use untreated individual septic tanks and oxidation ponds (Din, 2013). For the mechanical WWT, activated sludge (AS), aerated lagoons, rotating biological contactors (RBC), extended aeration (EA) and trickling filters are the current treatment technologies used. Malaysia’s current strategy is to reduce individual untreated wastewater by planning towards proper centralised treatment system. With more WWTP facilities to be built, it is in a good position to directly adopt well-developed technologies for sustainable wastewater treatment. This is because, more than 90% of current wastewater treatment technologies in Malaysia only involve conventional treatment (i.e. without energy recovery). This type of treatment cannot achieve sustainable operation for the rapid growth municipal WWTPs. Moreover, many resources are required such as energy and money for transportation, treatment and final disposal of sludge. As mentioned before, due to the chemical energy contained in wastewater, it is seen as valuable fuel to supplement power generation in Malaysia. However, how policy and environmental regulation from the government of developing country can best serve in improving sustainability. Upon the UN Climate Conference in Paris 2015, Malaysia has striven to reduce 45% of its carbon emission intensity by the year of 2030. Previously, it has introduced feed-in tariff (FiT) in 2004 and subsequently establishes the Sustainable Energy Development Authority (SEDA) Act in 2011 to fulfill the national aspiration towards achieving energy self-sufficiency and mitigating climate change. As in 2014, only 3.3% renewable energy produced in Malaysia where 96.7% consumed energy generated from the fossil fuel (British Petroleum, 2016). Based on this situation, Malaysia has adopted a target of 11% installed renewable energy capacity by 2020. Since water sector consumes 3-5% of total energy consumption of the country, it is an important factor in leading Malaysia to sustainable development, which we could include renewable energy and nutrient recovery in WWTP. Moreover, with the possibly strong municipal wastewater due to the implementation of separate sewer collection system, and hot climate throughout the year with temperature ranging from 22 ^{o}C to 32 ^{o}C, this situation is more favourable to adopt anaerobic digestion, anammox treatment, and A-B process. This is due to more energy could be recovered from stronger municipal wastewater, less or no energy is required by anaerobic digestion and anammox, and treatment efficiency is higher due to higher bacteria activity at a higher temperature.

On the other hand, the previous survey in 2005 by National Hydraulic Research Institute of Malaysia (NAHRIM) identified that 62% of lakes and reservoirs in Malaysia were in serious eutrophic condition. While, in 2013, out of 473 rivers monitored by Department of Environment Malaysia (DOEM), there are 72% polluted and 6% were classified as heavily polluted (Huang et al., 2015; Ariffin & M Sulaiman, 2015). As such, Department of Irrigation and Drainage of Malaysia is working towards cleaner water bodies such as introducing River of Life Project in 2012 which requiring cleaner effluent, especially from WWTP even though there is no concrete decision yet on the improvement of the wastewater effluent standard. Current effluent discharge limit to the river is 20-50mgN/L, 20-50mgBOD$_5$/L and 120-200mgCOD/L, but phosphorus limit is only required when discharging into the stagnant water bodies with 5-10mg P/L. Meanwhile, due to rapid expansion in crop production in Malaysia (e.g. rubber, oil-palm, cocoa etc.), there are huge increasing amount in the importation of phosphate fertilisers such as from China and Australia amounting £28.8 million in 2005 and £58.8 million in 2011. Based on this situation, P recovery to fertiliser from WWTP is a favoured option which could be considered. Finally, most of the sludge from WWTP in Malaysia is disposed of in the landfill which could impose potential risk and pollution of the underground of water and soil. Therefore, these situations would require more research and planning towards sustainable technology which could reduce the volume of sludge and other environmental impacts. However, the existing technologies from developed country should be carefully evaluated by considering the difference in culture, land, climate and economic. Currently, there are lacking of policies and regulation in Malaysia regarding the resource recovery from water and wastewater sector. Therefore, based on the future result of this research, the suggestions of new regulation and policy can further be explored in country-wide basis. For instance, economic incentives to enhance technology and market for nutrient recovery from WWTP can be proposed and brought about through regulation.

Although Malaysia is not as ambitious as China now, how to develop sustainable wastewater treatment in Malaysia in the global strategic platform of sustainable development is still pressing. The research on sustainable wastewater treatment from the system level is still very new while little work has been done in Malaysian context with the consideration of local factors, specifically on the overall environmental impact of wastewater treatment. Therefore, a detail review of Malaysia information and the related local wastewater situation is further discussed in this section.

In Malaysia, Environmental Quality Act 1974 (Act 127) is the primary federal legislative for water quality. As for sewage, the latest regulations are set in the Environmental Quality (Sewage) regulations 2009, which applicable to any premises discharge sewage into Malaysian waters except for housing development with less than 150PE. Therefore, those treatment system developed after 2009, have a stricter standard in terms of concentration limit and numbers of parameters regulated (Ariffin & M Sulaiman, 2015). For example, phosphorus limit has been introduced for the first time in 2009 with 5mg/L for standard A and 10mg/L for standard B. The other standard parameters included are such as BOD, COD, suspended solid, pH, oil and grease and NH_3-N. Indah Water Konsortium Sdn Bhd (IWK) is currently the biggest wastewater treatment operator in Malaysia where it manages more than 70% of wastewater treatment management. The list of all effluent discharge limit are shown in **Table 4**.

Table 4. Environmental Quality (Sewage) Regulation 2009 for new sewage treatment system (Malaysia)

Parameter	Unit	Standard A	Standard B
Temperature	^oC	40	40
pH value	-	6.0-9.0	5.5-9.0
Biochemical oxygen demand (BOD_5) at 20^oC	mg/L	20	50
Chemical oxygen demand (COD)	mg/L	120	200
Total suspended solids (TSS)	mg/L	50	100
Oil and grease (O&G)	mg/L	5	10
Ammoniacal nitrogen, AMN (river)	mg/L	10	20
Ammoniacal nitrogen, AMN (stagnant water body)	mg/L	5	5
Nitrate-nitrogen (river)	mg/L	20	50
Nitrate-nitrogen (enclosed water body)	mg/L	10	10
Phosphorus (stagnant water body)	mg/L	5	10

For sewage sludge production from WWTPs, Malaysia generates approximately 5 million m^3 per year. However, the amount has been predicted to reach 7 million m^3 per year by 2022, by Indah Water Konsortium. Sewage sludge / biosolids, is the sludge waste that has been produced after wastewater is treated in a wastewater treatment facility. These sludge is usually in a dilute suspension form, which typically contains 0.25 to 12% of solid (IWK, 1997). Pathogens, heavy metals and toxic pollutants present in the untreated wastewater. For example, sewage sludge also contains high amounts of heavy metals such as lead, cadmium, nickel, chromium and copper due to its industrial origin (Raymond & Felix, 2011). This is why most countries strictly regulate the usage of sewage sludge in agriculture or as a soil amendment because of its potential of being harmful to humans, animals, and the environment (Odegaard et al., 2002). Sewage sludge also comprises of organic matter (e.g. COD) and nutrient (e.g. nitrogen and phosphorus) that makes it suitable to be used as an organic fertiliser (Singh & Argawal, 2007). But, sewage application to land for a long period may result to the accumulation of heavy metals in soil. The increase amount of heavy metals are dangerous because they are usually non-degradable.

The environmental impacts from WWTP such as greenhouse gas emission, toxicity and acidification potentials is not properly measured since they are not regulated by the environment agency. To date, these environmental impacts have been ignored in the regulatory. Hence, it is a need for detailed life cycle inventory and assessment for identifying a correct environmental burden from WWTP. For the life cycle development in Malaysia, by the initial reviewing, there have been limited existing databases provided for local life cycle inventory involving wastewater management. The Malaysian Life Cycle Inventory Database (MY-LCID) has been established in 2005 by the Malaysian government under SIRIM Berhad (Scientific and Industrial Research Institute of Malaysia) but it is still at the very beginning stage and seeking to enhance its contents to wider aspects. This research could provide holistic operational databases acquired from variety sources including operational parameters, site sampling database, government websites, technical reports and local journal articles. The database is up-to-date and reflects the current environmental performance of wastewater treatment.

In overall summary, the role of environmental impact and cost assessment is importance for the sustainable development of WWTP. The technologies that will be adopted towards sustainable WWT could not only be assessed based on the single factor. Thorough environmental impacts have to be evaluated to provide guidance for future policy and water industry to find the trade-off of environmental factors and move towards to sustainable development.

2.6 Summary of literature review and knowledge gaps

Based on this review, LCA has been used as an effective and efficient methodology for the environmental impact of WWTPs. However, LCA applied for WWTPs is still relatively new compared with other manufacturing processes. The question of how WWTPs can implement LCA to achieve reliable results of their environmental impact still needs further research. Additional questions on how to implement LCA in developing countries such as Malaysia to provide guidance to policy makers and WWTP on operations and upgrading still remain and prove to be challenging. This thesis identified three main knowledge gaps of LCA for WWTPs and three challenges in the life cycle assessment methodology to address for WWTPs in developing countries such as Malaysia: (1) there is a need to assess the influence of seasonality (i.e. dry and wet season) on the environmental impact, (2) there is a need to investigate toxicity impacts from WWTPs, and (3) there is a need to evaluate environmental sustainability of different processes for upgrading. Therefore, discussion about each knowledge gap is provided below.

a. The seasonal effect on LCA:

   Firstly, it is well known that rainfall affects the wastewater flowrate to WWTPs especially with combined sewer systems and their treatment efficiency, but it is still unclear and not conclusive what influence rainfall has on the environmental profiles of WWTPs with LCA. Understanding the rainfall influence on environmental impacts is important because it could provide some guidance on sewer system selection, trade effluent received, and operation adjustment of WWTPs to alleviate the negative impacts from rainfall. Furthermore, a rainfall study will result in better practice guidelines for LCA by recommending whether it is necessary to conduct LCA in dry and wet seasons to provide a more accurate picture about the environmental impact of WWTPs from rainfall. Secondly, previous studies on the environmental impacts of rainfall focused on the combined sewer overflow but ignored the normal WWTP operations without combined sewer overflow during wet seasons with rainfall, even though WWTP operations and the effluent in wet seasons are quite different from those in dry seasons. Thirdly, when rainfall effects were investigated, wastewater strength was not considered, which may lead to the different results. Fourthly, there is a so-called ‘common sense’ or ‘intuition’ that rainfall only affects the operations of WWTPs with combined sewer systems but not those with separate sewer systems where storm runoff is collected separately, so minimum effects from rainfall are expected on wastewater quantity and quality to WWTPs. Rainfall effects on wastewater quality and quantity are more complicated. Fifthly, according to the main challenges and research gaps identified by the review paper published recently on life cycle assessment of wastewater treatment in developing countries (Gallego-Schmid et al., 2019), a lack of LCA of WWTPs exists in some developing countries (i.e. the representation of geographically different countries), as well as detailed information about the influent, the effluent, and the sludge produced, restricting the development of the best practice guidelines of LCA for the region.

b. Inclusion of metals and PPCPs for a toxicity impact study using LCA:

The risk of micropollutants that exist in wastewater were rarely considered for LCA-WWTP especially in tropical weather with low strength wastewater. The inclusion of micropollutants is important for identifying the toxicity impact especially to human health, as most previous studies only focused on the impact by electricity and chemical consumption. Therefore, it is important to know the dominant type of micropollutants of metals and PPCPs from wastewater treatment that is contributing to the environment both from direct and indirect emissions. Secondly, the average concentrations of pollutants are lower in developing countries than in developed countries, and this poses three interesting questions: (i) what levels of PPCPs and metals are in Malaysian wastewater?; (ii) how do they influence the toxicity of wastewater?; and (iii) if necessary, what measures need to be taken in the future for more stringent discharge of treated wastewater? To answer these questions, sampling campaign must be conducted because the data from the literature especially from developed countries could not represent the situation in a developing country with different weather conditions. Thirdly, information about PPCPs and metals in developing countries is limited, resulting in difficulty of LCA application in this region. Therefore, it is necessary to conduct sampling with consideration of existing technology used and local wastewater quality. This would provide relatively accurate data to life cycle inventory to execute a complete LCA assessment and also extend the data of PPCPs and metals to developing countries for further toxicity study in the future. Finally, the contribution of PPCPs and metals to toxicity categories remains unclear compared with indirect emissions such as electricity and chemicals used for wastewater treatment. Furthermore, it is important to conduct LCIA methods comparison in toxicity assessment for better result interpretation. Therefore, the toxicity impact of PPCPs and metal emissions needs to be assessed from a large centralised WWTP with low strength wastewater to provide useful information for LCA practice.

c. Using LCA to guide the upgrading designs of WWTPs:

It is important to upgrade existing WWTPs for nutrient removal and resource recovery for more efficient treatment, but identifying the impacts or trade-offs is also important for future reference, an aspect which is rarely discussed. Secondly, there as a lack of comparisons of environmental and economic impacts for the integrated nutrient (i.e., nitrogen and phosphorus) removal and resource recovery. For example, energy and P recovery could further reduce the other environmental impact within the same treatment scheme, but the economic cost is uncertain due to additional chemical and electricity consumption. The real trade-off between these upgrading systems needs to be identified for future implementation strategies towards more efficient and sustainable WWTPs. This is because, to achieve true sustainability, an assessment from an integrated perspective is needed where the environmental impacts of WWTP should not exceed its benefits (Zang et al., 2015). Thus, the comprehensive design for upgrading and a method for evaluating both environmental and economic burdens are needed to provide useful information for policy makers and practitioners on the rectification or upgrading of WWTPs. Thirdly, the lack of environmental impact weighting for different phases of operation leads to difficulties in identifying environmental burden hotspots. Most studies remain limited to single-unit operations such as sludge treatment without conducting a comprehensive impact from the whole treatment. Thus, it is crucial to investigate the hotspot impact from upgrading treatment to identify which process has the most burden. Finally, few studies have been conducted in developing countries especially when involving the integration of nutrient removal and resource recovery. Thus, a comprehensive assessment for evaluating both environmental and economic burdens from site-specific data is needed to provide useful information for upgrading wastewater treatments plants in terms of technical, environmental, and economic impacts.

Table 5. Overview of existing study of LCA in wastewater management from 2006 to 2019

No	**Reference**	**Journal**	**Country/ Area**	**Goal**	**FU**	**Processes considered**	**Sludge Disposal**	**Data Source/inventory**	**LCIA method & tool**	**Impact Category**	**Scale**
1	Hao et al., 2019	Water Research	China	To evaluate environmental impacts of a WWTP and compare with resource recovery option	PE/ year	Construction, operation, demolition	-	Foreground data: WWTP Background data : *Chinese* life cycle database	Tool: - Method: CML2001	GWP, EP,AP,ADP,HTP,	200,000m^3/day
2	Li et al., 2019	Journal of Environmental Management	China	To provide assessment of environmental impacts involving 126 PPCPs in advanced wastewater treatment by LCA	1 m^3 /day	Construction and operation	-	Foreground data: WWTP Background data : Gabi	Tool: Gabi 6.0 Method: Usetox and Traci	AP, EP, HTP, GWP,OLDP, FEP	-
3	Awad et al., 2019	Science of the Total Environment	Egypt	To study environmental performance of different scenarios in developing country.	1 m^3 /day	Construction and operation	-	Foreground data: WWTP Background data : Ecoinvent	Tool: - Method: CML2000	AP,GWP, EP, POP, OLDP, DARP, TEP,FEP	40,000 m^3/day
4	Delre et al., 2018	Journal of Cleaner Production	Denmark and Sweden	To investigate the contribution of direct CH4 and N2O to annual carbon footprint of seven WWTPs.	1 Mg of input, 1kg carbon, N & P removed	Operation	On-site incineration and application to agricultural land	Foreground data: WWTP Background data : Ecoinvent, EASETECH, ELCD	Tool: EASETECH v2.3.6 Method: IPCC 2006	GWP	-
5	Pradel et al., 2018	Science of the Total Environment	France	To assess impact of recovered Phosphorus from WWTP	1 kg of struvite recovered	Construction and operation	Use for fertiliser	Foreground data: WWTP Background data : Ecoinvent v2.2	Tool: Gabi v6 Method: CML-IA	ADFFP, AP,EP,FEP,MEP,TEP,HTP,OLDP, POP	300,000 PE
6	Amann et al., 2018	Resources, conservation and recycling	Austria	To analyse impact of P recovery form WWTP	PE/year	Operation	-	Foreground data: Literature Background data : Ecoinvent v2.2	-	GWP, AP	-

No	Reference	Journal	Country/ Area	Goal	FU	Processes considered	Sludge Disposal	Data Source/inventory	LCIA method & tool	Impact Category	Scale
7	Bai et al., 2017	Journal of Cleaner Production	China	To investigate how, and to what extent, the LCA results could be influenced by the adoption of various LCA methodologies, via a case study of a representative WWTP in China	10,000 m^3 of waste-water	Operation, sludge treatment	-	Foreground data:WWTP Background data :Ecoinvent V2.1, chinese life cycle database(CLCD)	Tool : - Method : CML and e-balance (China)	EP,FWEP,HTP,OLDP,GWP,ADP,ACP	-
8	Padilla et al., 2017	Journal of Cleaner Production	Mexico and Canada	To compare the environmental performance of two WWTP technologies across all environmental impact categories in Latin America and the Caribbean	1 m^3 /day	Construction and operation	-	Foreground data: WWTP Background data : Ecoinvent, national database and literature	Tool: - Method: Impact 2002 and Recipe	MEP,GWP,FWEP, PM	-
9	Lorenzo-Toja et al., 2016	Science of the Total Environment	Spain	To set new benchmark regarding environmental performnace of wwtp (different climatic region - atlantic and mediterranian) for summer/winter	1 m^3 /day	construction, operation, sludge treatment	-	Foreground data:WWTP Background data :ecoinvent 2.2 & spanish electricity production	Tool : Simapro Method : CML 2001, USES-LCA (heavy metals and PPCPs)	EP,GWP,OLDP,HTP,MEP,FWEP	25,000PE (atlantic), 70,000PE (mediterranian)
10	Fang et al., 2016	Water Research	Denmark	Evaluation to capture necessary infrastructure additions, operational changes and reuse option for EBPR2 and sidestream microalgae cultivation in photobioreactor	1 m^3 /day	construction, operation, sludge treatment	Incinerator and microalgal fertiliser	Main database: Operating reports of an existing process, databases and model result. Background data: Ecoinvent (swiss and European market)	Tool : EASETECH Method : ILCD 2011, Usetox (human toxicity)	GWP, ACP, TEP, MEP, POF, Etox, Htc, Htnc, PM, RD	-
11	Pintilie et al., 2016	Journal of Cleaner Production	Spain	To identify and quantify the main environmental contributors derived from the treatment of urban	1 m^3 /day	Operation, sludge treatment	Agriculture	Foreground data:WWTP Background data :ecoinvent 3.1 and literature	Tool : Monte Carlo simulation	TA,CC,FE,ME,POF, MD,FD,OD,TT,WD ,CED	132,000PE

No	Reference	Journal	Country/ Area	Goal	FU	Processes considered	Sludge Disposal	Data Source/inventory	LCIA method & tool	Impact Category	Scale
				wastewater and water reclamation opportunities in Tarragona, Spain					Method : CML 2001,		
12	Meneses et al., 2015	Journal of Cleaner Production	Spain	The main environmental contributors derived from the treatment of urban wastewater and water reclamation opportunities in Tarragona, Spain	1 m^3 / year	Operation, sludge treatment	Agriculture	Main database: Benchmark simulation model2 Background database: Ecoinvent-sludge transportation and Spanish Energy for electricity production, literature	CML2000	AP,GWP,EP,PHO, DAR,ODP,TAETP	-
13	Piao et al., 2015	Journal of Cleaner Production	Korea	Evaluates several wastewater treatment plant (WWTP) processes including an integrated sludge management system and waste sludge disposal methods in a large city based on life cycle analysis (LCA) and economic efficiency analysis (EEA)	1 m^3 /day	Operation, sludge treatment	-	Foreground-operation of WWTP. Background- LCI database of Korean ministry of environment	Tool : Gabi Method : CML 2001,	AP,EP,GWP,HTP	CAS-340,000 m^3/d A_2O-680,000m^3/d MLE-80,000m^3/d
14	Risch et al., 2015	Water Research	France	To propose a holistic, life cycle assessment (LCA) of urban wastewater systems (UWS) based on a comprehensive inventory including detailed construction and operation of sewer systems and wastewater treatment plants (WWTPs)	1 day of operation	construction, operation, sludge treatment	-	Foreground-operation of WWTP. Background-Ecoinvent	Tool: Simapro Method: Recipe v1.07	TA,CC,FE,ME,POF, MD,FD,OD,TT,WD ,CED	5,200PE
15	Zang et al., 2015	Journal of Cleaner Production	China	Review of the LCA studies dealing	various	various	various	various	Usetox,Recip e,EDIP97,CM	HT,FET,FAET,MAE T,AP,EP,etc	various

No	Reference	Journal	Country/ Area	Goal	FU	Processes considered	Sludge Disposal	Data Source/inventory	LCIA method & tool	Impact Category	Scale
				with biological (activated sludge) WWTPs					L,IMPACT20 02,USES-LCA		
16	Yoshida et al., 2014	Water Research	Denmark	To investigate how the basis of inventory data affects the outcome of a WWTP LCA by using specific WWTP located in Denmark based on TRENS system	1 m^3 /day	Operation, sludge treatment	-	Foreground-operation of WWTP Background: European Pollutant Release EPRTR) and Transfer Regis- try, Danish emission monitoring, state of the art LCA, Ecoinvent v2.2	Tool : EASETECH Method : ILCD 2011,	GWP, AP, EP, PHO, ETP, PM	265,000 PE
17	Niero et al., 2014	Journal of Cleaner Production	Denmark	Compare four types of wastewater treatment plants	1 m^3 /day	Operation, sludge treatment	Incinerator, Agriculture	Foreground data:WWTP Background data :ecoinvent 2.2, ELCD & Danish Environmental Protection Agency	Tool : Monte-carlo Method : ILCD 2011, IPCC, Recipe, UseTox, CML2002	AD,AC,EU,GWP,O DP,HT,TE,MET,FE T,PO	Between 20,000PE to 100,000PE
18	Rodriguez -Garcia et al., 2014	Science of the Total Environme nt	Spain	Compare three side-stream technologies treating anaerobic digestion supernatant at two different levels, as independent levels processes and as part of a modelled WWTP	1 m^3 /day	Operation, sludge treatment	Landfill	Foreground data: WWTP Background data:ecoinvent 2.2, (Swiss centre for life cycle inventory 2012)	Tool : Biowin Method : CML2002	AD,AC,EU,GWP,O DP,HT,TE,MET,FE T,PO	-
19	Kalbar et al., 2013	Water and Environme nt Journal	India	Comparative 4 wwt tecnologies	PE/year	Operation, sludge treatment	Land application, etc.,	Foreground data:WWTP Background data :ecoinvent 2.2 and literature	Tool:- Method:CM L2 baseline 2000	AP,GWP,EP,FWAT ,HT,MAET,ADP,TE	ASP:200k PE, UASB-FAL:300k PE, CW:30k PE, SBR:100k PE
20	Coromina s et al., 2013	Water Research	Spain	Comprehensive review of 45 papers dealing with WWT and LCA	m^3 or ML	Various	Various	Data for the inventory is collected from lab or pilot facilities as well as real plants, estimation from experts, relevant literature and/or LCA da- tabases	Tool: Excel	Various	Various

No	**Reference**	**Journal**	**Country/ Area**	**Goal**	**FU**	**Processes considered**	**Sludge Disposal**	**Data Source/inventory**	**LCIA method & tool**	**Impact Category**	**Scale**
21	Daelman et al., 2012	Water Research	Netherlands	To determine the contribution of methane to the greenhouse gas footprint of a wastewater treatment plant and to suggest measures to curb methane emissions.	-	Operation, sludge treatment	-	Foreground data:1-year measurement campaign	-	GHG	36,0000PE
22	Gupta & Singh., 2012	Journal of Water Sustainability	India	Evaluate and quantify the greenhouse gases, mainly methane and nitrous oxide, emissions from the wastewater treatment system	-	Operation, sludge treatment	-	Foreground data: WWTP	Tool:- Method:IPCC 2006	GHG emissions	33 MLD
23	Corominas et al., 2012	Biotechnology and Bioengineering	Spain	To demonstrate the importance of using process-based dynamic models to better evaluate GHG emissions	-	Operation, sludge treatment	-	-	Tool: Benchmark Simulation Model Platform No. 2 (BSM2)	GHG emissions	-
24	Listowski et al., 2011	Journal of Water Sustainability	Korea	Development of a comprehensive impact assessment of gaseous emission from urban wastewater infrastructure and treatment facilities	-	Operation, sludge treatment	-	Foreground data: WWTP	Method: Technical Guidelines (DCCEE, 2010)	GHG emissions	-
25	Foley et al., 2010	Water Research	Australia	To analyse ten different wastewater treatment scenarios, covering six process configurations and treatment standards ranging from raw sewage to advanced nutrient removal	-	Construction, operation, sludge treatment	agriculture	Foreground data: WWTP Background data :ecoinvent 2.2 and literature	Tool: Biowin simulator	GHG	-

No	Reference	Journal	Country/ Area	Goal	FU	Processes considered	Sludge Disposal	Data Source/inventory	LCIA method & tool	Impact Category	Scale
26	Zhang et al., 2010	Bioresource Technology	China	Illuminate the env. the benefit of a WWT and reuse project using LCA model	1 m^3 /day	Construction, operation and demolition	-	Foreground data:WWTP Background data :Chinese database for construction material	Tool: -, Method: eco-indicator99	Energy use	-
27	El-Sayed et al., 2010	Cleaner Production	Egypt	Develop scenarios to improve the total environmental performance and the sustain-ability of Alexandria's urban water system	1 m^3	Operation	-	Foreground data:WTP & WWTP Background data :Literature	Tool : Simapro Method: Eco-indicator	Various	Various scale of water and wastewater treatment
28	Pasqualino et al., 2009	Environmental Science and Technology	Spain	Identifies the environmental impact of aWWTPin order to determine the environmental loads associated with the plant's operation and compare the total environmental impact of the various stages in both water and sludge treatment lines	1 m^3 /day	Operation, sludge treatment and disposal	Incinerator, Agriculture, landfill,compost plant	Foreground data:WWTP Background data:ecoinvent 2.2, (Spanish energy mix and the European model for transport and water)	Tool : SiSOSTAQUA Method : CML2002	AP,GWP,EP,PHO, DAR,ODP,ETP	144,000 PE
29	Renou et al., 2008	Cleaner Production	France	Evaluate the env. performnace of a full scale WWTP	1 m^3 of ww/ year	Operation, sludge treatment	Agriculture	Foreground data: operation, Background data: estimation(air emission) for chemical & electricity	Tool : Simapro Method:CML2000,Eco-indicator99, EDIP96,EPS, Eco-points97	GWP,ARD,AP,EP, TP	140,000 PE
30	Hospido et al., 2008	The International Journal of Life Cycle Assessment	Spain	The environmental evaluation of the most common technical options for urban wastewater.	PE	Operation, sludge treatment	-	Foreground data: operation, Background data: ecoinvent	Tool : Simapro Method:CML2000,	EU,OP, GWP,ACP,AC,PO, AD, TOXILOGICAL (HT,FET,MET,TET)	72,000 to 125,000 PE

No	Reference	Journal	Country/ Area	Goal	FU	Processes considered	Sludge Disposal	Data Source/inventory	LCIA method & tool	Impact Category	Scale
31	Köhler et al., 2007	Environmental Science and Technology	Germany	To provide a modular gate-to-gate inventory model for industrial wastewater purification in the chemical and related sectors	1 m^3	Operation	-	Foreground data: operation, Background data: ecoinvent	-		>500,000m^3
32	Halleux et al., 2006	Proceedings of LCE	Belgium	To assess the env. impact of WWTP by using LCA methodology	1 m^3	Construction, operation,sludge treatment	-	Foreground data: operation	Tool:- Method: eco-indicator 99, CML and Impact 2002+	HT,FWT,MET,TE,EU,AC,GW,FF	170,000PE

3 Assessing environmental impacts of large centralised wastewater treatment plants with combined or separate sewer systems in wet/dry seasons by using LCA

3.1 Introduction

Municipal wastewater treatment plants mainly deal with domestic wastewater, but it is a very common practice worldwide that storm runoff, through a combined sewer system, is combined with domestic wastewater for treatment. During wet weather, the untreated wastewater together with storm runoff could overload wastewater treatment plants (WWTPs), leading to overflow of wastewater directly into receiving waters. Even without overflow, rainfall still can affect environmental impacts from WWTPs by changing wastewater quality, quantity and treatment performance Life cycle assessment (LCA) is an efficient tool to evaluate environmental impacts from WWTPs. LCA is known as a technique for a holistic environmental assessment of a product or system. Since 1990s, LCA has been applied to the field of wastewater treatment (Corominas et al., 2013). In a study with LCA, Risch et al. (2018) reported that loads from storm events contributed significantly to eutrophication and ecotoxicity of WWTPs in freshwater. In addition, the compositions and strength of wastewater to WWTP change accordingly with the variation of rainfall which could affect wastewater treatment performance and the quality of effluent to the environment. Moderate to strong correlations were observed between rainfall intensity and pollutant concentrations in influent as well as rainfall intensity and volumetric flow rate of wastewater at 24 WWTPs in Georgia state, America with combined sewer systems (Mines et al., 2006). The square of correlation coefficient, R^2, between flow rate and average monthly rainfall ranged from 0.21 to 0.85, indicating that the flow rates of wastewater to WWTPs with combined sewer systems in different catchment areas were affected by rainfall intensity to different extents (Mines et al., 2007). It is believed that highly pollutant loaded influent in dry season can usually have satisfactory levels of pollutants removal while diluted influent by storm water is prone to cause operational issues (Lorenzo-Toja et al., 2015; Risch et al., 2018), and lower treatment efficiency. In many cases, however, lower effluent pollutant concentrations were reported from WWTPs during wet weather due to the dilution of wastewater (Joel, 2017; Li et al., 2017). Wastewater characteristics (e.g. concentrations of pollutants) in influent are one of the most important parameters to affect wastewater treatment efficiency, and effluent quality, leading to different environmental impacts from WWTPs. So far, the vast majority of LCA studies of WWTPs, however, were based on the dry weather conditions without considering rainfall effects, which does not enable a holistic view at the scale of the year with the temporal variability of environmental burdens. This is particularly important to the vulnerable receiving waters as dry weather-based environmental impact assessment might overestimate or underestimate the environmental burdens such as eutrophication and ecotoxicity.

Due to the importance of rainfall effects on flow rate and pollutant concentrations of wastewater influent, treatment performance in WWTPs and pollutant concentrations in effluent, a few of studies evaluated the effects of rainfall on the environmental impacts of wastewater treatment plants. Nevertheless, conclusions from these studies are not consistent. For example, for a Spanish municipal WWTP, Moreira et al. (2004) concluded that the differentiation of wet (humid) and dry seasons for environmental analysis was not necessary because the data variability in each season had turned out to be more significant than the variation caused by rainfall. Lorenzo-Toja et al. (2016), however, found that Atlantic region with the highest rainfall resulted in the least environmental impact when they studied WWTPs with LCA in different regions of Spain with different rainfall intensity (i.e. from 300mm to >1000mm). Results from three-year data in a WWTP, China, with a subtropical monsoon climate showed five chosen impacts, (e.g. abiotic depletion potential (ADP), acidification potential (AP), eutrophication potential (EP), global warming potential (GWP) and photochemical ozone creation potential (POCP) increased almost linearly with monthly precipitation when the monthly precipitation was below 200mm/month (Li et al., 2017). This result indicates higher environmental burdens in the wet season. These contradictory results about rainfall effects (i.e. no impact, positive impact or negative impact) indicate that some key factors that might influence environmental impacts by LCA are still not fully understood. Some possible factors are identified as below. The rainfall effects on WWTPs should be closely related to how much it can cause the changes of influent characteristics including flow rate and concentrations instead of the absolute precipitation amount. Secondly, one of the most important factors affecting the efficiency of WWTPs has been revealed to be the characteristic of the influent particularly wastewater strength (Lorenzo-Toja et al., 2015). Rainfall during wet weather does not only affect wastewater strength in influent and effluent by dilution, but also treatment performance. These in turn affect environmental impacts from WWTPs. Thirdly, choosing different functional units might lead to different LCA results on the study of rainfall as influent wastewater quality is changed by rain, but not reflected by some functional units. Per m^3 treated wastewater is a mostly used functional unit for LCA analysis of WWTPs. However, it is argued that per m^3 treated wastewater could not reflect the influent quality or wastewater treatment efficiency in WWTPs (Corominas et al., 2013), making the comparison between two systems with different influent quality or different wastewater treatment efficiency difficult. Instead, per kg pollutant removed such as per kg of chemical oxygen demand equivalent (COD-eq.) removed (Wang et al., 2018) or per kg of phosphate (PO_4^{3-}eq) removed (Rodriguez-Garcia et al., 2011) could be a better functional unit when considering different influent quality or treatment efficiency for the comparative studies. Per population equivalent (P.E.) could also be considered when reflecting the difference of flow rate of influent and the associated load (Gallego et al., 2008; Kalbar et al., 2013). The comparison between two different functional units, e.g. per m^3 treated wastewater and per kg PO_4^{3-}eq removed, resulted in contrasting results in terms of main environmental impacts (Rodriguez-Garcia et al., 2011), highlighting the importance of the selection of functional unit in different scenarios. It is thus suggested that LCA studies on WWTPs are preferably carried out using more than one functional unit to deepen understanding of the system under study and to avoid misleading conclusions (Zang et al., 2015). For the study of rainfall effects on the environmental burdens from WWTPs, assessing different functional units is important because the influent quality and quantity changed by rainfall could affect the treatment performance due to the dilution of the influent and the disturbance to biological treatment.

This study aims to investigate the influence of rainfall on the environmental impacts of WWTPs by using LCA in two scenarios, i.e. large centralised WWTPs with high strength wastewater and low strength wastewater, respectively, but with similar rainfall effects on influent flow rate. Meanwhile, different functional units would be studied to evaluate their influence on LCA results in the scenarios with/without rainfall.

3.2 Materials and methods

3.2.1 The selection and description of two case studies

A pre-screening assessment in this study found that the correlation coefficients between monthly rainfall intensity and influent flow rate of wastewater to two WWTPs, i.e. a Malaysian Sewage Treatment Plant (MSTP) in Penang, Malaysia, and Millbrook Wastewater Treatment Work (MWTW), in Southampton, the United Kingdom, are similar. In addition, the strength of wastewater in MSTP and MWTW are distinctive. Thus, these two WWTPs were selected to study the effects of rainfall on the environmental impacts of WWTPs with different wastewater strength.

MSTP receives domestic wastewater of 800,000-population equivalent (PE) with a flow rate varying between 111,191 and 149,584 m^3/day throughout the year 2016. Wastewater enters into MSTP from a separate sewer system. MSTP mainly consists of grit and grease screening, sequencing batch reactor for pollutant removal, gravity belt thickener, anaerobic sludge digester, and biosolids dewatering. This type of WWTP is widely used in Malaysia and is considered as a typical wastewater treatment plant. The treated water is discharged into the river nearby, while the sludge produced is sent to a landfill located 47 km away. The operation data in 2016 was used in this study. Daily rainfall data in 2016 was retrieved from the Malaysian Meteorology Department in the MSTP catchment area. The average monthly rainfall and temperature data from the year 2010 to 2016 were obtained from the web source: (www.worldweatheronline.com) for the comparison of the seasonal pattern. MWTW with a combined sewer system has a wastewater treatment capacity for 140,000 PE with a flow rate varying between 35,028 and 49,563 m^3/day throughout the year 2017. This facility includes primary settlement, Bardenpho process for COD and nitrogen removal, secondary settlement, sludge thickening, dewatering and anaerobic digestion incorporated with biogas collection and energy recovery systems. Methanol is dosed as an external carbon source for denitrification, and polymer is used for thickening and centrifuges while lime is used for sludge disinfection. Biosolids after digestion are sent for various land application. Rainfall and temperature data in the year 2013 to 2017 in Southampton was obtained from the weather website (www.worldweatheronline.com). **Figure 2** shows the schematic diagrams of MSTP and MWTW.

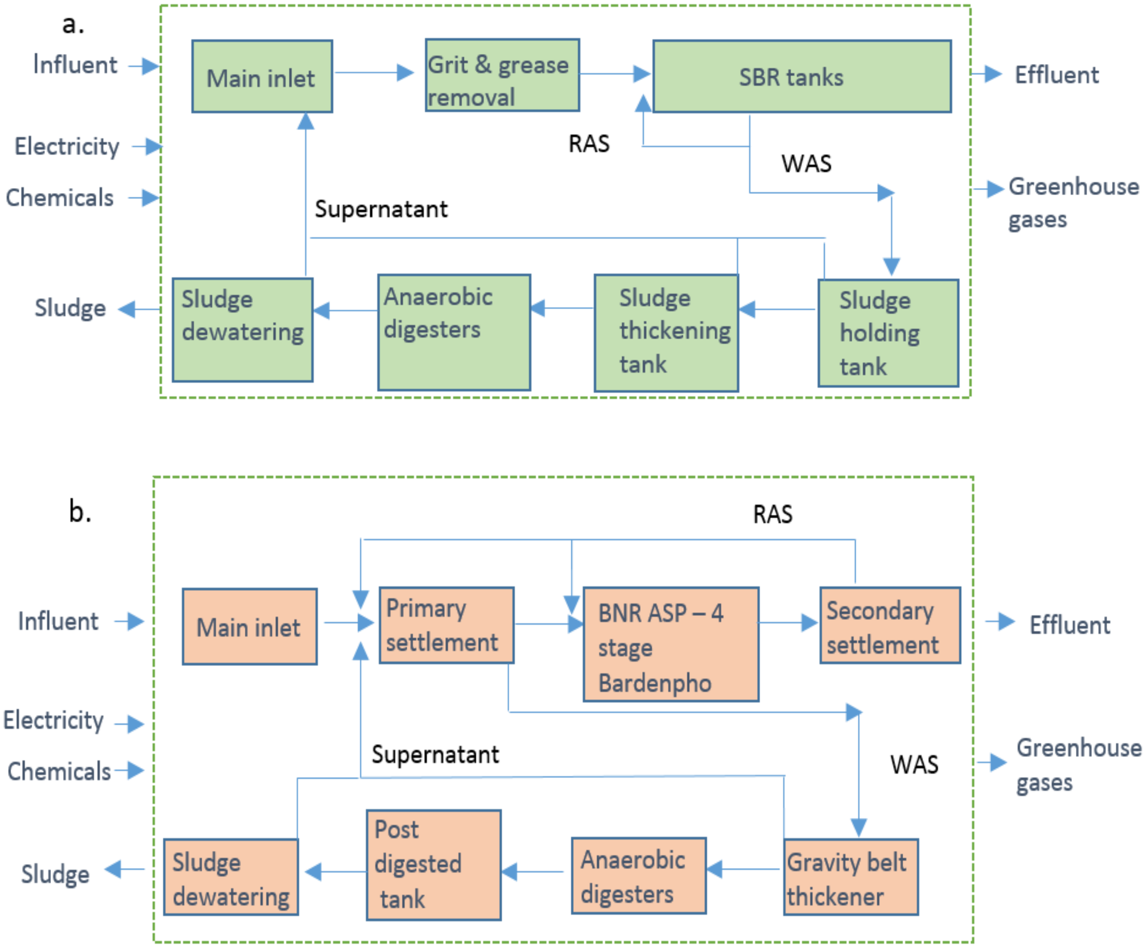


Figure 2. The system boundary of: a) Malaysian STP and b) Millbrook WTW in this study

Note: (SBR = sequencing batch reactor; BNR-ASP = biological nutrient removal-activated sludge process; RAS = return activated sludge, WAS = waste activated sludge)

Table 6 provides wastewater quality and quantity entering into the MSTP in the year 2016 and MWTW in the year 2017. The storm flow (maximum) was 1.36 times of the dry weather flow (minimum) in MSTP, which is similar to 1.42 times in MWTW. Influent mass load (kg/month) entering into the MSTP and MWTW in the dry seasons were at least 1.6 and 2.2 times, respectively, of that in wet season. The ratios of mass and pollutant concentration in dry to wet season were higher in MWTW probably because various pollutants were carried in by stormwater runoff to the treatment plant through a combined sewer system in rainy days (Li et al., 2017). High strength or low strength wastewater is the measure of concentration of pollutants (e.g. BOD, COD, TSS and TN) in the sewage. According to the review concerning wastewater strength in developed countries and developing countries by Gallego-Schmid & Tarpani, 2019, average influent BOD, COD and SS concentration is 251, 551 and 252 mg/L respectively, in developed countries are higher than those in developing countries with 209, 410 and 190 mg/L, respectively. In addition, BOD, COD, SS, N, and

P vary in wide ranges in either developed or developing countries. The strength of wastewater into each WWTP in this study falls well within the pollutant concentration range in developing and developed countries as reported by Gallego-Schmid & Tarpani, 2019, respectively. For example, the strength of wastewater into MWTW, UK is at the upper limit of the range in developed countries, while the strength of wastewater into MSTP, Malaysia is at the bottom limit of the range in developing countries. These two WWTPs are thus ideal for the study if wastewater strength plays a role when studying rainfall effects on environmental impacts.

Table 6. The fluctuations of pollutant concentration, mass load and flow rate of wastewater into Malaysian STP (year 2016) and Millbrook WTW (year 2017), respectively

	Concentration (mg/L)			Mass (kg/month)		
Malaysian STP	Maximum	Minimum	Ratio	Maximum	Minimum	Ratio
Flow (m^3/month)	$4.55x10^6$	$3.34x10^6$	1.36	32gBOD/head	16gBOD/head	2.00
BOD_5	141.00	69.00	2.04	$5.25x10^5$	$2.59x10^5$	2.03
COD	530.40	229.50	2.31	$1.16x10^6$	$4.65x10^5$	2.49
TSS	150.00	49.00	3.06	$5.82x10^5$	$1.75x10^5$	3.33
TN	32.50	20.00	1.63	$1.30x10^5$	$8.10x10^4$	1.60
Millbrook WTW	Maximum	Minimum	Ratio	Maximum	Minimum	Ratio
Flow (m^3/month)	$1.49x10^6$	$1.05x10^6$	1.42	100gBOD/head	19gBOD/head	5.26
BOD_5	459.00	91.8	5.00	$5.00x10^5$	$1.21x10^5$	4.13
COD	1215.00	327.50	3.71	$1.41x10^6$	$4.35x10^5$	3.24
TSS	625.00	168.00	3.72	$8.04x10^5$	$1.96x10^5$	4.10
TN	71.00	35.40	2.00	$9.13x10^4$	$4.13x10^4$	2.20

Notes: BOD5 (five-day biochemical oxygen demand); COD (chemical oxygen demand); TSS (total suspended solids); TN (total nitrogen)

3.2.2 Correlation analysis of wastewater indicators

In this study, twelve months of operation data in MSTP in the year 2016 and in MWTW in the year 2017 were evaluated with a statistical method to correlate different parameters (Mines et al., 2007; Li et al., 2017). Average monthly rainfall was plotted against average monthly influent flow rate in both plants. Trend lines and the square of correlation coefficient R^2 were determined using linear regression analysis in both plants. In addition, the Pearson coefficient's correlation analysis between rainfall intensity, sewage temperature, power consumption, volumetric flow rate and other pollutant parameters in influent at a monthly basis was conducted using SPSS software v24.

3.2.3 Life cycle analysis

3.2.3.1 Goal and scope

The goal of this study is to investigate and compare the effect of rainfall from dry season and wet season on the environmental impacts from large centralised municipal wastewater treatment plants with different influent wastewater strength. Since this study focuses on rainfall effect on the life cycle environmental impacts from WWTP operation, construction and demolition stages, as well as landfilling sludge are not considered because they are same regardless of rainfall. However, transport of sludge to landfill was included. For this selection, 'gate-to-gate' analysis is adopted which begins with the wastewater influent physically entering into WWTPs, and ends with the effluent discharged into water bodies and transport of biosolids to landfill. The illustrated system boundary for this LCA - WWTP study is shown in **Figure 3.** In general, the system boundary is limited to wastewater treatment operations with wastewater flow rate and pollution loads in a foreground system, and energy and chemical consumption (e.g. electricity and chemical production) in a background system.

3.2.3.2 Functional unit

1m^3 of treated wastewater was used as a functional unit first, which is widely adopted for LCIA in WWTPs (Piao et al., 2016; Lorenzo-Toja et al., 2016; Rahman et al., 2016; El-Sayed et al., 2010; Niero et al., 2014). It is believed that the functional unit as per m^3 of treated wastewater, however, does not consider the change of wastewater flow rate to WWTPs (Piao & Kim, 2016) or wastewater treatment efficiency. Therefore, functional unit 2 (FU2) defined as 1 kgPO_4^{3}-eq removed was used as well for a better comparison with the change of wastewater flow rate. FU2 was also used by

(Rodriguez-Garcia et al., 2011) and (Comas Matas, 2012). The eutrophying substances, i.e. chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) in wastewater were converted to $kgPO_4^{3}$-eq using the characterisation factor from eutrophication potential impact category as defined in the CML-IA baseline v3.04 methodology.

3.2.3.3 Life cycle inventory (LCI)

The operation data of MSTP in 2016 and the data of MWTW in 2017 were considered in this study. The life cycle inventory consists of monthly electricity consumption, monthly volume of wastewater treated and daily influent and effluent characteristics. The life cycle inventory (LCI) consists of following parameters as shown in **Table 7**.

Table 7. Summary of the data sources for life cycle inventory

Indirect inputs of resources including energy and chemical consumed for wastewater treatment and sludge treatment, as well as sludge transportation. Background data were obtained from the Ecoinvent v3.3 database as described below:		Direct influent pollutants as inputs and effluent pollutants as outputs which consist of COD, nitrogen and phosphorus.	Direct gases emissions from the plant as outputs, which mainly include CO_2, CH_4 and N_2O. They were calculated according to the Intergovernmental Panel on Climate Change guideline (IPCC, 2006) based on the 100-year time horizon.
a.	Electricity production: Malaysia and the United Kingdom were selected from the Ecoinvent v3.3 database.		a. N_2O was mainly generated from biological nitrogen removal process
b.	Chemical production: Data on the processes of methanol and lime were selected from the ELCD and Ecoinvent v3.3 database. For polyelectrolytes, a similar production process for acrylonitrile was taken from the Ecoinvent v3.3 (Rodriguez-Garcia et al., 2011; Lorenzo-Toja et al., 2016).		b. CH_4 was from anaerobic wastewater and/or sludge treatment (Masuda et al., 2015)

c. A lorry with a capacity of 3.5-7.5 metric ton was selected as transport vehicle for the disposal of sludge and wastes produced from both WWTPs. Chemical transportation to the site is excluded due to small proportion to environmental impact (Lorenzo-Toja et al., 2016) with less than 5% emission compared to the sludge transportation value (Rodriguez-Garcia et al., 2011)

All inventory data are provided in **Table 8** (FU1) and in **Table 9** (FU2). The LCI for FU1 and FU2 were calculated as example below:

For FU1: Daily inventory components/sources such as electricity (kWh), chemicals (kg), transportation (t.km), direct emission of $CO_2/CH_4/N_2O$ (kg) and direct emission of COD/TN/TP (kg) were divided by daily inflow to WWTP (m^3). For example:

a. LCI for electricity equals to: kWh.day /m^3.day.

For FU2: Daily inventory components/sources such as electricity (kWh), chemicals (kg), transportation (t.km), direct emission of $CO_2/CH_4/N_2O$ (kg) and direct emission of COD/TN/TP (kg) were divided by mass pollutants removed per day in (kgPO_4^{3-}eq./day). For example:

b. LCI for chemicals consumption equals to: kg.day / kgPO_4^{3-}eq.day.

Table 8. Life cycle inventory (LCI) data in Malaysian STP and Millbrook WTW according to per functional unit (1 m^3 of treated wastewater)

Inventory components	Malaysian MSTP		Millbrook MWTW		Unit/ m^3
	Dry season[A]	Wet season[B]	Dry season (summer)[C]	Wet season (winter)[D]	
1.Electricity consumption	2.58E-01± 9.0E-2	2.38E-01± 1.7E-2	6.11E-01± 5.9E-2	4.86E-01± 4.2E-2	kWh
2.Transportation of sludge and waste	6.48E-03± 1.9E-6	6.47E-03± 4.7E-7	2.70E-02± 9.0E-3	2.62E-02± 8.0E-3	t.km
<u>Polymer consumption</u>					
3.Methanol	-	-	7.12E-03± 4.0E-3	6.93E-03± 3.0E-3	kg
4.Polyelectrolyte	5.15E-04± 1.8E-8	5.15E-04± 1.3E-8	3.62E-03± 7.0E-3	3.52E-03± 6.8E-3	kg
5.Lime	-	-	8.19E-02± 8.4E-3	7.97E-02± 7.5E-3	kg
<u>Emission to air</u>					
6.Carbon dioxide (biogenic)[E]	9.69E-02± 7.8E-3	8.17E-02± 1.5E-3	3.84E-01± 5.5E-2	3.07E-01± 4.1E-2	kg
7.Methane, CH_4	1.11E-03± 3.3E-4	1.09E-03± 8.6E-4	2.40E-03± 9.6E-4	2.00E-03± 8.2E-4	kg
8.Dinitrogen monoxide, N_2O	4.87E-04± 7.4E-5	4.40E-04± 2.1E-5	5.40E-04± 8.3E-5	5.10E-04± 7.1E-5	kg
<u>Emission to water</u>					
9.Total COD	5.10E-02± 8.0E-3	4.28E-02± 1.0E-2	4.61E-02± 1.0E-2	3.20E-02± 9.0E-1	kg
10.Total nitrogen	1.08E-02± 2.5E-3	7.63E-03± 2.2E-3	9.00E-03± 2.5E-3	6.95E-03± 1.8E-3	kg
11.Total phosphorus [F]	2.20E-03	1.10E-03	1.1E-03	8.00E-04	kg
<u>Combined sewer overflow (CSO)</u>					
12.Total COD [G]	x	x	x	4.34E-01	kg
13.Total nitrogen [G]	x	x	x	2.55E-02	kg
14.Total phosphorus [G]	x	x	x	4.00E-03	kg

[A] From January to March 2016; [B] From September to November 2016

[C] From June to July 2017; [D] From January to February 2017

[E] Carbon dioxide emission from the biological process in WWTP is considered biogenic origin by IPCC guideline and was not included in the LCA analysis (IPCC Guidelines for National Greenhouse Gas Inventories, 2006)(IPCC Guidelines for National Greenhouse Gas Inventories, 2006)

[F] 1 set of TP data

[G] 1 set of inventory data from Millbrook WTW management for CSO

Table 9. Life cycle inventory (LCI) data of Malaysian STP and Millbrook WTW per functional unit 2 (eutrophication reduction – 1 kg PO_4^{3-} eq.)

Inventory components	Malaysian MSTP		Millbrook MWTW		Unit/ kgPO_4^{3-}eq
	Dry season[A]	Wet season[B]	Dry season (summer)[C]	Wet season (winter)[D]	
1.Electricity consumption	1.80E+01	1.47E+01	9.15E+00	1.28E+01	kWh
2.Transportation of sludge and waste	4.52E-01	3.99E-01	4.03E-01	6.91E-01	t.km
Polymer consumption					
3.Methanol	-	-	5.40E-02	9.27E-02	kg
4.Polyelectrolyte	3.60E-02	3.18E-02	1.06E-01	1.83E-01	kg
5.Lime	-	-	1.22E+00	2.10E+00	kg
Emission to air					
6.Methane, CH_4	1.46E-01	1.15E-01	1.64E-01	2.20E-01	kg
7.Dinitrogen monoxide, N_2O	2.04E-02	1.80E-02	3.00E-02	3.96E-02	kg
Emission to water					
8.Total COD	3.37E+00	2.51E+00	6.55E-01	8.00E-01	kg
9.Total nitrogen	7.00E-01	4.47E-01	1.26E-01	1.73E-01	kg
10.Total phosphorus	1.46E-01	6.44E-02	1.57E-02	2.00E-02	kg

[A] From January to March 2016; [B] From September to November 2016

[C] From June to July 2017; [D] From January to February 2017

3.2.3.4 Life cycle impact assessment (LCIA)

Life cycle impact assessment (LCIA) was conducted with the characterisation factors from CML-IA baseline v3.04 methodology. As wastewater treatment plants mainly generate climate change-related impacts and environmental quality issues (Renou et al., 2007), seven characterisation impact categories such as eutrophication potential (EP), ozone layer depletion potential (ODP), freshwater ecotoxicity potential (FEP), human toxicity potential (HTP), global warming potential

(GWP), abiotic depletion (fossil fuel) potential (ADFP) and acidification potential (AP) were chosen as the main assessment categories.

3.2.3.5 Life cycle interpretation and sensitivity analysis

The LCA results were interpreted to assess the contribution of each component in the inventory, e.g. electricity consumption, chemicals consumption, transportation and direct emission of pollutants and GHGs, to each environmental impact category. MSTP and MWTW are expected to have 30 to 40-year operational lifetime. Within the 40 years operation of MSTP and MWTW, there will be variation in the mass load of pollutants which could affect the electricity consumption, chemicals consumption and nutrient concentration in the effluent. Sensitivity analysis was thus conducted to assess how the ±10% variations in inventory data such as electricity consumption, nitrogen and phosphorus concentrations in effluent, and chemicals consumption, affect LCA impact category results. This sensitivity analysis could identify the accuracy of inventory data of wastewater treatment plant with a long design life. FU1, i.e. per m^3 treated wastewater was selected for this analysis to facilitate the comparison with the results from other studies.

3.3 Results and discussion

3.3.1 Multivariate correlation between various parameters of wastewater in two WWTPs

Rainfall affects the wastewater flow rate to WWTP particularly with a combined sewer system receiving storm runoff. It can further affect operation in WWTP and quality of effluent to water bodies. A positive linear relationship between the average monthly rainfall intensity and the average monthly influent flow rate into MWTW, Southampton, UK, with a combined sewer system was found as shown in **Figure 3a.** This is plausible as the high rainfall intensity directly results in the storm runoff into the sewer system, and thus increases the influent flow rate. This result is consistent with those reported in other geographical areas with combined sewer systems. For example, Li et al. (2017) reported a linear relationship between influent flow rate to WWTP and rainfall precipitation with a combined sewer system in Yangtze, Eastern China, where the average yearly precipitation is 1100mm, comparable with 879 mm in Southampton, UK, in this study. Both WWTPs have similar P.E., e.g. around 186,000 –200,000. However, the influent flow rate to WWTP increases by 1480 m^3 per mm precipitation in the Yangtze, China, while by 2793 m^3/mm, twofold higher, in this study to MWTM in Southampton, UK. Mines et al. (2006) correlated the rainfall intensity and influent flow rates to 24 WWTPs with combined sewer systems in Georgia state, America, and found similar linear relationships, but the slopes of regression lines range from 540 to 8100 m^3/mm precipitation in different locations. This is mainly because that the change in flow rates to WWTP caused by rainfall with a combined sewer system relies on both the precipitation amount and hydrogeologies e.g. soil condition for filtration (Metcalf & Eddy, 2004), sewer pipe conditions, runoff from the city, and the catchment area. The increase rates in influent rate by rainfall to WWTPs with a combined sewer system in different catchments vary, but a linear relationship can well describe the effects of precipitation on the flow rate of influent to WWTP.

For WWTPs with a separate sewer system, a general impression is that rainfall should not cause much change in wastewater flow rate because storm runoff is collected separately. Thus, there lacks studies on the rainfall effects on influent flow rate to WWTPs with separate sewer systems. In this study However, a linear relationship was established as well between rainfall intensity and flow rate to MSTP, Penang with a separate sewer system (**Figure 3b.**), and a similar increasing rate as MWTW in Southampton, UK, with a combined sewer system, was found. This contrasts with the general impression that rainfall does not cause much flow rate change to WWTP with a separate sewer system, indicating the complexity of the actual situation with regard to the effect of rainfall intensity on the influent flow rate to WWTPs. In this study, the precipitation in MSTP, Penang, is 2200mm yearly, which is much higher than 879mm in the catchment area with MWTW, Southampton. It is thus speculated that water saturation in the soil in Penang, Malaysia, might be higher, leading to more infiltration to the sewer system although it is meant to collect domestic wastewater only. The investigation on the specific reasons for this is beyond the scope of this study, but results here clearly suggest that combined or separate sewer system is not the only decisive factor to determine the effect of rainfall on influent flow rate to WWTPs. To the best of my knowledge, the findings here about rainfall effect on influent flow rate to a WWTP with a separate sewer system are reported for the first time. The comparison of rainfall effect on influent flow rate to WWTPs with a separate sewer system and a combined sewer system in two locations were investigated for the first time, and similar results were obtained. This highlights the necessity to study the rainfall effect on WWTPs even with a separate sewer system.

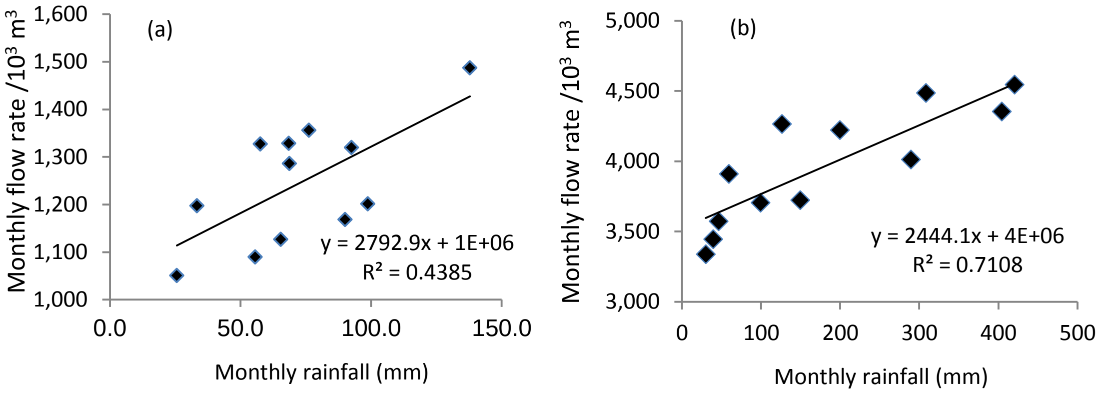


Figure 3. The linear relationship between the average monthly influent volumetric flow rate and the average monthly rainfall intensity for: a) Millbrook WTW and b) Malaysian STP

The great dependence of influent flow rate on rainfall can lead to the changes of wastewater quality (pollutant concentrations) and quantity (flow rate), thus further affects the environmental impact of WWTPs due to the changed power consumption for pumping and aeration, and treatment performance. To understand the relationship between different parameters, the correlations between rainfall, temperature, power consumption, and wastewater influent characteristics were carried out and the results are shown in **Table 10**. When the Pearson correlation coefficient, r moves from 0 to ±1.0, the correlation becomes stronger. In detail, r with 0 to ±0.49 indicate weak linear relationship, while r with ±0.5 to ±1.0 indicate moderate to strong linear relationship (Li et al., 2017). From **Table 10**, a strong correlation between rainfall and the influent flow rate was found with a Pearson coefficient of 0.84 for MSTP, Penang, with a separate sewer system and 0.66 for MWTW, Southampton, with a combined sewer system. Like linear regression, the correlation is even stronger in MSTP, Penang, with a separate sewer system, which is probably due to higher rainfall intensity and larger catchment area in Penang, Malaysia. In addition, there is a moderate correlation between rainfall intensity and power consumption in MSTP with r as 0.54 but only 0.07 in MWTW. It is believed that a weak correlation between rainfall and power consumption for MWTW was from lower precipitation in Southampton at an average monthly of 78.5mm.

Both plants exhibited negative correlations between influent flow rate, and influent BOD_5, TCOD, TSS and TN concentrations, indicating a dilution of wastewater by rainfall. This result is in agreement with the findings reported for 24 WWTPs in the USA mainly with combined sewer systems by Mines et al. (2006), who found low to moderate negative correlation between influent flow rate and concentrations of BOD and TSS in the influent. Although rainfall dilutes wastewater in terms of pollutant concentrations, the correlations between rainfall and pollutant mass load (e.g. kg/day) in both Malaysian STP and MWTW are mostly positive. This suggests increased total pollutant mass loads in rainy days, especially in MSTP, due to the pollutants taken in by runoff, which increases the treatment burdens to WWTPs. The correlation between influent flow rate and mass loads of pollutants in MSTP with a separate sewer system was relatively weaker probably due to pollutant filtration by soil before infiltration. These results further indicate the complexity of the correlation between influent flow rate and pollutants (Nesmerak & Blazkova, 2014).

In addition, mass loads and pollutant concentrations in influent were highly correlated to the energy consumption in both plants. In Malaysian STP, the correlations were high between power consumption and pollutant loads of BOD_5m, TCODm, TSSm and TNm with r as 0.81, 0.82, 0.69, and 0.8, respectively, while there were relatively moderate values of correlation, e.g. r, between 0.41 and 0.69 in MWTW. The power consumption in WWTPs is not fixed the year around, and WWTP uses more energy when it deals with higher pollutant mass loads. It seems that MSTP with bigger capacity (i.e. for an average of 588,000 PE) is more affected. Both plants also exhibited a positive moderate correlation between power consumption and influent pollutant concentrations (mg/L), which further proved the good correlation between power consumption and the pollutant characteristics. The correlation between influent flow rate and power consumption was moderate with r of 0.44 in MSTP, while it is only 0.03 in MWTW. With the high rainfall intensity in the MSTP catchment area, the treatment plant consumed higher energy with higher inflow while there was little power consumption change with the inflow change in MWTW.

Table 10. Pearson correlations between average monthly sewage temperature, rainfall, the flow rate of wastewater, power consumption, and influent pollutant concentrations and mass loads in Malaysian STP and Millbrook WTW

Malaysian STP	Temperature	Rainfall	Inflow	Power	BOD_5m	TCODm	TSSm	TNm
Temperature	1.00	0.02	0.21	0.33	0.28	0.21	-0.18	0.07
Rainfall	0.02	1.00	0.84	0.54	0.26	0.42	0.28	0.58
Inflow	0.21	0.84	1.00	0.44	0.13	0.23	0.06	0.51
Power	0.33	0.54	0.44	1.00	0.81	0.82	0.69	0.80
BOD_5c	0.16	-0.15	-0.34	0.57	0.89	0.76	0.43	0.32
TCODc	0.15	0.11	-0.13	0.69	0.89	0.93	0.54	0.46
TSSc	-0.25	0.03	-0.22	0.54	0.43	0.46	0.96	0.46
TNc	-0.11	0.07	-0.14	0.60	0.61	0.58	0.71	0.78

Millbrook WTW	Temperature	Rainfall	Inflow	Power	BOD_5m	TCODm	TSSm	TNm
Temperature	1.00	-0.40	-0.37	0.61	0.10	0.5	0.55	0.30
Rainfall	-0.40	1.00	0.66	0.07	0.25	0.03	-0.09	0.06
Inflow	-0.37	0.66	1.00	-0.03	0.17	-0.02	-0.05	0.25
Power	0.61	0.07	-0.03	1.00	0.48	0.69	0.57	0.41
BOD_5c	0.24	0.02	-0.14	0.47	0.95	0.61	0.37	0.34
TCODc	0.56	-0.12	-0.24	0.64	0.55	0.97	0.91	0.79
TSSc	0.60	-0.20	-0.23	0.56	0.33	0.93	0.98	0.85
TNc	0.47	-0.21	-0.17	0.43	0.36	0.90	0.95	0.91

Notes: Highlighted values in grey is the correlation values that are higher than ±0.5. (Inflow = influent flow rate, power = electricity consumption, m = mass load, c = concentration)

Finally, it is found that influent flow rates correlate negatively to the effluent quality in both MSTP and MWTW (**Table 11**), suggesting that the reduction in the concentrations of pollutants in effluent is also from the dilution by rainfall. These results indicate that for either combined or separate sewer system, rainfall does affect wastewater influent flow rate, wastewater influent and effluent quality, and power consumption, which further influence the overall environmental impact from WWTPs. Therefore, using one set of data from industry-standard simulation software or from short-period sampling to do static environmental impact assessment with LCA might cause some bias. Thus, it is very necessary to split the whole year as a wet and dry season to see how rainfall in wet and dry seasons with different sewer systems affects wastewater treatment and environmental impact with real dynamic data to provide a basis for further methodology development and validation, as well as the improvement of the LCA practice.

Table 11. Pearson correlations between average monthly sewage temperature, rainfall, the inflow of wastewater, power consumption and effluent pollutant concentrations at Malaysian STP and Millbrook WTW

MSTP	Temperature	Rainfall	Inflow	Power	BOD_5	COD	TN	TSS
Temperature	1.00							
Rainfall	0.02	1.00						
Inflow	0.21	0.84	1.00					
Power	0.33	0.54	0.44	1.00				
BOD_5	-0.23	-0.23	-0.39	-0.10	1.00			
COD	-0.35	-0.02	-0.32	0.08	0.77	1.00		
TN	-0.75	-0.28	-0.36	-0.79	0.19	0.12	1.00	
TSS	-0.32	-0.35	-0.53	-0.30	0.53	0.75	0.37	1.00
MWTW								
Temperature	1.00							
Rainfall	-0.19	1.00						
Inflow	-0.39	0.63	1.00					
Power	0.57	0.16	0.14	1.00				
BOD_5	0.62	-0.61	-0.50	0.49	1.00			
COD	0.34	0.30	-0.02	0.38	0.08	1.00		
TN	0.25	0.19	-0.16	0.04	0.07	0.38	1.00	
TSS	0.04	0.29	0.01	0.30	0.04	0.54	0.05	1.00

3.3.2 Rainfall effects on wastewater quality, energy and chemical consumption in two WWTPs

The monthly rainfall intensity versus months in 2016 was plotted to identify wet and dry seasons in MSTP catchment area, (**Figure 4**), from which dry season was identified from January to March with the lowest rainfall intensity while wet season from September to November. To validate the consistency of wet and dry seasons over years, the average rainfall intensity from 2010-2016 was further analysed to identify wet and dry seasons. The results from 2010-2016 are consistent with the year 2016's rainfall pattern, indicating that 2016 is a year with a typical dry season and wet season. An earlier study on Penang in the year 2000 (Ahmad Jailani, 2004) showed the same wet and dry seasons. Similarly, the monthly rainfall pattern in 2017 was compared with that from the year 2013 to 2017 (**Figure 5**) in the MWTW catchment area, and June to July was identified as dry season (summer as well) while January to February is wet season (winter as well).

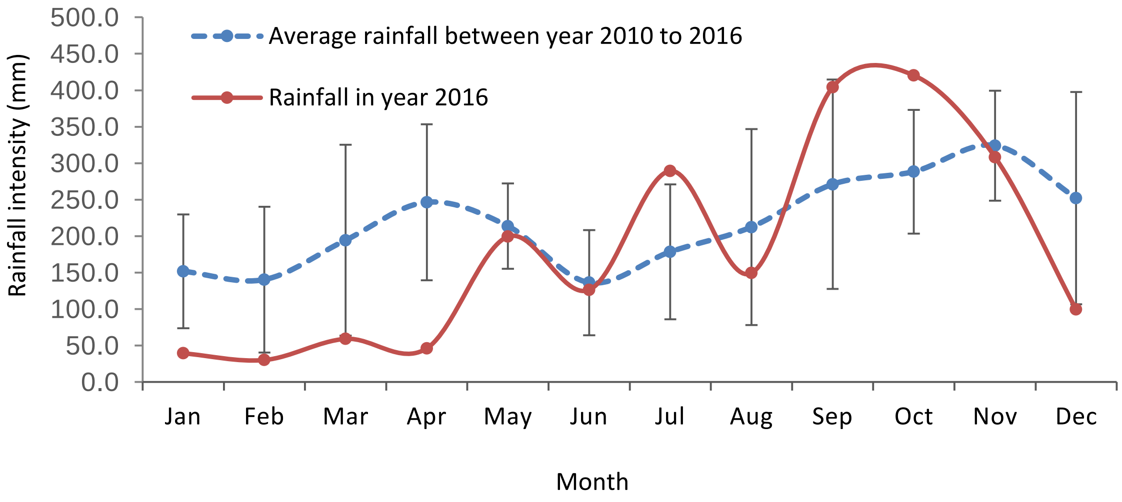


Figure 4. The comparison of average monthly rainfall data for Malaysian STP in the year 2016 (from Malaysian Meteorological Department) and average rainfall data from the year 2010 to 2016 with standard deviation (from international weather website: www.worldweatheronline.com).

Note: Dry season was identified from January to March while the wet season was from September to November in Penang, Malaysia. The average air temperature in MSTP (Malaysia) is consistent throughout the year ranging from 26 ^{o}C to 30 ^{o}C.

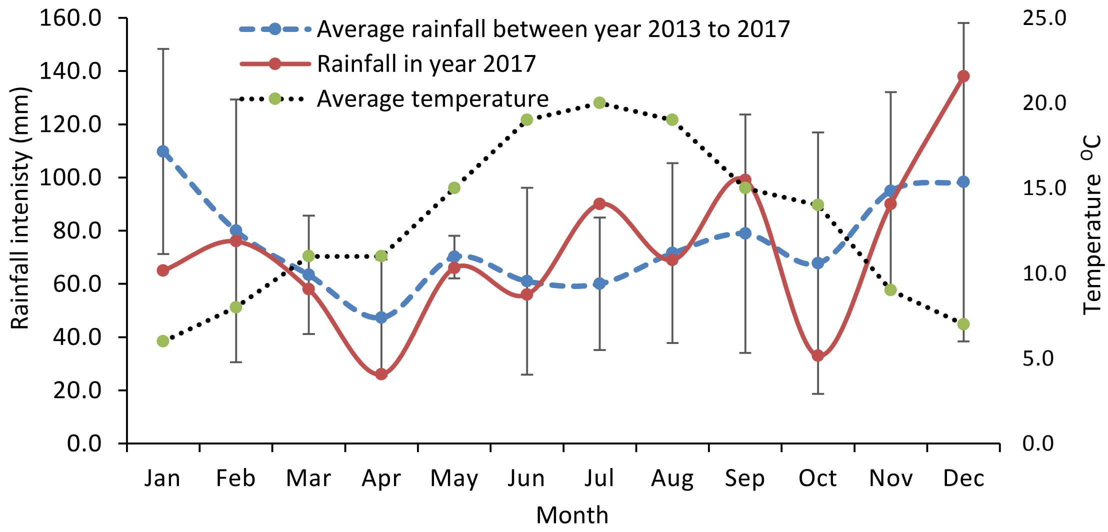


Figure 5. The comparison of average monthly rainfall data for Millbrook WTW in the year 2017 and average rainfall data from the year 2013 to 2017 with standard deviation (from international weather website: www.worldweatheronline.com)

Note: Winter (wet) was identified in January and February while summer (dry) was in June and July in Southampton, UK

Table 12 shows the comparison between dry and wet seasons in terms of influent and effluent pollutant concentrations and other parameters in MSTP and MWTW. Higher flow rate and power consumption were found in the wet season than in the dry season at MSTP (**Table 12**). A 20.1% increase in the flow rate in the wet season was found compared to the dry season at MSTP although a separate sewer system is used, while the flow rate in MWTW with a combined sewer system only increased by 11.2% in the wet season. This highlights that the rainfall effect on wastewater flow rate depends on not only the type of sewer system but also rainfall intensity and other factors.

The influent pollutant concentrations in dry and wet seasons are relatively stable in MSTP while they varied significantly in MWTW from 12.8 to 57.2%. This might be because that wastewater in MSTP has low strength pollutants even in the dry season while the wastewater strength in MWTW is much higher, leading to more susceptibility of influent pollutant concentrations to rainfall's dilution. Although the higher temperature in summer (dry season) should be more efficient for the biological treatment to produce better effluent in MWTW, the concentrations of effluent pollutants are higher in summer (dry season). This might be due to much higher influent pollutant concentration in dry season. In MWTW with a combined sewer system, the combined untreated sewage with storm runoff during wet season overflows to rivers when influent flow rate is over 6 times of dry weather flow. **Table 12** shows a sample of CSO discharge to the water body on 14th February 2017. Although the pollutant concentrations in CSO in MWTW during a storm event are much lower than the influent concentrations due to dilution, pollutant concentrations are still much higher than the effluent in both seasons. This suggests a risk posed by untreated CSO to public health and the environment. Since the data on the discharge amount and frequency of CSO in MWTW are not available, LCA analysis in this study does not include the environmental impact from CSO. In addition, this can facilitate the comparison between two WWTPs with f^{oc}uses on wet and dry seasons only in this study without considering CSO.

Table 12. Comparison of average monthly parameters, influent and effluent pollutant concentrations in Malaysian STP and Millbrook WTW

	Malaysian STP			Millbrook WTW			
	Dry season[A]	Wet season[B]	Difference (%)[E]	Dry (summer season)[C]	Wet (winter season)[D]	Difference (%)[E]	CSO[F]
Rainfall (mm)	43.0 ±14.8	378.0 ±60.5	88.6	62.2 ±14.4	84.4 ±32.2	26.2	
Flow rate (m^3)	3.6E+06± 3.0E+05	4.5E+06± 9.8E+04	20.1	1.2E+06± 1.3E+05	1.3E+06 ± 1.3E+05	11.2	4.8E+04/day
Power consumption (kWh)	9.2E+05± 4.5E+04	1.1E+06± 5.9E+04	13.8	7.2E+05± 2.4E+04	6.5E+05 ± 2.6E+04	-10.0	
Sewage temperature (^oC)	21.5	20.2	6.1	20.5 ±2.2	12.4 ±1.3	39.5	
<u>Influent</u>							
TBOD (mg/L)	86.0±11.5	77.7±8.5	9.7	327.5±186.0	213±36.1	35.0	161.0
TCOD (mg/L)	178.7±65.0	177.3±40.6	0.7	1157.5±81.3	495.5±39.6	57.2	434.0
TSS (mg/L)	96.7±27.9	79.7±13.1	17.6	595.3±42.1	259.8±22.3	56.4	360.0
TN (mg/L)	23.5±2.2	22.3±0.7	5.0	70.3±1.0	47.6±1.2	32.3	25.5
TP (mg/L)	4.0±0.08	3.3±0.07	17.5	6.3	4.2	12.8	4.0
<u>Effluent</u>							
TBOD (mg/L)	8.9 ±1.6	4.7 ±2.4	47.2	8.4 ±3.1	3.7 ±0.1	56.0	161.0
TCOD (mg/L)	51.0 ±23.5	42.8 ±10.2	16.1	46.1 ±3.7	32 ±0.1	30.6	434.0
TSS (mg/L)	21.2 ±9.9	14.9 ±8.0	30.0	7.5 ±0.2	6.5 ±2.5	13.3	360.0
TN (mg/L)	10.8 ±4.2	7.6 ±3.5	29.3	9.0 ±0.9	6.9±1.1	22.8	25.5
TP (mg/L)	2.2±0.04	1.2±0.04	45.5	1.1	0.8	27.3	4.0

[A] From January to March 2016; [B] From September to November 2016
[C] From June to July 2017; [D] From January to February 2017
[E] Wet season used as a reference; [F] One set of data on combined sewer overflows (crude effluent) at >6DWF (dry weather flow)

3.3.3 Seasonal comparison of seven life cycle environmental impact categories in MSTP and MWTW using FU1 (1m^3 treated wastewater) and FU2 (eutrophication reduction – 1 kg PO_4^{3}-eq)

3.3.3.1 Environmental impact of two WWTPs in wet and dry seasons using FU1

The environmental impact assessment results of two WWTPs are shown in **Figure 6.** It can be seen that environmental impacts of all categories are lower in the wet season than that in the dry season with the difference less than 19.6% except for eutrophication potential (EP), which is 39 % lower in MSTP and 25 % lower in MWTW in the wet season. This seems straightforward as there are lower pollutant concentrations in the effluent due to the dilution by rainfall (Joel et al., 2017) in the wet season with per m^3 treated wastewater as the functional unit for comparison. Meanwhile, there are no significant changes in operational conditions such as chemical, power consumption and transportation against the increased flow rate in the rainy season (Piao & Kim., 2016). The difference between wet and dry seasons suggests a necessity to do seasonal LCA assessment, especially when considering eutrophication to the environment.

Direct emissions of COD, TN and TP in effluent contribute 99% to EP in both treatment plants during dry and wet seasons. Obviously, to reduce EP impact, it is important to increase TN and TP effluent discharge standards. The current P discharge standard of 1-2 mg/L in the UK (Lesjean et al., 2003) cannot comply with the EU Water Framework Directive 2000/60/EC to reach 'good' ecological standard in the country's watercourses (Howell, 2010; Vlachopoulou et al., 2014). Much stricter phosphorus limits such as 0.1 mg/L (for large wastewater treatment works) and 0.5 mg/L (for small sites) are thus set to be imposed in the UK (Jarvie et al., 2006; Howell, 2010). This is expected to reduce eutrophication in watercourses greatly. But there is no expected discharge requirement of phosphorus to rivers/streams in Malaysia (DOE Malaysia, 2010) in the near future, and it is thus expected that eutrophication in watercourses will still be a problem. EP impact from MWTW in both seasons is lower than that in MSTP although the influent nutrient concentrations in MWTW are 2-3 times higher. This is mainly because MWTW adopts Bardenpho treatment for nitrogen and phosphorus removal to a certain degree while MSTP process is operated only for COD and SS removal. It needs to be pointed out that the eutrophication would be 81% more in MWTW in the senario of combined sewer overflows (CSO) due to the direct raw sewage emission to natural water bodies (**Figure 7**). The other six life cycle impact categories, however, have comparable results between CSO occurrence and the normal winter (wet) condition. This implies that the direct discharge of untreated wastewater to the environment during storm event mainly causes eutrophication. To reduce this impact, the UK is promoting sustainable urban drainage system to reduce CSO frequency or root out the occurrence of CSO from the source (Stovin et al., 2007).

For the other 6 environmental impact categories, energy consumption is the main contributor, dominating in both plants during both seasons. Since the wastewater strength in MWTW is much higher than that in MSTP, and meanwhile MWTW adopts technology for nutrient removal, the electricity consumption in MWTW for treating per m^3 wastewater is 0.55 kWh while it is only 0.26 kWh in MSTP. Nitrogen removal demands more aeration thus more electricity for nitrification. The environmental impact caused by energy consumption is also related to the energy source for electricity generation. 93% of electricity production in Malaysia is depending on fossil fuel while it is only 58% in the UK with the other 42% from renewable and nuclear power.

Electricity consumption accounts for 96% to ozone layer depletion potential (ODP) in MWTW (**Figure 6b**). ODP is 19.6% higher in the dry season than the wet season due to 20.5% higher of electricity consumption per functional unit (1 m^3 treated wastewater) in the dry season. ODP value in MWTW was 99.1% higher than that in MSTP due to 53% higher energy consumption in MWTW per m^3. This result in MWTW is comparable to those reported by Godin et al. (2011) and Lorenzo-Toja et al. (2016) that high energy consumption per functional unit of 1 m^3 ranging from 0.4 to 0.7 was used. Both plants have no chemical addition for phosphorus removal, thus, the contribution to ODP is mainly from electricity consumption. It has been reported that the addition of ferric chloride for phosphorus removal or flocculation can contribute to more than 90% of ODP (Greg et al., 2016; Lorenzo-Toja et al., 2016) because the production of ferric chloride leads to high emission. Thus, based on LCA analysis, appropriate process/chemicals could be chosen in WWTPs to reduce negative environmental impact.

For freshwater ecotoxicity potential (FEP) category (**Figure 6c**) and Human toxicity potential (HTP) category (**Figure 6d**) in both plants, dry and wet seasons do not show an evident difference because of the nearly similar electricity consumption and chemical consumption during these two seasons. But FEP and HTP in MSTP are much higher than those in MWTW and electricity accounts for 99% share while in MWTW chemical consumption contributes to a certain degree. This is mainly because the electricity generation in the UK is less dependent on fossil fuel, which results in smaller FEP and HTP because FEP and HTP are mainly from fossil fuels.

For GWP impact, MWTW is 35% higher than MSTP due to high electricity consumption per m^3, chemical consumption for denitrification, and higher direct emission from high strength wastewater. Regarding seasonality, GWP in the dry season is 7.6% higher in MSTP and 14.2% higher in MWTW, respectively. This difference is mainly caused by the difference in energy consumption per functional unit due to the seasonal difference in influent quality as well as the wastewater strength. The dilution effect from storm runoff is more effective to relatively high strength wastewater in MWTW to result in a less environmental impact in the wet season due to the reduced power consumption and the less direct emission due to the reduced wastewater strength. GWP values in this study ranging from 0.40 to 0.73 $kgCO_2eq/m^3$ are in accordance with those reported by (Rodriguez-Garcia et al., 2011; Corominas et al., 2013; Lorenzo-Toja et al., 2016) with GWP ranging from 0.44 to 0.71 $kgCO_2eq/m^3$. This suggests a consistent GWP range from WWTPs. It has to be pointed out that apart from electricity consumption, direct emission from wastewater treatment processes is also an important contributor to GHG emission. However, it is believed that this direct emission is usually underestimated by the calculation guided by IPCC. Based on the actual measurement on-site, direct emission could contribute up to 71% of the total GHG (Delre et al., 2019). This poses a great challenge to WWTPs to optimize the treatment process to reduce direct emission especially N_2O from nitrogen removal process and CH_4 from sewage and sludge handling.

For abiotic depletion (fossil fuel) potential (ADFP) (**Figure 6f**), MSTP presents a slightly better result than MWTW because MWTW uses more electricity per m^3 treated water but lower fossil fuel percentage in the grid. Again, the season difference, i.e. 16.4%, is more obvious in MWTW than in MSTP. **Figure 6g** shows that the main contribution to acidification potential (AP) is also from the electricity consumption in both treatment plants with a 40% higher impact in MSTP. This is attributed to emissions of gases such as sulfur dioxide, sulfur monoxide and nitrogen oxides from fossil fuel combustion for electricity generation in Malaysia. Chemical consumption only accounts for 6.3% in MWTW and 0.1% in MSTP respectively. AP in Dry season is 7.5% and 18.8% higher than a wet season in MSTP and MWTW, respectively. In terms of the electricity consumption in different processes, **Figure 8** shows that average electricity consumption in secondary treatment is the highest with 59% and 65% in MSTP and MWTW respectively. Secondary treatment is the most energy intensive process due to high energy use for aeration in both WWTPs, although MSTP only remove organic matter. Therefore, effort to reduce electricity consumption should mainly focus on secondary treatment for both WWTPs.

From the LCA assessment above, it is found that a higher percentage of fossil fuel for electricity generation results in higher impacts in terms of categories of FEP, HTP, AP and ADFP. Therefore, moving the electricity generation from fossil fuels to renewable energy definitely benefits environment impact from WWTPs just as the UK did in the last few decades (UK Energy, 2017). This is obviously a nation-level strategy on energy use and environmental protection. However, if WWTPs are able to recover energy from wastewater as much as possible to cover its own energy consumption, it will bring down environmental impacts in these 4 categories. For EP and GWP, they are more dependent on treatment performance and final effluent emission to the environment. More advanced treatment results in lower EP but higher GWP due to the increased chemical and energy consumption for advanced treatment. There is a trade-off between them. Meanwhile, the direct emission to GWP should not be neglected although it is still not common to be included in most studies on LCA. With regard to the seasonality effect by LCA analysis, it can be found that wet season in both plants has a less environmental impact than the dry season. This is mainly due to the dilution from storm runoff, thus the lower emission from effluent to the environment. In addition, less electricity is consumed to treat per $1m^3$ wastewater during wet seasons due to the dilution of pollutants. MWTW shows a more obvious difference between two seasons while MSTP is more or less comparable except for EP category. From the raw sewage data, we can see that the strength of sewage to MWTW is much higher than that to MSTP, and the dilution during wet season plays a much obvious role in MWTW for reduced electricity consumption as well as reduced pollutant concentrations. Therefore, raw sewage strength is a key factor to lead to different environmental impact in the dry and wet seasons. This can well explain the contradictory results from the literature. Some studies reported lower environmental impact in a wet season than in dry season (Moreira et al., 2004; Mines et al., 2007; Joel et al., 2017), while Lorenzo-Toja et al. (2016) reported higher environmental impact in a wet season (winter). Therefore, it is very necessary to do LCA analysis with the consideration of rainfall effect on the sewage dilution especially when sewage has high strength to reflect real environmental impacts from WWTPs in different seasons. In addition, site-specific LCA for WWTP is also necessary to reflect the accuracy of environmental impact profile with different precipitation intensity (Yoshida et al., 2014).

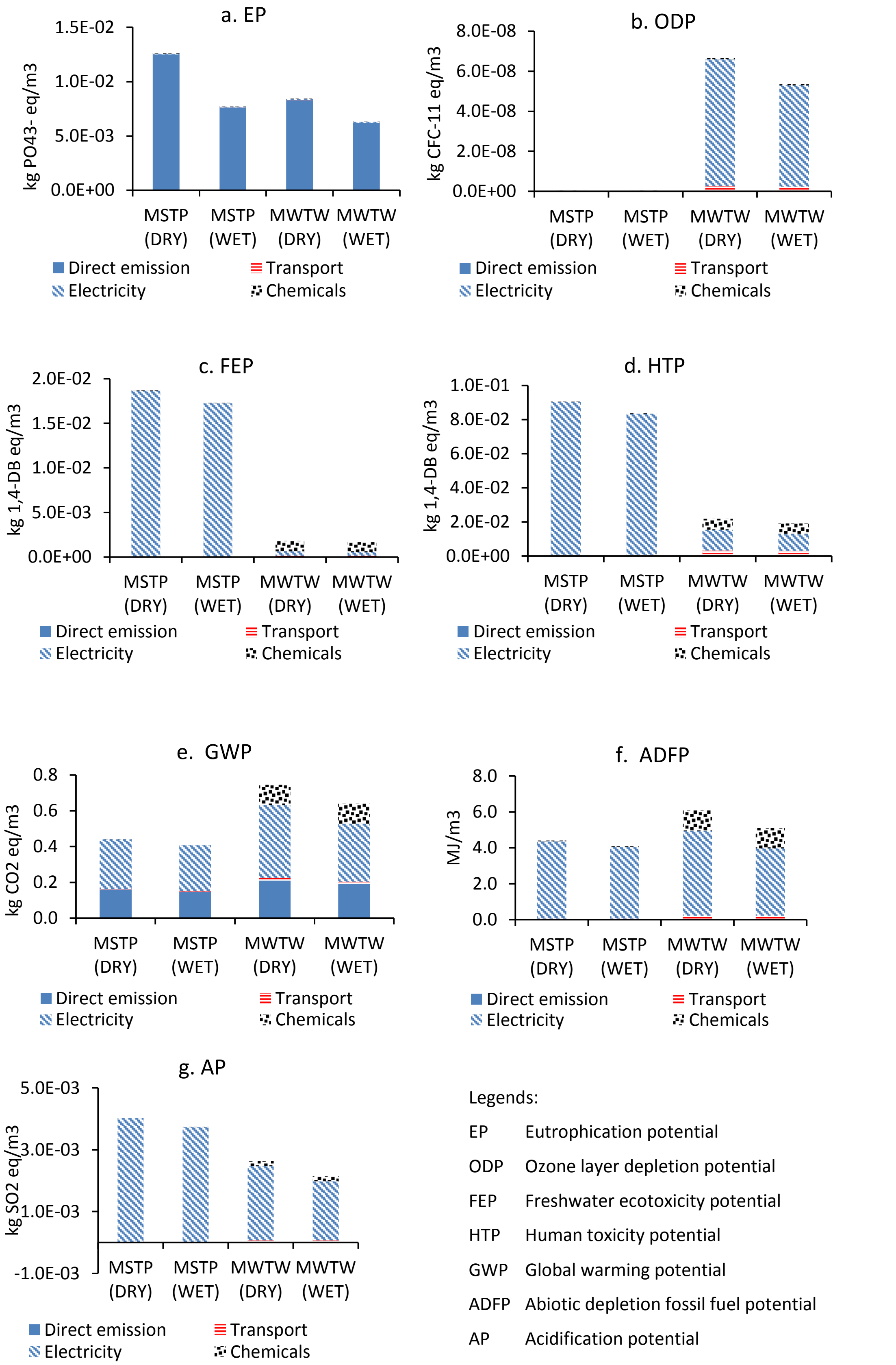


Figure 6. Environmental impact assessment in seven categories at Malaysian STP and Millbrook WTW in both dry and wet seasons by using FU of 1 m^3 treated wastewater

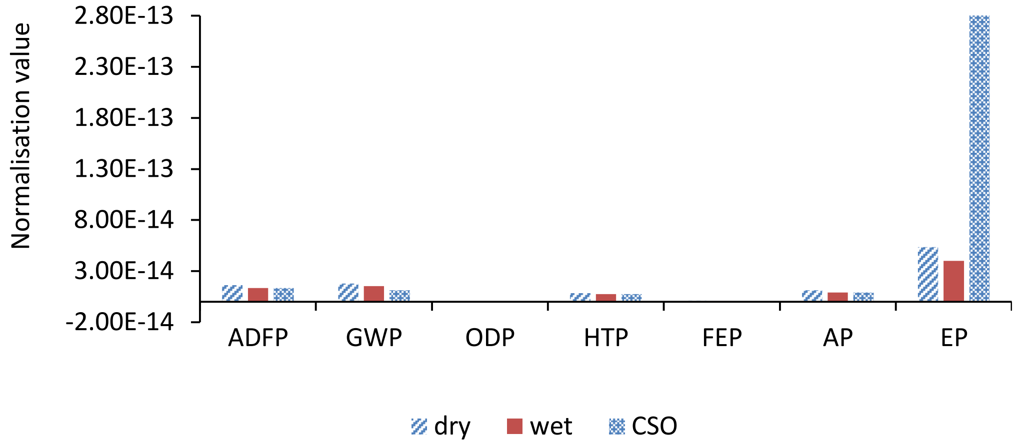


Figure 7. The comparison among dry season, wet season and combined sewer overflow (CSO) occasion in terms of seven impact categories in Millbrook WTW using functional unit of 1m^3 treated wastewater (FU1)

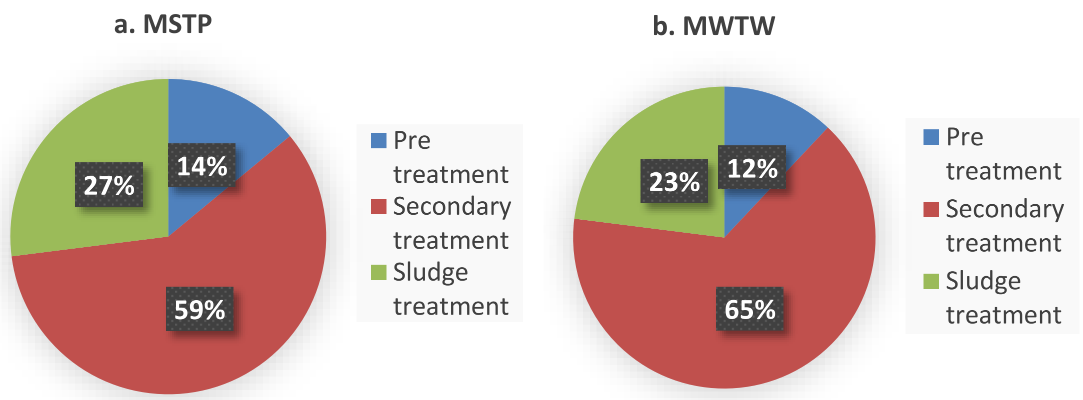
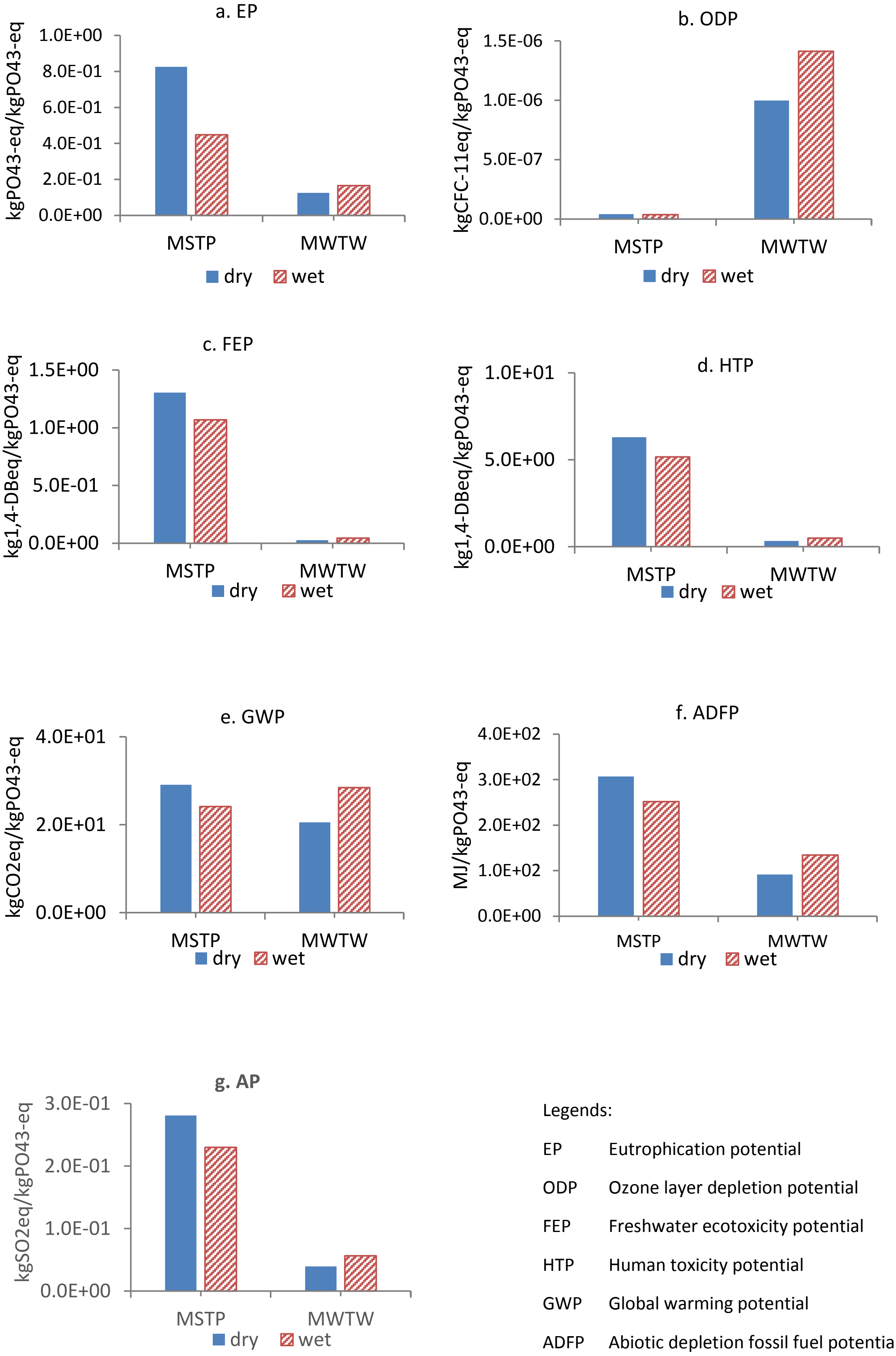


Figure 8. The contribution of electricity consumption in different processes, i.e. pre-treatment, secondary treatment and sludge treatment for a) MSTP and b) MWTW

3.3.3.2 Environmental impact of two WWTPs in wet and dry seasons using FU2

Environmental impact analysis using FU2 was conducted to compare the impact difference of by using two functional units for both WWTPs in dry and wet seasons. It can be seen from **Figure 9** that except for ODP, MWTW still exhibits lower environmental impacts using FU2 compared to MSTP due to its high nutrient removal efficiency and better effluent quality. In MSTP, each environmental category in dry and wet seasons shows a similar trend when using FU1 and FU2. This suggests that no much difference is caused by adopting different functional units to WWTP with low strength wastewater. MWTW, however, demonstrates higher environmental impact in the wet season than the dry season with FU2, which is contrary to that by using FU1. For example, the lower EP from MWTW in the dry season using FU2 reflects a higher pollutant removal efficiency than in wet season, indicating that rainfall in wet season negatively affects wastewater treatment efficiency although it plays a dilution role. This result is in agreement with the study by Rodriguez-Garcia et al. (2014) who compared nitritation-anammox, nitrite shortcut and struvite crystallization processes for the supernatant treatment from anaerobic sludge digestion using two functional units, i.e. FU1 (per m^3 treated wastewater) and FU2 (kg PO_4^{3}-eq removal). It was found that struvite crystallization process has the lowest eutrophication (EP) impact using FU1 due to the cleanest effluent (partially due to much lower influent pollutant concentrations) but the highest EP using FU2 due to the lowest removal of COD and N, and the least efficient in terms of EP reduction. In addition, a higher difference in all impacts ranging from 25% to 39% between dry and wet seasons is found by using FU2 compared to FU1.

It can be seen that, the selection of appropriate functional unit is prominent as the total treated water discharge volume to the environment is more in the wet season than the dry season, leading to the possible higher total pollutant mass load to the environment. It is noteworthy that although FU1 has been widely used for seasonal LCA assessment of WWTP (Piao & Kim, 2016; Lorenzo-Toja et al., 2016; Li et al., 2017; Risch et al., 2018), effects from the variation of influent compositions and flow could not be reflected very well if only using per unit volume as a functional unit (Rodriguez-Garcia et al., 2011; Piao et al., 2016). The functional unit as per kgPO_4^{3}-eq removed based on eutrophying substances removal (e.g. COD, TN and TP) is believed to reflect the wastewater treatment performance better as pollutant removal from wastewater is the main objective of a WWTP to meet effluent limits by the legislation (Comas Matas, 2012). Thus, considering pollutant removal efficiency during the wastewater treatment process, using FU2 as 1 kgPO_4^{3}-eq removed is more appropriate for an environmental impact assessment. It also makes the direct comparison between different WWTPs, or different seasons more meaningful as it is mainly based on pollutant removal by minimising the effect from influent compositions and flows.



Legends:

EP	Eutrophication potential
ODP	Ozone layer depletion potential
FEP	Freshwater ecotoxicity potential
HTP	Human toxicity potential
GWP	Global warming potential
ADFP	Abiotic depletion fossil fuel potential
AP	Acidification potential

Figure 9. Environmental impact assessment in seven categories at Malaysian STP and Millbrook WTW in both dry and wet seasons by using FU of 1 kgPO_4^{3-} eq. removed

3.3.3.3 Detailed comparison of EP and GWP categories between FU1 and FU2

Based on **Figure 7**, eutrophication potential (EP) and global warming potential (GWP) are two categories that are mostly affected by direct emissions from WWTPs. Therefore, they were further analysed to investigate the detailed contribution of each substance such as the contributions of COD, TN and TP to EP and, the contribution of CH_4, and N_2O to GWP. These could be further compared with indirect emissions from electricity and chemical consumption. As shown in **Figure 10,** TP in the MSTP effluent contributed 54% in a dry season and 45% in a wet season to eutrophication category, respectively, with both functional units. TN is the second-highest contributor in MSTP with a contribution of 36% and 43% in a dry and wet season, respectively, followed by COD (11%) and negligible impact (<1%) from the electricity and chemical consumption (indirect impact) in eutrophication. Rodriguez-Garcia et al. (2011) also highlighted the negligible impact of electricity and chemical consumption in this category. In MWTW, TN and TP present roughly comparable contributions to EP in both seasons using either FU. EP results by using FU2 show significantly lower EP in MWTW than MSTP due to considerable removal of pollutants including nutrients but MSTP does not have nutrient removal. This result suggests that FU2 reflects more effort made by the plant for pollutant/nutrients removal instead of the actual effluent emission only as FU1 does. The result from Piao & Kim. (2016) also highlighted that their WWTP B using A2/O process with higher nutrient removal rate had a 30% lower EP impact compared to WWTP A with conventional activated sludge when using FU of 1 kg TN removed. The big difference of EP in dry and wet seasons in MSTP also suggests that nutrient removal in the dry season is more important than wet season to reduce EP.

For GWP category (**Figure 10c and 10d**), MWTW has much higher GWP than MSTP using FU1 while GWPs in both plants are comparable by using FU2. This suggests again that FU selection is important for the comparison between different WWTPs with different influent compositions. The strength of wastewater to MWTW is higher than that to MSTP, resulting in almost double electricity consumption for treating per m^3 wastewater. In addition, the additional chemical dose in MWTW for denitrification also contributes to GWP. Thus, it is plausible that GWP in MWTW is higher than MSTP when the comparison is based on per m^3 treated wastewater. When the comparison is based on per kgPO_4^{3}-eq removed, however, it is found that MWTW is more environmentally efficient for the pollutant removal. This means that less environmental impact is caused by removing the same amount of pollutant. In addition, indirect contribution to GWP using FU2 is much smaller than using FU1 in MWTW, making the direct contribution from CH_4 and N_2O in treatment process more predominate (2 times more than that using FU1). Rodriguez-Garcia et al. (2014) also reported the higher percentage of direct emission to GWP with N-removal technology when using FU2, proving that direct emission could be dominant in the GWP impact category of WWTP. Nowadays, more on-site measurement of CH_4 and N_2O emission (Masuda et al., 2015; Schaubroeck et al., 2015; Piao et al., 2016) indicates that the direct emission of CH_4 and N_2O based on IPCC guidelines is underestimated. The direct GHG emission from a studied WWTP can contribute 75% to GWP with 53% from N_2O and 22% from CH_4 according to the average site-specific emission factor from the Korea Environmental Corporation Report 2008 (Piao et al., 2016). With $1m^3$ treated water as FU, the emission of N_2O and CH_4 from Piao et al., 2016 is 3.5 and 5.5 times higher, respectively, than those in this study calculated based on IPCC guideline. The higher percentage of direct emission poses a great challenge to reduce GWP in WWTPs because currently there are still no widely accepted strategies, which can mitigate CH_4 and N_2O emissions effectively from wastewater treatment processes. In addition, the higher GWP in MWTW in wet season suggests a less efficient pollutant removal.

Overall, EP in MWTW is smaller than MSTP with both FUs due to the nutrient removal process. EP and GWP in MWTW in dry and wet seasons showed contrasting trends when using FU1 and FU2, respectively, indicating that MWTW is more sensitive to the selection of different functional units. This is probably stronger wastewater to MWTW with nutrient removal process, which is more affected by dilution from rainfall.

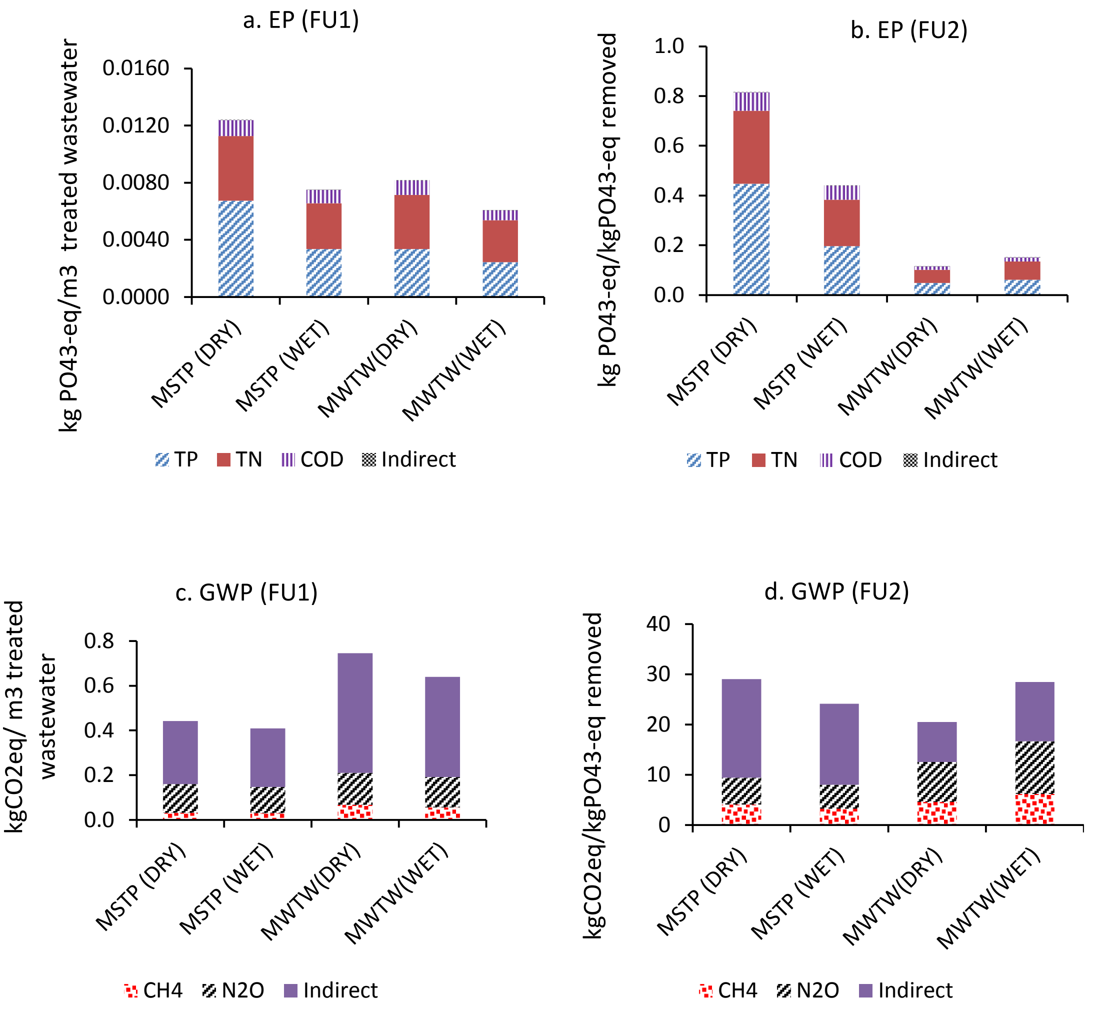


Figure 10. Comparison of eutrophication potential (EP) and global warming potential (GWP) at Malaysian STP and Millbrook WTW in the dry and wet seasons by using FU1 (per m^3) and FU2 (per $kgPO_4^{3-}$-eq. removed), respectively

Legends: EP = Eutrophication potential, GWP = global warming potential

3.3.4 Sensitivity analysis

Environmental impact assessment results in two plants highlight that nutrients in effluent and electricity consumption are the major factors to affect environmental impacts. **Table 13** shows how environmental impacts in MSTP and MWTW in the dry season (as example) were affected by varying ±10% of selected inventory component values such as nutrient concentrations and electricity consumption or chemical consumption. Data from FU1 was selected for this analysis to facilitate the comparison with the results with other studies. Environmental impact categories such as FEP, HTP, GWP, ADFP, and AP varied from ±7.4% to ±9.9 % to respond to the change in electricity consumption by ±10%. The response to ±10% change in electricity consumption in MWTW was even less obvious, ranging from ±2.8% to ±9.5% in six categories except for eutrophication. The less sensitivity to electricity consumption values in MWTW is mainly due to lower fossil fuel percentage used for electricity generation in the UK compared with Malaysia (**Table 14**). This result is in agreement with Piao et al. (2016) that electricity consumption caused the most sensitive change to acidification and human toxicity potentials in all WWTPs studied. Eutrophication potential (EP) changed by ±9.1% in MSTP and by ±8.5% in MWTW to respond to ±10% change in TP and TN concentrations in the effluent while the other six categories are almost unaffected. Finally, the chemical consumption shows less effects on all the categories with the highest FEP change by ±6.1%. In general, the variation of electricity and nutrients in the effluent by 10% will not cause an environmental impact change more than 10%, suggesting a less sensitivity of environmental impact results to inventory data.

Table 13. Sensitivity analysis results by changing selected inventory data by ±10% of in Malaysian STP and Millbrook WTW in the dry season according to 1 m^3 of treated wastewater

	MSTP (%)		MWTW (%)		
Inventory components	Electricity consumption	TN and TP in the effluent	Electricity consumption	TN and TP in the effluent	Chemical consumption*
Eutrophication potential, EP	±0.50	±9.05	±0.10	±8.50	±0.02
Ozone layer depletion potential, ODP	±0.06	±0.01	±9.52	±0.00	±0.10
Fresh water ecotoxicity potential, FEP	±9.95	±0.00	±2.78	±0.01	±6.11
Human toxicity potential, HTP	±9.88	±0.00	±5.56	±0.00	±2.86
Global warming potential, GWP	±7.39	±0.00	±5.27	±0.02	±2.05
Abiotic depletion (fossil fuels) potential, ADFP	±9.79	±0.00	±7.73	±0.00	±1.87
Acidification potential, AP	±9.95	±0.00	±9.10	±0.00	±0.62

Note: TN = Total nitrogen, TP = total phosphorus; Sensitivity analysis on chemical consumption was not conducted in MSTP since chemical consumption contributes less than 1% to all environmental impacts categories

Table 14. The national electricity generation mix in Malaysia and United Kingdom

Energy source	Malaysia (%)[a]	United Kingdom (%)[b]
Natural gas	45	41
Coal	41	11
Oil	7	-
Renewable energy (incl. biomass, wind, solar)	7	27
Nuclear	-	15
Others (e.g. interconnector)	-	6

[a] Source: Ecoinvent v3.3
[b] Source: Ecoinvent v3.3 and www.mygridgb.co.uk

3.4 Conclusion

The influence of rainfall on the environmental impacts of two large centralised WWTPs with different wastewater strengths and sewer systems but similar rainfall effects on influent flow rate was investigated by using LCA in this study. Meanwhile, two different functional units were evaluated to see how the selection of functional units affect LCA results in the circumstance of rainfall effects. The results are summarised as below.

- The coefficients between monthly rainfall and the influent flow rate are similar at around 2500 m^3 influent flow rate/mm precipitation although two WWTPs have different sewer systems and wastewater strengths. This disclose that rainfall intensity affects the quantity and quality of influent to WWTPs, but the extent of effect is not directly determined by rainfall intensity or sewer system, i.e. if it is a combined or a separate sewer system.
- Based on the life cycle analysis from two large centralised WWTPs, nutrients in effluent and electricity consumption are the major factors to affect the environmental impacts, while chemical consumption and transportation has minimal impact on the environment due to the little consumption of chemicals.
- When per m^3 treated wastewater was used as the functional unit, all environmental impact categories in MSTP except eutrophication potential are almost similar in dry and wet

seasons while MWTW shows higher environmental burdens in the dry season than a wet season for all seven environmental impact categories.

- When per kgPO_4^{3-}eq. removed was used as the functional unit, all seven environmental impacts in MWTW showed higher values in the wet season than the dry season, while the selection of either of functional units has no influence on the environmental impact categories in MSTP.

The results from this study demonstrate that rainfall effects on the environmental impact of WWTPs are more effective in MWTW with higher wastewater strength. The contrasting results of environmental impacts in MWTW during wet and dry seasons by using two different functional units suggest that the selection of functional unit is dependent on the comparison purpose, such as the impact of WWTPs effluent to the environment only, or the combined effects from effluent and WWTP treatment efficiency. This work identified the importance of wastewater strength and functional units to the studies of rainfall effects on the environmental profile of WWTPs, which could serve as a basis for further rainfall studies with different coefficients between rainfall intensity and inflow rate, advanced treatment and others. In addition, the environmental impact assessment in this study provides guidance for a better eutrophication potential control especially in vulnerable receiving waters in different seasons.

4 Evaluation of life cycle toxicity assessment methods of municipal wastewater treatment plants with the inclusion of direct emissions of heavy metals and PPCPs

4.1 Introduction

Toxicity impact assessment from municipal wastewater treatment has attracted great attention in recent years especially when more and more contaminants of emerging concern (CEC) are detected from municipal wastewater. According to European Economic Community 1991 (EEC, 1991), municipal wastewater treatment plants contribute a considerable amount of pollutants to the natural environment through disposal of sludge and discharge of effluent to water bodies. Thousands of prescribed drugs such as antibiotics, contraceptives, personal care products (e.g. soap, fragrances and sunscreen agents) and beta-blockers used daily (Munoz et al., 2008), which finally go to sewage works after consumption. In the year 2000, the EU framework directive identified 33 priority pollutants in the aquatic environment including heavy metals such as nickel, chromium, lead and mercury. In addition, pharmaceuticals such as carbamazepine, triclosan, bisphenol-A and ibuprofen are regarded as priority personal care products (PPCPs) by EU framework directive 2007 for discharge monitoring because of their regular detection and possible effects on human health (Archer et al., 2017). Although these priority pollutants are present in very low concentrations, their continued release from wastewater effluent to the environment is believed to have potential to cause long term hazards to human and the environment (Bolong et al., 2009; Alfonsín et al., 2014). A typical sewage treatment plant, however, is designed to remove organic matters and nutrients but not micropollutants such as pesticides, pharmaceuticals, and heavy metals (Gallego-Schmid & Tarpani, 2019). Therefore, assessing toxicity from sewage treatment plants started to increasingly gain attention in the last decade to see what degree of hazards that micropollutants or other priority pollutants which are not targeted for removal by almost all sewage treatment plants might cause, and if measures need to take particularly in vulnerable and sensitive areas. To achieve this purpose, life cycle assessment (LCA) has been used as a potential approach, but it is mainly for developed countries (Munoz et al., 2008; Lorenzo-Toja et al., 2016a).

A wide range of factors influence the types and quantities of PPCPs and metals in wastewater such as catchment sizes, lifestyles, economic development levels, local medical and farming practices. In addition, wastewater treatment technologies adopted in sewage treatment plants can also affect the concentrations of PPCPs and metals in effluent because it has been reported that some PPCPs such as acetaminophen and caffeine could be removed in good efficiencies by microbial biodegradation or sorption to sludge even though they are not targeted at (Sin et al., 2009). It is thus expected that toxicity effects from sewage plants might vary region by region. So far, most studies on toxicity impacts of sewage plants by LCA were carried out for developed countries (Lorenzo-Toja et al., 2016; Shimako et al., 2017; Emara et al., 2018). Only recently a complete LCA study on toxicity was conducted in a developing country i.e. in China by Li et al., 2019, but all the inventories or databases including metals and 126 PPCPs were from secondary data (i.e. from various literature in developed and developing countries). As highlighted by Gallego-Schmid & Tarpani (2019), the key parameters such as influent, effluent and sludge produced, and the technologies adopted for wastewater treatment have medium to high influence on the LCA of wastewater treatment. The lack of such real key information would make LCA of wastewater less representative for local situations. Therefore, sampling campaign is more preferred than computing or modelling particularly for PPCPs and metals as they vary significantly with wastewater treatment technologies, locations and seasons, and are more unpredictable (Luo et al., 2014). As reported in a review paper by Li et al. (2017), the average concentrations of BOD, COD, SS, TN and TP in developing countries are lower than those in developed countries. Rashid & Liu (2020) shows that wastewater in Malaysia is even more diluted and almost at the lower limit of the ranges of these parameters in developing countries as described by Gallego-Schmid & Tarpani (2019). This poses interesting questions, which are: i) what levels of PPCPs and metals in Malaysian wastewater, ii) how they influence the toxicity of wastewater, and iii) if necessary measures need to take in the future for more stringent discharge standards of treated wastewater. Obviously, to answer these questions, sampling campaign has to be carried out because the data from literature especially from developed countries could not represent the situation in Malaysia. Meanwhile, information about PPCPs and metals in Malaysian municipal WWTPs is completely missing. Therefore, to study toxicity of PPCPs and metals from wastewater in Malaysia, it is very necessary to do sampling with the consideration of technology used, which does provide relatively accurate data to life cycle inventory to execute LCA, and also extend the data of PPCPs and metals to developing countries for further toxicity studies.

Compared with impact categories such as acidification potential (AP) and global warming potential (GWP), the study on toxicity with LCA is relatively new and more challenging. Large discrepancies between the life cycle impact assessment (LCIA) methods regarding toxicity impact were reported

(Renou et al., 2007; Pizzol et al., 2011) and the comparison between different models was carried out to identify the sources of differences (Renou et al., 2007; Niero et al., 2014; Piao et al., 2016). The Society for Environmental Toxicology and Chemistry (SETAC) and United Nations Environment Program (UNEP) have introduced the Life Cycle Initiative for LCA practitioners to apply more effective life cycle practice (Rosenbaum et al., 2008). As a result, USEtox was developed and recommended as a scientific consensus model after a comparison between several models such as IMPACT 2002+, WATSON, USES-LCA, EDIP, BETR, EcoSenee, and CalTox for assessing toxicity-related effects in LCA (Rosenbaum et al., 2008; European commission, 2013). However, due to the complexity of computing characterization factors (CFs), it was clearly pointed out in the USEtox that CFs provided are only interim instead of recommended for metals, dissociating and amphiphilic substances (Rosenbaum et al., 2008). In addition, available CFs for PPCPs in existing USEtox model are very limited and some of the modelling on fate, exposure and impact pathways of chemicals is inaccurate (Emara et al., 2018). Thus, for the emissions of PPCPs (i.e. most are dissociating and amphiphilic chemicals) and metals i.e. main contributors in WWTPs to toxicity impacts, the comparison between different models are still necessary in the current situation to improve the understanding about different LCIA methods for WWTP-LCA. Even though the above mentioned problems exist, toxicity assessment can still not only provide indicative effects of chemicals from different products or processes on human and ecosystems (Pedrazzani et al., 2019), but also prioritise/rank heavy metals or PCPPs for removal. The toxicity assessment by considering emissions of PPCPs and metals in developing countries, however, is very little (Sin et al., 2009). This poses a great challenge to understand the toxicity impacts in developing countries with different wastewater quality and technologies adopted. Toxicity impact is more important to regions than global warming as toxicity can directly cause impact to human and ecosystems in more vulnerable and disadvantaged areas. Furthermore, it is still not very clear about the contribution of PPCPs and metals to toxicity categories compared with indirect emissions such as electricity and chemicals used for wastewater treatment. Therefore, this study aims to assess toxicity impact of PPCPs and metal emissions from a large centralised wastewater treatment plant in Malaysia with low wastewater strength, and to provide useful information for LCA toxicity assessment practice by identifying the importance and contribution of PPCPs and metals and comparing LCIA models.

4.2 Materials and method

4.2.1 The selection and description of the case study

A centralised municipal wastewater treatment plant (Malaysian STP) in Penang, Malaysia was selected as a case study in this research to investigate the life cycle toxicity impacts in developing countries and also to provide useful information for LCA toxicity assessment practice. Malaysian STP treats domestic wastewater with average flow rate of 148,950 m3/d (i.e. 662,002 population equivalent) in 2017. The operation of Malaysian STP are including of grit and grease screening (primary treatment), sequencing batch reactor (secondary treatment by COD removal), gravity belt thickener, anaerobic digester, and biosolids dewatering (**Figure 11**). This type of wastewater treatment technology and configuration are widely adopted in Malaysia. The treated sewage is discharged into the river nearby, and the sludge is sent to a landfill located 47 km from Malaysian STP.

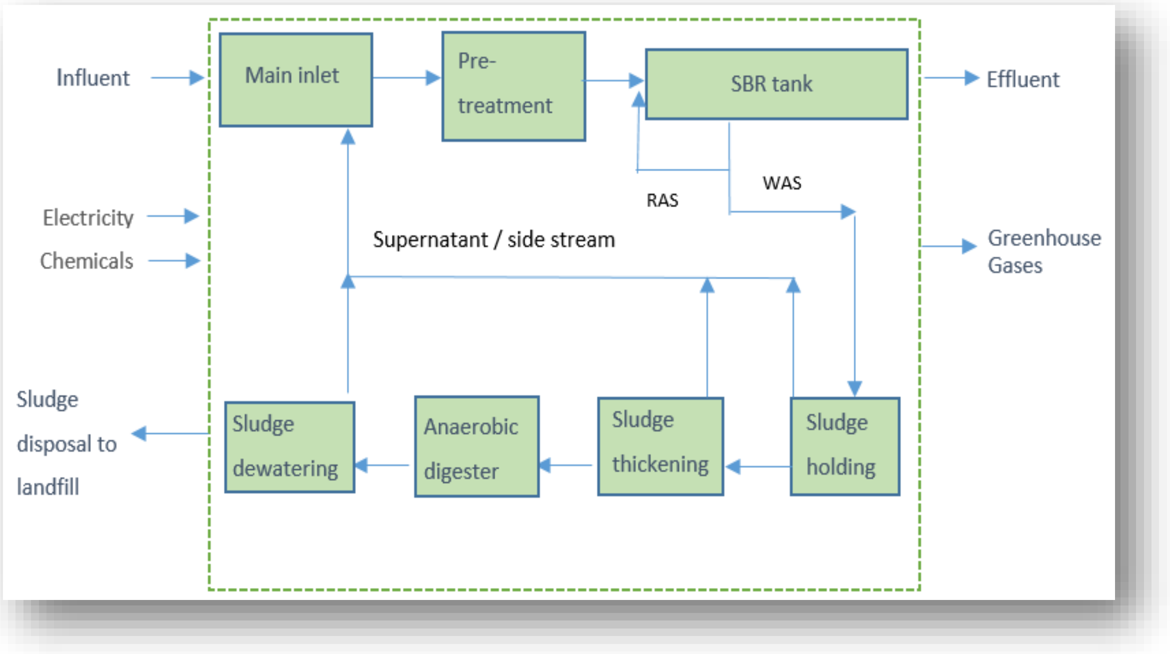


Figure 11. Schematic diagram of Malaysian STP in the system boundary of this study.

Note: For life cycle analysis, the pollutants measured in influent and effluent of MSTP are organic matters, nutrient, heavy metals and PPCPs. Meanwhile, the pollutants measured from sludge are heavy metals. (SBR = sequencing batch reactor; RAS = return activated sludge; WAS = waste activated sludge)

The selection of types of heavy metals and PPCPs as toxic pollutants in this study was based on their occurrence in Malaysia found by (Al-Odaini et al., 2010; Tan at el., 2015), their importance worldwide as highlighted by Üstün, 2009, the risks they pose to human and the environment, and the availability of their analytical methodology in Malaysia. 10 pharmaceuticals and personal care products and 9 heavy metals were selected as they are the most investigated and high-risk pollutants identified in various researches (Lorenzo-Toja et al., 2016; Yang et al., 2017; Yoshida et al., 2014; Petrie et al., 2014), and they are detectable in the wastewaters in Malaysia (Al-Odaini et al., 2010; Tan et al., 2015). The list of all selected substances for the study includes organic matters, suspended solids, nutrients, heavy metals and PPCPs, which is shown in **Table 15**.

The process and operation data were provided by Malaysian STP's manager. To get the data of identified pollutants above, influent, effluent and sludge of Malaysian STP were sampled in August 2017 to provide the required data for this study. Glass bottles were generally used to keep the samples of influent and effluent, but polytetrafluoroethylene (PTFE) bottles were used for samples intended for the analysis of heavy metals. PTFE were selected/used to avoid chemical interaction between metal ions and glass. Samples for the analysis of PPCPs were collected by 1-litre amber crystal bottle. The function of amber crystal bottle is to avoid photo-degradation of PPCPs. Sewage samples from influent and effluent were retained in iceboxes with average temperature of 4oC. For sludge analysis, 200g of sludge was collected from the sludge collection area. Finally, all samples were sent to the laboratories for analytical determination by ALS Technichem (M) Sdn Bhd (for PPCPs) and National University of Malaysia, UKM (for other pollutants). In the laboratories, sewage were filtered using Whatman GF/F filter papers to remove the suspended particulate matter. Next, sewage samples were extracted by solid-phase extraction (SPE) process using Oasis MCX cartridges ($3cm^3$/60mg). Since PPCPs in the influent and effluent of WWTP were main concerns for the pollution of receiving waters (Lorenzo-Toja et al., 2016; Li et al., 2019), PPCPs in the sludge were not analysed. The analytical methods used for all chosen pollutants in Malaysian STP are listed in **Table 15**.

Table 15. Lists of pollutants analysed from Malaysian STP and the relevant analytical methodologies used

No	Compounds	Analytical methods	Reference	Unit	Location of sampling
1	Total biochemical oxygen demand, $TBOD_5$	Standard methods-5210 B electrode method ysi meter	(Method5210, 2001)	mg/L	Influent, and effluent of Malaysian STP
2	Total chemical oxygen demand, TCOD	Hach-method 8000-Reactor Digestion method	(Method8000, 2014)	mg/L	
3	Total suspended solids, TSS	Standard methods – 2540D	(Method2540D, 2007)	mg/L	
4	Oil and grease, OG	Standard methods - 5520 B	(Method5520, 1999)	mg/L	
5	Total nitrogen, TN	Kjehdahl's method	(Method351, 1978)	mg/L	
6	Total phosphorus, TP	Microwave digestion method HPR-EN-11 (ICP-MS)	(Method3051, 2007)	mg/L	
7	Ammoniacal nitrogen, AMN	HACH method 8038 – Nessler method	(Method8038, 2017)	mg/L	
8	Nitrate	HACH method 8171 – Cadmium reduction method	(Method8171, 2014)	mg/L	
9	Sulfate	HACH method 8171 – Cadmium reduction method	(Method8171, 2014)	mg/L	
10	Heavy metals in wastewater (e.g. Cd, Cr, Cu, Fe, Pb)	ICP-MS	(Method3051, 2007)	µg/L	
11	Heavy metals in sludge (e.g. Cd, Cr, Cu, Pb, Zn)	Microwave digestion method HPR-EN-11 (ICP-MS)	(Method3051, 2007)	mg/kg	Dry sludge of Malaysian STP
12	Pharmaceuticals (carbamazepine, diclofenac, nonylphenol mixture, trimethoprim)	W-PHALMS05 LCMS	(Method1694, 2007)	µg/L	Influent and effluent of Malaysian STP
13	Pharmaceuticals (ibuprofen, 17α-ethinylestradiol, estrone, 17β- estradiol)	W-PHALMS06 LCMS	(Method1694, 2007)	µg/L	
14	Personal care product (bisphenol-A)	W-AEOGMS01 LCMS	(Method1694, 2007)	µg/L	
15	Personal care product (triclosan)	W-PESLMS04 LCMS	(Method1694, 2007)	µg/L	

4.2.2 Life cycle assessment

4.2.2.1 Goal and scope

The objective of this research was to investigate the life cycle toxicity impact of PPCPs and metals in low strength wastewater of a big centralised wastewater treatment plant in Malaysia. Since this study focused on toxic pollutants to the life cycle environmental impact from the plant's operation, construction and demolition stages were not considered because these two stages contribute negligible pollutants to water. 'Gate-to-gate' assessment was adopted comprising from raw wastewater entering Malaysian STP as influent, to the effluents discharged into nearby river, as well as sludge disposal to the landfill. Specifically, this system boundary was including wastewater treatment operations (i.e. wastewater volume, pollution loads and direct emissions to air and water bodies) as foreground system, while transport, electricity production and chemical production as background system.

4.2.2.2 Functional units

Functional unit (FU) based on volume, i.e. 1 m^3 of treated wastewater was used in this study. This FU has been commonly adopted for LCA-WWTPs including for toxicity potential impacts in many studies such as by Piao et al., 2016; Lorenzo-Toja et al., 2016; Rahman et al., 2016; Niero et al., 2014. The selection of volume as FU in the main analysis was because, the focus of this study is on influent and effluent which basically the data does not change. The results based on volume could also be used for easy comparison with other LCA studies. Functional unit 2 (FU2) defined as 1 kgPO_4^{3}-eq removed was used for comparison with the result from functional unit 1 (FU1) to identify the difference. FU2 was also used by Rodriguez-Garcia et al., 2011 and Comas Matas, 2012. For calculation, mass load of eutrophying substances removed, i.e. chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) in MSTP were converted to kgPO_4^{3}-eq using the characterisation factor from eutrophication potential impact category as defined in the CML-IA baseline v3.04 methodology. The illustrated system boundary for this LCA - WWTP study is shown in **Figure 11**.

4.2.2.3 Life cycle inventory (LCI)

The data of plant operation and performance were from both plant managers and sampling conducted in Malaysian STP in August 2017, which were converted to life cycle inventory (LCI). LCI in this study are comprises of the following parameters:

1) The direct pollutants emission including TCOD, TN, TP, metals and PPCPs in influent and effluent; and metals in sludge

2) The direct gas emissions from wastewater treatment such as CO_2, CH_4 and N_2O as outputs; calculated based on 100-year time horizon, by referring to the Intergovernmental Panel on Climate Change guidelines (IPCC, 2006). N_2O emission was mainly generated from biological nitritation and denitrification which might occur in WWTPs, while CH_4 emission was from anaerobic/sludge treatment (Masuda et al., 2015)

3) The indirect resources such as electricity consumption for pumping, aeration, stirring, chemical consumption and sludge disposal. Background data for indirect emissions were selected from the Ecoinvent v3.3 as described below:

a. Electricity production was taken from the Ecoinvent v3.3 based on Malaysia database.
b. Chemical production: For polyelectrolyte, a similar production process of acrylonitrile was selected from the Ecoinvent v3.3 as suggested by Rodriguez-Garcia et al., 2011.
c. Transport vehicles: Lorry with a capacity of 3.5-7.5 metric tons was selected for the disposal of sludge/biosolids in landfill.

To understand how micropollutants in developing countries particularly with highly diluted wastewater affect LCA results, an extended life cycle inventory (LCI) including toxic pollutants such as metals and PPCPs was used. In order to study effects of heavy metals and PPCPs on toxicity assessment, three scenarios were analysed: Scenario 1 did not consider heavy metals and PPCPs in LCI, which was typical in the most studies on LCA of wastewater treatment plants; scenario 2 included ten PPCPs and nine heavy metals in the liquid effluent; and scenario 3 encompassed both heavy metals and PPCPs in the effluent and heavy metals in the sludge. LCI data for all three scenarios are shown in **Table 16**.

Table 16. Life cycle inventories (LCI) of Malaysian STP in three scenarios with/without including direct emissions of metals and PPCPs. Values are presented based on 1 m^3 of treated wastewater (scenarios 1, 2 and 3) and 1 $kgPO_4^3$ -eq. (scenario 3 only) as a functional unit

No.	Description	FU1 Scenario 1	FU1 Scenario 2	FU1 Scenario 3	FU2 Scenario 3	Unit/m^3
	Indirect Inputs:					
1	Electricity consumption	2.05E-01	2.05E-01	2.05E-01	1.12E+01	kWh
2	Transportation of sludge	6.47E-03	6.47E-03	6.47E-03	3.53E-01	t.km
3	Polymer (polyelectrolyte)	5.15E-04	5.15E-04	5.15E-04	2.81E-02	kg
	Emissions to air:					
4	Carbon dioxide, CO_2 (biogenic)[a]	8.90E-02	8.90E-02	8.90E-02	4.85E+00	kg
5	Methane, CH_4	1.10E-03	1.10E-03	1.10E-03	5.99E-02	kg
6	Dinitrogen monoxide, N_2O	4.60E-04	4.60E-04	4.60E-04	2.51E-02	kg
	Emissions to water:					
7	Total chemical oxygen demand, TCOD	4.47E-02	4.47E-02	4.47E-02	2.43E+00	kg
8	Total nitrogen, TN	1.50E-02	1.50E-02	1.50E-02	8.17E-01	kg
9	Total phosphorus, TP	1.05E-03	1.05E-03	1.05E-03	5.74E-02	kg
10	Mercury, Hg	-	5.00E-09	5.00E-09	2.72E-07	kg
11	Cadmium, Cd	-	2.14E-07	2.14E-07	1.17E-05	kg
12	Lead, Pb	-	1.69E-06	1.69E-06	9.19E-05	kg
13	Copper, Cu	-	2.14E-06	2.14E-06	1.16E-04	kg
14	Nickel, Ni	-	6.81E-06	6.81E-06	3.71E-04	kg
15	Zinc, Zn	-	9.98E-05	9.98E-05	5.44E-03	kg
16	Chromium VI, Cr VI	-	2.13E-06	2.13E-06	1.16E-04	kg
17	Arsenic, As		6.00E-09	6.00E-09	3.27E-07	kg
18	Antimony, Sb		1.00E-07	1.00E-07	5.45E-06	kg
	<u>*PPCPs*</u>					
20	17α-ethinylestradiol	-	7.0E-10	7.0E-10	3.81E-08	kg
21	17β-estradiol	-	6.0E-10	6.0E-10	3.27E-08	kg
22	Bisphenol-A	-	6.5E-08	6.5E-08	3.54E-06	kg
23	Carbamazepine	-	2.6E-08	2.6E-08	4.77E-10	kg
24	Diclofenac	-	8.0E-08	8.0E-08	4.36E-06	kg
25	Estrone	-	5.0E-09	5.0E-09	2.72E-07	kg
26	Ibuprofen	-	1.8E-08	1.8E-08	9.80E-07	kg
27	Nonylphenol mixture	-	3.2E-08	3.2E-08	1.74E-06	kg
28	Triclosan	-	5.0E-09	5.0E-09	2.72E-07	kg
29	Trimethoprim	-	9.0E-09	9.0E-09	4.90E-07	kg
	Emissions to soil:	-	-			kg
30	Mercury, Hg	-	-	4.97E-09	2.71E-07	kg
31	Cadmium, Cd	-	-	4.02E-08	2.19E-06	kg
32	Lead, Pb	-	-	1.15E-06	6.27E-05	kg
33	Copper, Cu	-	-	4.52E-06	2.46E-04	kg
34	Nickel, Ni	-	-	2.12E-06	1.16E-04	kg
35	Zinc, Zn	-	-	1.61E-05	8.78E-04	kg
36	Chromium VI, Cr VI	-	-	1.33E-06	7.25E-05	kg
37	Arsenic, As	-	-	5.56E-09	3.03E-07	kg
38	Antimony, Sb	-	-	5.15E-07	2.80E-05	kg

Notes: [a] Carbon dioxide emission from biological processes in WWTPs is considered biogenic based on the IPCC guideline and was not included in the LCA analysis (IPCC Guidelines for National Greenhouse Gas Inventories, 2006)

4.2.2.4 Life cycle impact assessment (LCIA) and interpretation

Life cycle assessment (LCA) was conducted for three scenarios by using CML-IA v3.04 methodology to identify the different environmental impacts between conventional data and extended data including metals and PPCPs. CML-IA method was chosen in the scenarios analysis because it had been widely used in LCA assessment of WWTPs for midpoint impact categories including toxicity impact by Renou et al., 2007; Munoz et al., 2008; Lorenzo-Toja et al., 2016 and it is an available method in various LCA softwares including SimaPro and Gabi. Although USEtox is recommended for toxicity analysis, CFs of dominant metals and PPCPs provided in USEtox are still interim (Pizzol et al., 2011), leading to no superiority compared with other models in terms of wastewater treatment plants. Furthermore, only human toxicity and freshwater ecotoxicity potentials are considered in USEtox. Thus, given the importance of heavy metals to terrestrial impact especially when biosolids are disposed to landfills or used in agriculture lands, CML-IA was selected for the main analysis because it provides terrestrial ecotoxicity as one of midpoint impact categories, and it is easier to compare with other researchers' results. In the scenarios comparison, eight potential impact categories such as global warming potential (GWP), eutrophication potential (EP), abiotic depletion (fossil fuel) potential (ADFP), ozone layer depletion potential (OLDP), acidification potential (AP), human toxicity potential (HTP), freshwater ecotoxicity potential (FEP) and terrestrial ecotoxicity potential (TEP) were chosen. The results were assessed/interpreted by the contribution of different components such as electricity consumption, chemicals consumption, transportation and direct emission of pollutants including GHGs to each environmental impact category.

For model comparison in detail toxicity assessment that includes metals and PPCPs, two LCIA methods namely CML-IA and USEtox were used to investigate and compare effects of the selected metals and PPCPs on toxicity impact categories, i.e. human toxicity potential (HTP), freshwater ecotoxicity potential (FEP) and terrestrial ecotoxicity potential (TEP), with the inclusion of local data of metals and PPCPs in the inventory. USEtox was selected for this analysis because it is the latest developed consensus-model for toxicity categories. The impact values obtained by different methods cannot be compared directly because different units are used in different models. For example, kg 1.4 dichlorobenzene (1.4-DB) eq. is used in CML-IA whilst USEtox shows results with the comparative toxic unit (CTU). Therefore, only relative scores as percentages within each model were calculated for the comparison purpose. CF values of metals and PPCPs by using USES-LCA and

USEtox are tabulated in **Table 17** and **Table 18**, which were referred to (Alfonsin et al., 2014; Ortiz de Garcia et al., 2017; Li et al., 2019) and the databases in CML-IA and USEtox via Simapro v8.5.

In addition, to further study the effects of LCIA methods on toxicity impacts mainly by heavy metals, five LCIA methods namely CML-IA, Recipe, IMPACT 2002+, EDIP 2003 and USEtox were used to compare results of midpoint toxicity impact categories such as human toxicity (HTP), freshwater ecotoxicity (FEP) and terrestrial ecotoxicity (TEP) potentials using LCI in scenario 3. These methods were selected due to the availability of suitable toxicity impacts categories in each method and their wide use in LCA-WWTP research for toxicity related studies (Pizzol et al., 2011; Lorenzo-Toja et al., 2016; Munoz et al., 2008; Li et al., 2019). In CML-IA/Recipe methods, human toxicity potential is considered, and ecotoxicity is separated into three impact categories: freshwater ecotoxicity potential (FEP); terrestrial ecotoxicity potential (TEP) and marine ecotoxicology potential (MEP). The impact of these categories is indicated as 1,4-dichlorobenzene (DCB) (Ref: http://cml.leiden.edu/software/data-cmlia.html and www.lcia-ReCiPe.net. EDIP 2003 method provide human toxicity and ecotoxicity potentials. Human toxicity potential in EDIP is however consist of three different exposure routes (i.e. HTP via water, HTP via soil and HTP via air). While, ecotoxicity potential in EDIP comprise of three different impact categories (i.e. acute FEP, chronic FEP, and chronic TEP), and the impact is expressed as volume (m^3) (Ref: http://www.lca-center.dk/cms/site.aspx?p=4441. IMPACT 2002+ method provides human toxicity as carcinogens and non-carcinogen, aquatic ecotoxicity and terrestrial ecotoxicity potentials. These impacts are expressed in different units (e.g. kg C_2H_3Cl eq. for human toxicity and kg TEG soil for terrestrial ecotoxicity potentials (Ref: http://www.impactmodeling.org. The USEtox method is comprises of six different emission compartments (i.e. rural air, urban air, seawater, freshwater, agricultural soil and natural soil). It consists of human toxicity with both cancer and non-cancer effects, and freshwater (aquatic) ecotoxicity potential (Rosenbaum et al., 2008). USEtox result is expressed as comparative toxic units (CTUh/kg) (Ref: www.usetox.org).

Characterisation factors (CFs) are available in several life cycle impact assessment (LCIA) methods, such as the International Reference Life Cycle Data System (ILCD) 2011, Impact 2002+, CML-IA, USEtox or EDIP 2003. CF models are built based on mechanisms of cause-effect chains starting from emissions to impacts to calculate CF values, which are the total results from environmental fate, exposure and the effects on the receiving compartment such as human, freshwater and terrestrial (Huijbregts et al., 2005). CFs were thus calculated by multiplying fate factor (FF) with exposure factor (XF), and effect factor (EF) as shown in **equation 4.2.2.4-1** (Hou et al., 2020; Rosenbaum et al., 2008; Hedberg et al., 2019). Fate factor (FF) represents the mobility of pollutants to the receiving compartments such as through ingestion by human, runoff to freshwater or adsorption in soil.

Exposure factor (XF) relates to the concentration of substances taken by the receiving compartment. Effect factor (EF) is related to the effect level in the receiving compartment. Fate factor and exposure factor are combined to form intake factor (IF) of a substance as shown in **equation 4.2.2.4-1**. For LCIA, CFs are multiplied with the inventory data (LCI) that emitted to air, water and soil compartments, to get the potential toxicity impact as shown in **equation 4.2.2.4-2** (Ortiz et al., 2013) which is expressed in specific unit contribution. However, not all of CFs of chemicals in the above mentioned models are available due to the data limitation, and some authors have to compute CFs of some chemicals with these models by collecting more fate, exposure and effect data from literature.

a. Characterisation factor (CF)

= fate factor (FF) x exposure factor (XF) x effect factor (EF)

= intake factor (IF) x effect factor (EF) (**Equation 4.2.2.4-1**)

b. Example of impact: Freshwater ecotoxicity potential (FEP)

$= (LCI_{air} \times CF_{air}) + (LCI_{water} \times CF_{water}) + (LCI_{soil} \times CF_{soil})$ (**Equation 4.2.2.4-2**)

Table 17. Characterisation factors (CFs) for 9 metals by CML-IA and USEtox. Emission compartment: air, freshwater (water) and soil

CFs by CML-IA	HTP(air)	HTP(water)	HTP(soil)	FEP(air)	FEP(water)	FEP(soil)	TEP(air)	TEP(water)	TEP(soil)
Unit	kg1.4DB eq.	kg1.4DB eq.	kg1.4DB eq.	kg1.4DB eq.	kg1.4DB eq.	kg1.4DB eq.	kg1.4DB eq.	kg1.4DB eq.	kg1.4DB eq.
Antimony, Sb	6.71E+03	5.14E+03	2.63E+03	3.72E+00	1.97E+01	9.98E+00	6.11E-01	1.66E-20	1.25E+00
Arsenic, As	3.48E+05	9.51E+02	1.02E+03	4.95E+01	2.07E+02	1.34E+02	1.61E+03	1.04E-17	3.34E+03
Cadmium, Cd	1.45E+05	2.29E+01	6.67E+01	2.89E+02	1.52E+03	7.76E+02	8.12E+01	1.42E-20	1.67E+02
Chromium VI, Cr VI	3.43E+06	3.42E+00	5.00E+02	7.69E+00	2.77E+01	2.10E+01	3.03E+03	2.27E-19	6.30E+03
Copper, Cu	4.30E+03	1.34E+00	1.25E+00	2.22E+02	1.16E+03	5.95E+02	6.99E+00	4.06E-21	1.44E+01
Lead, Pb	4.67E+02	1.23E+01	2.93E+02	2.40E+00	9.62E+00	6.53E+00	1.57E+01	4.77E-22	3.25E+01
Mercury, Hg	6.01E+03	1.43E+03	1.08E+03	3.17E+02	1.72E+03	8.48E+02	2.83E+04	9.30E+02	5.60E+04
Nickel, Ni	3.50E+04	3.31E+02	1.98E+02	6.29E+02	3.24E+03	1.69E+03	1.16E+02	1.03E-18	2.39E+02
Zinc, Zn	1.04E+02	5.84E-01	4.22E-01	1.78E+01	9.17E+01	4.77E+01	1.20E+01	2.53E-21	2.46E+01
CFs by USEtox	HTPcancer (air)	HTPcancer (water)	HTPcancer (soil)	HTPnon-cancer(air)	HTPnon-cancer(water)	HTPnon-cancer(soil)	FEP(air)	FEP(water)	FEP(soil)
Unit	CTUh	CTUh	CTUh	CTUh	CTUh	CTUh	CTUe	CTUe	CTUe
Antimony, Sb	n.a	n.a	n.a	1.87E-04	3.64E-04	1.83E-04	4.89E+02	1.22E+03	6.15E+02
Arsenic, As	4.24E-04	3.69E-04	1.95E-04	1.71E-02	2.73E-02	1.45E-02	1.16E+04	2.78E+04	1.47E+04
Cadmium, Cd	2.52E-04	1.59E-06	8.08E-07	4.45E-02	4.27E-04	2.17E-04	3.92E+03	9.71E+03	4.94E+03
Chromium VI, Cr VI	4.70E-03	1.06E-02	5.34E-03	7.48E-04	2.40E-05	1.20E-05	5.17E+02	1.29E+03	6.50E+02
Copper, Cu	n.a	n.a	n.a	1.32E-05	8.63E-07	4.55E-07	2.31E+04	5.52E+04	2.92E+04
Lead, Pb	2.66E-05	3.42E-07	2.02E-07	9.32E-03	1.20E-04	7.08E-05	1.74E+02	3.75E+02	2.21E+02
Mercury, Hg	6.89E-03	1.20E-04	8.48E-05	8.15E-01	1.42E-02	1.00E-02	1.21E+04	2.21E+04	1.56E+04
Nickel, Ni	5.62E-05	3.83E-05	1.97E-05	3.16E-06	2.15E-06	1.11E-06	6.08E+03	1.49E+04	7.66E+03
Zinc, Zn	n.a	n.a	n.a	1.52E-02	1.28E-03	7.02E-04	1.67E+04	3.86E+04	2.11E+04

Reference: CML-IA (http://cml.leiden.edu/software/data-cmlia.html); USEtox (www.usetox.org)

Note: n.a means that the substance does not have impact on human toxicity cancer (HTPcancer)

Table 18. Characterisation factors (CFs) for 10 PPCPs by CML-IA and USEtox. Emission compartment: freshwater environment

Toxicity categories	CFs by CML-IA						CFs by USEtox					
	HTP	Ref.	FEP	Ref.	TEP	Ref.	HTcancer	Ref.	HTnon-cancer	Ref.	FEP	Ref.
Unit	kg1.4DB eq.		kg1.4DB eq.		kg1.4DB eq.		CTUh		CTUh		CTUe	
17α-ethinylestradiol	2.37E+04	a	1.53E+08	a	9.33E-04	a	3.76E-04	c	1.63E-01	c	1.69E+06	b,c
17β-estradiol	2.59E+03	a	8.40E+08	a	4.04E-05	a	1.14E-03	c	1.04E-03	c	1.84E+08	b,c
Bisphenol-A	1.51E+00	d	1.57E+01	d	2.38E-08	d	4.66E-07	c	7.00E-07	c	5.16E+03	c
Carbamazepine	4.79E-01	a	2.32E+00	a	2.20E-07	a	n.a*	c	7.68E-06	c	8.54E+02	b,c
Diclofenac	6.30E+00	a	1.58E+02	a	6.63E-07	a	1.16E-04	c	1.15E-04	c	2.67E+03	b,c
Estrone	6.12E+02	a	2.17E+05	a	5.37E-04	a	n.a*	c	4.86E-04	c	2.14E+04	b,c
Ibuprofen	4.88E+00	a	1.54E+05	a	2.60E-02	a	n.a*	e	3.71E-07	e	2.09E+02	b,e
Nonylphenol	1.51E-01	d	2.67E+01	d	1.93E-06	d	n.a*	e	3.38E-07	e	1.47E+04	e
Triclosan	9.48E+01	a	3.55E+06	a	4.45E-01	a	n.a*	c	2.21E-07	c	1.06E+05	b,c
Trimethoprim	1.26E-01	a	3.05E+01	a	4.11E-08	a	9.42E-07	c	5.66E-07	c	4.74E+02	b,c

Ref. = reference; (References: a = Alfonsin et al., 2014; b= Garcia et al., 2017; c= Li et al., 2019; d = CML-IA (http://cml.leiden.edu/software/data-cmlia.html); e = USEtox (www.usetox.org).

Note: n.a means that the substance does not have impact on human toxicity cancer (HTcancer)

4.2.2.5 Sensitivity analysis

Throughout the operation of Malaysian STP, there is potential of variation in the concentration of pollutants in sludge, influent and effluent that affecting electricity consumption. Thus, sensitivity analysis was conducted to assess ±10% variations of priority data such as metals in sludge, metals and PPCPs in effluent, and electricity consumption to toxicity impact results. For metals in effluent and sludge, the most affected/toxic pollutants such as nickel, zinc and copper were selected. While for PPCPs in effluent, the top 3 toxic pollutants such as 17α-ethinylestradiol, 17β-estradiol and triclosan were selected. Each of the selected pollutant were analysed for sensitivity to identify if the increase/reduction in concentration of each pollutant can cause more or less changes in toxicity potential results. FU1, i.e. per m^3 treated wastewater was selected for this analysis to facilitate the comparison with the results from other studies.

4.3 Results and discussion

4.3.1 Occurrence of heavy metals and PPCPs in wastewater and their removal by wastewater treatment processes

Although sewage plants are not designed for the removal of soluble PPCPs and heavy metals, these pollutants can still be removed in treatment processes to some extents. The removal efficiencies of PPCPs and heavy metals in this study with the SBR system varied from 9% to 99% except for lead, zinc, cadmium, and trimethoprim as shown in **Table 19**. Heavy metals cannot be degraded because they are inorganic elements, but they could be removed by mineralisation or adsorption by activated sludge, leading to heavy metal transfer from water to sludge. A large variation in heavy metal removal efficiencies ranging from 8.8% to 73.0% was found in this study. The results of heavy metal removal efficiencies in this study were generally lower than those reported by Üstün, 2009. They examined the removal of heavy metals such as cadmium, chromium, zinc, nickel and lead in an activated sludge process with a continuous aeration tank in Turkey and found that the removal efficiencies ranged between 47% and 95%. Nonetheless, Lorenzo-Toja et al. (2016) found a larger variation in removal efficiencies of heavy metals in Spain, ranging from 33% to 97%. **Table 20** shows the comparison of heavy metal concentrations and removal efficiencies in developing and developed countries with different wastewater strengths. Unlike BOD and nutrient removal efficiencies, which are usually lower in developing countries, no clear patterns in heavy metal concentrations and removal efficiency were found between developing and developed countries.

This highlights the dependence of heavy metal concentrations on local situations and thus necessity of sampling for heavy metal studies due to the complexity of metal migration between aqueous solutions and sludge. In addition, in this study, it was found that concentrations of lead, zinc, and cadmium in the effluent even higher than those in the influent, resulting in negative removal efficiencies. This phenomenon might be due to the solubilisation/hydrolysis of particulates in biodegradation process, desorption of soluble metals from particulates in the wastewater treatment process, or dissolution from metal precipitates in wastewater treatment conditions. These multiple possibilities suggest complexity and unpredictability of heavy metal removals from aqueous solutions. In terms of toxicity, the transfer of heavy metals from aqueous wastewater to sludge means that toxicity could transfer from wastewater to land if sludge is finally disposed of in a landfill or applied in agriculture land, which can transfer back to freshwater by runoff.

Table 19. Measured concentrations of organic pollutants, heavy metals and PPCPs in influent, effluent and sludge at Malaysian STP

No	Substances	Influent concentrations (mg/L)	Effluent concentrations (mg/L)	Removal efficiency (%)	
1	$TBOD_5$	126.00 ±1.20	10.12 ±0.42	92.0	-
2	TSS	174.00	13.50	92.0	-
3	TCOD	433.00 ± 2.00	44.67 ±1.15	90.2	-
4	O&G	15.00	3.00	80.0	-
5	TP	2.59 ±0.07	1.05 ±0.04	60.0	-
6	TN	27.00	15.00	44.5	-
	Heavy metals:	Influent concentrations (mg/L)	Effluent concentrations (mg/L)	Removal efficiency (%)	Dry weight sludge (mg/kg)
7	Arsenic, As	0.022±0.009	0.006±0.005	73.0	0.081±0.008
8	Antimony, Sb	0.25±0.011	0.10±0.009	60.0	7.5±0.33
9	Mercury, Hg	0.012±0.006	0.005±0.009	57.1	0.072±0.007
10	Copper, Cu	4.797±0.145	2.138±0.026	55.4	65.90±0.66
11	Chromium VI, Cr VI	3.763±0.086	2.133±0.079	43.3	19.40±0.08
12	Nickel, Ni	7.472±0.415	6.814±0.076	8.8	30.90±0.66
13	Lead, Pb [a]	0.966±0.027	1.688±0.015	-42.8	16.80±0.21
14	Zinc, Zn [a]	64.565±4.227	99.829±7.046	-35.3	234.80±7.60
15	Cadmium, Cd [a]	0.160±0.018	0.214±0.018	-25.2	0.60±0.01
	PPCPs:	µg /L	µg /L	%	
16	17α-ethinylestradiol	0.083	0.001	99.2	-
17	17β-estradiol	0.024	0.001	97.6	-
18	Bisphenol-A	2.690±1.070	0.065±0.026	97.6	-
19	Carbamazepine	0.049	0.026	46.9	-
20	Diclofenac	0.191	0.080	57.9	-
21	Estrone	0.168	0.005	97.0	
22	Ibuprofen	0.344±0.138	0.018±0.001	94.8	-
23	Nonylphenol mixture	0.081	0.032	60.5	
24	Triclosan	0.475±0.142	0.005±0.001	98.9	-
25	Trimethoprim [a]	0.028±0.005	0.036±0.007	-22.2	-

Note: [a] Pollutant concentrations in the effluent are higher than those in the influent. (BOD = biochemical oxygen demand, COD= chemical oxygen demand, O&G = oil and grease, TSS = total suspended solids, TN = total nitrogen, TP = total phosphorus). Standard deviations (±) are based on triplicate analysis of samples

Table 20. Comparison of concentrations of heavy metals and their removal efficiencies between developed and developing countries with different wastewater strengths

List of heavy metals	Developed countries			References	Developing countries			References
	Influent (mg/L)	Effluent (mg/L)	Removal efficiency %		Influent (mg/L)	Effluent (mg/L)	Removal efficiency %	
1.Arsenic, As	3.49	3.41	3	Yoshida et al., 2014/Denmark*	0.022	0.006	73.0	This study (COD= 433mg/L)
2.Cadmium, Cd	0.12	0.06	50	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	0.160	0.200	-25.2	This study (COD= 433mg/L)
						4.2		Asgharipour & Azizmoghaddam., 2012/Iran (COD = 191mg/L)
3.Chromium VI, Cr VI	0.57	0.23	59.6	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	3.763	2.133	43.3	This study (COD= 433mg/L)
	0.0019-0.0032			Drozdova et al., 2019/chezh republic (COD= 640 – 657 mg/L)				
	0.011			Yoshida et al., 2014/Denmark*				
4.Copper, Cu	6.1	4.1	32.8	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	4.797	2.138	55.4	This study (COD= 433mg/L)
	0.013-0.065			Drozdova et al., 2019/Czech republic (COD= 640 – 657 mg/L)		0.34		Asgharipour & Azizmoghaddam., 2012/Iran (COD = 191mg/L)
	0.097			Yoshida et al., 2014/Denmark*	3.1	0.8	74.2	Ramadan et al., 2016/Egypt*
5.Antimony, Sb	0.173	0.204	-20.4	Hargreaves et al., 2016/UK (COD = 622mg/L)	0.25	0.1	60.0	This study (COD= 433mg/L)

Heavy metals	Developed countries			References	Developing countries			References
	Influent (mg/L)	Effluent (mg/L)	Removal efficiency %		Influent (mg/L)	Effluent (mg/L)	Removal efficiency %	
6.Lead, Pb	0.001-0.038			Drozdova et al., 2019/ Czech republic (COD= 640 – 657 mg/L)	0.970	1.700	-42.8	This study (COD= 433mg/L)
	0.017			Yoshida et al., 2014/Denmark*	1.15	0.7	39.1	Ramadan et al., 2016/Egypt*
	0.0044			Yoshida et al., 2014/Denmark*		0.02		(Asgharipour & Azizmoghaddam., 2012)/Iran (COD = 191mg/L)
7.Mercury, Hg	0.0049			Yoshida et al., 2014/Denmark*	0.012	0.005	58.3	This study (COD= 433mg/L)
8.Nickel, Ni	2.0	1.13	43.5	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	7.47	6.81	8.83	This study (COD= 433mg/L)
	0.003-0.005			Drozdova et al., 2019/Czech republic (COD= 640 – 657 mg/L)		0.24		(Asgharipour & Azizmoghaddam., 2012)/Iran (COD = 191mg/L)
	0.018			Yoshida et al., 2014/Denmark*				
9.Zinc, Zn	36.2	1.9	94.8	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	64.600	99.800	-35.3	This study (COD= 433mg/L)
	0.114-0.403			Drozdova et al., 2019/ Czech republic (COD= 640 – 657 mg/L)	20	11.3	43.5	Ramadan et al., 2016/Egypt*
	0.373			Yoshida et al., 2014/Denmark*		0.71		(Asgharipour & Azizmoghaddam., 2012)/Iran (COD = 191mg/L)

Note: * means that COD value is not available in the study

PPCPs were chosen as contaminants of emerging concern to analyse in this study due to their continuous release into aquatic ecosystems where their effects may go unnoticed (Lorenzo-Toja et al., 2016). In addition, a study by Munoz et al. (2008) concluded that PPCPs are more toxic than other priority pollutants such as polycyclic aromatic hydrocarbons (PAHs), biocides, and organic priority pollutants such as trichlorobenzene. PPCPs are organic chemicals, which can be degraded by activated sludge to certain extents or removed from aqueous solution through adsorption by activated sludge or air stripping (Sagban, 2014). **Table 19** shows that all tested PPCPs were removed with efficiencies ranging from 46.9% to 99.2%, except for trimethoprim. The removal efficiencies of micropollutants vary from one study to another as it greatly depends on influent concentrations, treatment technologies adopted (Kasprzyk-Hordern et al., 2009), and operation conditions of treatment facilities (Ustun, 2009). For instance, Kasprzyk-Hordern et al. (2009) reported that a local WWTP in the UK with an extended aeration oxidation ditch had over 85% removal efficiencies for 37 pharmaceuticals, 13 personal care products, 3 illicit drugs, and 2 endocrine disruptors, indicating that this type of treatment provided good removal efficiencies for PPCPs. In addition, a review by Yang et al. (2017) highlighted that conventional wastewater treatment in seven different countries removed 21 PPCPs with efficiencies ranging from 74% to 99% with an exception of trimethoprim, which had negative removal efficiency with a higher concentration in the effluent than in the influent, which is the same as this study. A similar phenomenon of negative PPCPs removals was reported by Üstün, 2009; Sun et al., 2014; Wang et al., 2015. Higher concentrations of PPCPs in effluent than influent may be attributed to the desorption of pollutants from particulates during treatment and low biodegradability levels (Üstün, 2009). In developed countries, more advanced processes are used for nutrients removal as well, which requires longer sludge retention times (SRTs) and thus might be beneficial for degradation of PPCPs due to long explosion to activated sludge. By contrast, WWTPs in developing countries usually do not have nutrient removal, resulting in shorter SRTs. It is reasonably speculated that long SRTs generally benefit to PPCPs degradation. However, similar to heavy metals, concentrations and removal efficiencies of PPCPs between different wastewater treatment plants in developed and developing countries do not show any clear patterns (**Table 21**), suggesting unpredictability of PPCPs concentrations and their removal efficiencies in sewage treatment plants.

Table 21. Comparison of concentrations of PPCPs and their removal efficiencies between developed and developing countries with different wastewater strengths

List of PPCPs	Developed countries			References	Developing countries			References
	Influent (µg/L)	Effluent (µg/L)	Removal efficiency %		Influent (µg/L)	Effluent (µg/L)	Removal efficiency %	
17α-ethinylestradiol	0.628	0.002	99.7	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	0.85	0.23	72.8	Tan et al., 2015/Malaysia*
					0.083	0.001	99.2	This study (COD= 433mg/L)
17β-estradiol	0.04	0.0005	98.8	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	0.024	0.001	97.6	This study (COD= 433mg/L)
	0.022	0.0028	87.2	Miège et al., 2009/France*				
Acetaminophen/ paracetamol	61	0.86	99	Benotti and Brownawell, 2007/USA*	4.24	0.115	97.3	Tan et al., 2015/Malaysia*
	211.4	117.3	44.5	Kasprzyk-Hordern et al., 2009/ UK*				
	23.2	0.1	99.0	Rosal et al., 2010/Spain (COD = 269 mg/L)				
Bisphenol-A	0.5	0.077	84.6	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	2.690	0.065	97.6	This study (COD= 433mg/L)
	0.42	0.086	79.5	Kasprzyk-Hordern et al., 2009/ UK*				
Caffeine	42	15.2	64	Benotti and Brownawell, 2007/USA*	5.860	0.108	98.2	Sui et al., 2011/China*
	22.9	1.18	94.9	Rosal et al., 2010/Spain (COD = 269 mg/L)	25.6	0.115	99.5	Tan et al., 2015/Malaysia*
					15.700	0.016	99.9	This study (COD= 433mg/L)
Carbamazepine	0.1	0.065	37	Benotti and Brownawell, 2007/USA*	0.0285	0.0163	42.8	Sui et al., 2011/China*
	0.103	0.104	-1.0	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)				

	0.129	0.117	9.8	Rosal et al., 2010/Spain (COD = 269 mg/L)				
Diclofenac	0.069	0.098	-42.0	Kasprzyk-Hordern et al., 2009/ UK*	0.286	0.185	35.3	Sui et al., 2011/China*
	1.34	0.68	49.3	Miege et al., 2009/France*	2.0	0.632	68.3	Tan et al., 2015/Malaysia*
	0.232	0.22	5	Rosal et al., 2010/Spain (COD = 269 mg/L)				
Estrone	0.257	0.01	96.1	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	4.2	0.36	91.6	Tan et al., 2015/Malaysia*
					0.168	0.005	97.0	This study (COD= 433mg/L)
Furosemide	1.48	1.16	21.6	Kasprzyk-Hordern et al., 2009/ UK*				
	0.413	0.166	59.8	Rosal et al., 2010/Spain (COD = 269 mg/L)				
Hydrochlorothiazide	2.5	1.18	53.2	Rosal et al., 2010/Spain (COD = 269 mg/L)	0.010	0.028	-180.0	This study (COD= 433mg/L)
Ibuprofen	17.4	0.12	99.3	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)	0.344	0.018	94.8	This study (COD= 433mg/L)
	1.681	0.263	84.4	Kasprzyk-Hordern et al., 2009/ UK*				
	14.6	1.96	86.6	Miege et al., 2009/France*				
	2.69	0.14	95	Rosal et al., 2010/Spain (COD = 269 mg/L)				
Metoprolol	0.075	0.069	8	Kasprzyk-Hordern et al., 2009/ UK*	0.103	0.105	-2	Sui et al., 2011/China*
	0.16	0.338		Miege et al., 2009/France*	0.62	0.17	72.5	Tan et al., 2015/Malaysia*
					0.045	0.062	-37.8	This study (COD= 433mg/L)
Nonylphenol mixture	0.04	0.07	-42	Gatidou et al., 2007*	0.081	0.032	60.5	This study (COD= 433mg/L)
Salbutamol	13	8.1	36	Benotti and Brownawell, 2007/USA*	0.008	0.003	62.5	This study (COD= 433mg/L)
	0.32	0.234	26.9	Kasprzyk-Hordern et al., 2009/ UK*				
Salicyclic acid	5.87	0.164	97.2	Kasprzyk-Hordern et al., 2009/ UK*				
Trimethoprim	0.3	0.12	60	Benotti and Brownawell, 2007/USA*				

	0.01	0.011	-10	Lorenzo-Toja et al., 2016/Spain (COD = 424mg/L)				
Triclosan	0.087	0.025	71.3	Kasprzyk-Hordern et al., 2009/ UK*	0.475	0.005	98.9	This study (COD= 433mg/L)
	0.38	0.15		Miege et al., 2009/France*				
	0.86	0,219	74.5	Rosal et al., 2010/Spain (COD = 269 mg/L)				

Note: * means that COD value is not available in the study

Table 22 further compares concentrations of metals and PPCPs in Spain and Malaysia with similar wastewater strength, i.e. an average COD concentration of 378 mg/L in influent (year 2017) in this study (Malaysia), and an average COD concentration of 424 mg/L in influent in the Betanzos WWTP in Spain (Lorenzo-Toja et al., 2016). This comparison revealed no pattern in terms of the concentrations of metals and PPCPs in both WWTPs although wastewater strengths are similar. Unlike lower nutrient concentrations in low strength sewage, some concentrations of metals and PPCPs were even higher in the Malaysian STP than those in the Spanish WWTP, indicating the unrelated relationship between concentrations of metals and PPCPs and wastewater strength. They might be closely related to local lifestyles and commodities, demographics and receiving of partial industrial and commercial wastewater in municipal wastewater treatment plants. Unlike general pollutants such as COD, BOD and nutrients, heavy metals and PPCPs show much specificity in each wastewater treatment plants. This further highlights the significance of sampling for studies of toxicity caused by PPCPs and heavy metals in a specific wastewater treatment plant instead of obtaining data from the published literature or simulation to represent the real effects of heavy metals and PPCPs on human and the environment. Although most PPCPs and heavy metal concentrations can be reduced to low levels in aqueous solution, they still have potential toxicity impact on the environment and human health even at low concentrations. In addition, the migration of pollutants from aqueous solution to sludge poses a challenge for safe sludge disposal. Therefore, the inclusion of local PPCPs and heavy metals into toxicity assessments is necessary to provide a holistic view of impact from a wide range of contaminants.

Table 22. Comparison of concentrations of metals and PPCPs in medium strength wastewater in Spain and Malaysia

Pollutants	Concentration range from influent to effluent of MSTP, Malaysia) [a]	Concentration range from influent to effluent of Betanzos WWTP, Spain [b]
Heavy metals	**(mg/L):**	**(mg/L):**
Arsenic	0.022-0.006	3.49-3.41
Antimony	0.25-0.1	Not measured
Mercury	0.012-0,005	0.15-0.57
Copper	4.8-2.1	6.1-4.1
Chromium VI	3.8-2.1	0.57-0.23
Nickel	7.5-6.8	2.0-1.1
Lead	0.97-1.7	5.0-0.39
Zinc	64.6-99.8	36.2-1.9
Cadmium	0.16-0.21	0.12-0.06
PPCPs	**(µg/L):**	**(µg/L):**
17α-ethynilestradiol	0.083-0.001	0.63-0.0007
17β-estradiol	0.024-0.001	0.04-0.0005
Bisphenol-A	2.69-0.065	0.14-not detected
Carbamazepine	0.049-0.026	0.10- 0.07
Diclofenac	0.19-0.08	Not measured
Estrone	0.168-0.005	0.329-0.004
Ibuprofen	0.34-0.018	17.4-0.028
Nonylphenol mixture	0.081-0.032	Not measured
Triclosan	0.475-0.005	0.23-not detected
Trimethoprim	0.028-0.036	0.067-0.011

Notes: [a] This study ; [b] Lorenzo-Toja et al.,2016

4.3.2 Inclusion of PPCPs and heavy metals for LCA of the wastewater treatment plant using the CML-IA method (FU1)

Three scenarios with different inventories (i.e. scenarios 1, 2 and 3) were studied to investigate the effects of inclusion of PPCPs and heavy metals in low strength wastewater of the Malaysian STP into LCA. For each impact category, the result was divided by the sum of the results from three scenarios; thus, 33.3% of each category represented the same results from the three scenarios. As seen in **Figure 12**, the results of impact categories such as abiotic depletion (fossil fuels) potential (ADFP), acidification potential (AP), eutrophication potential (EP), global warming potential (GWP) and ozone layer depletion potential (OLDP) were similar in all three scenarios with or without heavy metals and PPCPs, indicating no impact from micropollutants on these five impact categories. However, including PPCPs and heavy metals in liquid and sludge changed toxicity category results such as freshwater ecotoxicity potential (FEP), terrestrial ecotoxicity potential (TEP) and human toxicity potential (HTP).

Human toxicity impact category by including metals and PPCPs in scenarios 2 and 3 was 3.2% and 4.0%, respectively, higher than that in scenario 1. The small difference is mainly because the indirect emissions from electricity production are the dominant contributors to HTP, accounting for 94%. This result is in agreement with the review done by Zang et al., 2015 on 56 studies related to LCA-WWTPs in both developing and developed countries, which reported that fossil-based electricity, chemical consumption, and sludge incineration (if there is) were the main sources of human toxicity.

However, both FEP and TEP increased significantly by including direct emissions of heavy metals and PPCPs in LCA. FEP increased by 74% in scenario 2 and 76% in scenario 3 compared with that in scenario 1. The increased values are straightforward to understand because the emissions of PPCPs and heavy metals from the effluent have a direct impact on freshwater ecotoxicity. Although the wastewater in this study was much diluted, micropollutants in the effluent at the Malaysian STP were not necessarily lower than those with higher strength wastewater reported in Literature (**Tables 17 and 18**). Metals in the effluent and from electricity generation contributed 65.3% and 23.7% to FEP, respectively, whereas PPCPs only contributed 11%. According to the LCA study by Lorenzo-Toja et al. (2016) for the WWTP with medium strength wastewater in Spain using activated sludge with the extended aeration, the impact of 11 metals and 19 PPCPs in the effluent to FEP was only 10%, whereas electricity and sludge composting contributed to 35% and 55%, respectively. Notably, the removal efficiencies of all toxic pollutants in their study were between 62% and 99%, much higher than those in this study. This indicates that good removal efficiency of toxic pollutants resulted in less contribution of effluent to FEP, linking wastewater treatment technology with FEP.

TEP increased by 88% from scenario 2 to scenario 3 when metals in sludge were considered in LCA. If PPCPs were included as well, it could be expected that the increase in TEP would be even higher. This finding highlights the importance of including data of sludge disposal in terrestrial ecotoxicity analysis. Gallego et al. (2008) reported that seven metals in sludge for agricultural land application from 13 WWTPs with extended aeration, biodenpho, and aerobic-anoxic technologies for less than 20,000 PEs in Spain were the main contributors to terrestrial ecotoxicity, with mercury and chromium contributing 51% and 31%, respectively. The high contribution of sludge metals to TEP indicates the importance of including metals in sludge to terrestrial ecotoxicity impact assessment regardless of the sizes of treatment plants and technologies used.

Overall, the results indicate that the inclusion of PPCPs and heavy metals lead to significant difference in terms of FEP and TEP assessment results even with highly diluted municipal wastewater. Thus, micropollutants and metals should not be omitted from studies of toxicity impacts particularly freshwater toxicity and terrestrial toxicity when sludge is landfilled or applied to the land.

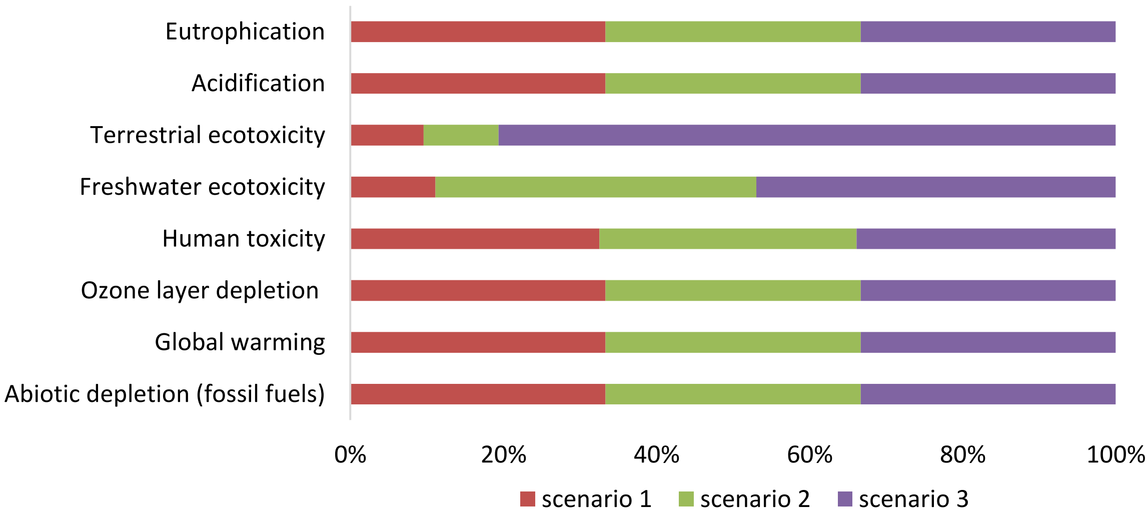
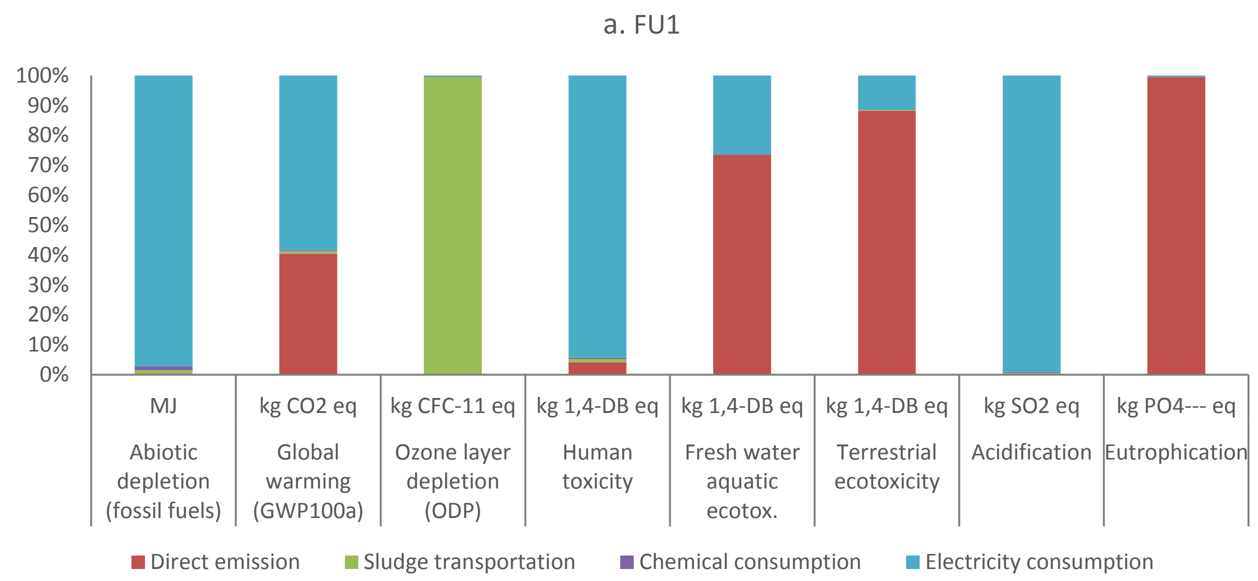


Figure 12. Comparison of eight environmental impact potentials of Malaysian STP with and without considering the direct emissions of heavy metals and PPCPs in three different scenarios, with 1 m^3 of treated wastewater as functional unit.

Note: (Scenario 1 did not consider heavy metals and PPCPs in LCI. Scenario 2 included 10 PPCPs and 9 heavy metals in the liquid effluent, and scenario 3 encompassed both heavy metals and PPCPs in the effluent and heavy metals in the sludge. For each category, the result was divided by the sum of results from three scenarios, thus 33.3% of each category represents the same results from three scenarios.)

4.3.2.1 Comparison of environmental impacts using FU1 (1 m^3 of treated wastewater) and FU2 (eutrophication reduction- 1 kgPO$_4$^{3}-eq) by CML-IA method

Environmental impact analysis using two functional units was conducted to compare the difference of impacts from MSTP. It can be seen from **Figure 13** that the contribution of each input to environmental impacts is similar in both results of FU1 and FU2. For example, electricity was the main contributor with >90% to abiotic depletion (fossil fuel) potential, human toxicity potential and acidification potential by both FUs. While, there is almost negligible contribution by chemical consumption in all eight impact categories by both FUs. This suggests that no much difference is caused by adopting different functional units to single input data of WWTP with low strength wastewater. The similar result of using per m^3 of treated wastewater, and eutrophication reduction (kgPO$_4$^{3}-eq) because the analysis does not involve the change of wastewater flow rate to MSTP. Further analysis in this study adopted FU1 since it has been widely used for LCA assessment of WWTP such as by Piao & Kim, 2016; Lorenzo-Toja et al., 2016; Li et al., 2017; Risch et al., 2018 and for easy comparison to other studies.



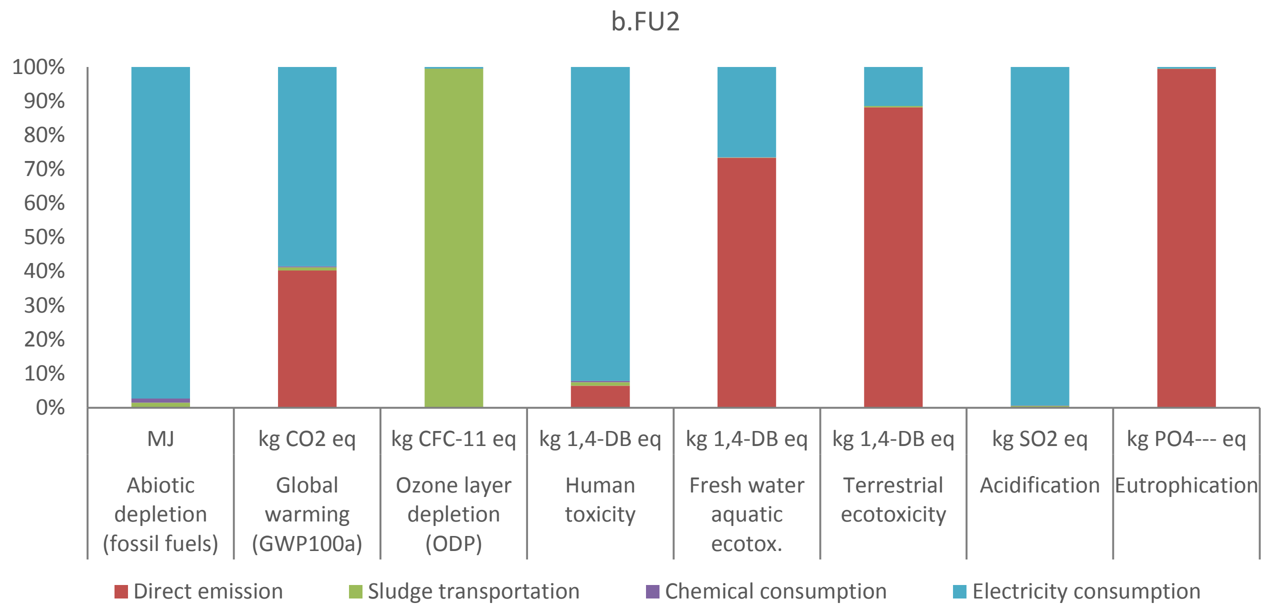


Figure 13. Comparison of eight impact potentials at Malaysian STP by using FU of a. 1m^3 and b. 1 kgPO_4^{3-} eq. removed by CML-IA method

4.3.3 Contribution of PPCPs and heavy metals to toxicity impact categories using CML-IA

It is well known that wastewater treatment is energy intensive. Meanwhile, chemicals are consumed in different operation units such as for denitrification, dewatering, or phosphorus removal. The processes for electricity generation and chemical production emit toxic substances, which cause indirect toxicity to humans and the environment. **Figure 14a** shows the contributions of direct emissions such as PPCPs and heavy metals and indirect emissions such as electricity, sludge transportation and chemicals used in the wastewater treatment plant in this study to toxicity impact categories. As seen, the indirect emissions from electricity consumption accounted for 94.4% of human toxicity potential, indicating negligible contributions from direct emissions such as PPCPs and heavy metals to human toxicity potential (HTP). **Figure 14b** shows the contribution of different processes (i.e pre-treatment, secondary treatment and sludge treatment) to human toxicity, freshwater ecotoxicity and terrestrial ecotoxicity potentials. Secondary treatment (SBR) is the highest contributor with an average 60% to all three toxicity categories. This result highlights the importance of reducing energy consumption in the secondary treatment to decrease human toxicity, freshwater ecotoxicity and terrestrial ecotoxicity potentials. The main toxic substances emitted from electricity generation in Malaysia were barium, hydrogen fluoride, and nickel when 93% of electricity was generated from fossil fuels. As reported by Almudena Hospido et al. (2008) on the human toxicity impact by the CML2000 method for 4 municipal WWTPs in Spain, energy consumption contributed to 85%, and the remaining 15% was from direct pollutants in effluent and sludge. Piao et al. (2016) reported that 70-89% of the impact to HTP by WWTPs in Korea was attributed to the emissions of nickel, barium, and nitrogen oxides from the generation of electricity. In Korea, 74% of electricity generated is from fossil fuels. All these consistent results indicate that the consumption of fossil fuels for electricity generation is much related to HTP results by WWTPs. Therefore, moving fossil fuels to clean energy such as natural gas and hydrogen, solar and wind energy can reduce human toxicity effectively. In addition, if WWTPs could achieve energy neutrality, i.e. energy recovered from wastewater can cover electricity consumed for treatment, HTP would be significantly reduced. Currently, energy recovery from wastewater and improving energy efficiency in WWTPs are being intensively studied. It is expected that the current trends to shift electricity generation from fossil fuels to that from renewable energy would mitigate HTP. Since the WWTP in this study was designed and operated for suspended solid and COD removal only, no chemicals were needed for denitrification or phosphorus removal. The only chemical used was acrylonitrile, which was added to enhance sludge dewatering. Acrylonitrile used in this WWTP had a negligible impact on toxicity. If more or different types of chemicals were used in different units in WWTPs, the toxicity impact of chemicals would be different from this study. For example, Niero et al. (2014) reported that ferric chloride consumption in WWTPs for phosphorus removal contributed 20 - 40% to human toxicity. This comparison indicates a trade-off between toxicity and eutrophication impact category when chemicals are used for nutrient removal. Thus, the technology selection for phosphorus removal should not be just focused on nutrient removal and chemical cost only. Toxicity can also be used as one of the important criteria for technology selection from a holistic point of view. Finally, the transportation contribution to toxicity was also negligible, which is in agreement with the study by Piao et al., 2015. This highlights that transportation is not the main concern to toxicity impact categories.

The FEP results are shown in **Figure 14a** and the dominant pollutants that contribute to FEP are shown in **Table 23**. Direct emissions of metals and PPCPs from the Malaysian STP contributed 76.3% to freshwater ecotoxicity with the toxicity caused mainly by nickel (34%), zinc (14.2%), and 17β-estradiol (7%) from the effluent. Almudena Hospido et al. (2008) reported that seven heavy metals in both effluent and sludge contributed 75% to freshwater ecotoxicity potential. However, their research did not include PPCPs in the analysis due to the lack of relevant information. It is found in this study that the contribution of PPCPs in the effluent to FEP was 11%, mainly from 17β-estradiol, triclosan, and 17α-ethinylestradiol, suggesting more concerns about the negative impact of heavy metals on water bodies. It needs to point out that 10 PPCPs were investigated in this study according to Malaysian local situation and only 3 of them shows obvious contribution. If that PPCPs number for toxicity assessment was expanded can result in higher toxicity contribution is dependent on the toxicity level of the included specific type of chemical. For example, Li et al. (2019) included 126 PPCPs for LCA toxicity assessment using the USEtox model in different advanced wastewater treatment processes in China such as ozonation, granular activated carbon adsorption and reverse osmosis, and found that only 5% of FEP was from this 126 PPCPs, in which the main contributors were 17α-ethinylestradiol and 17β-estradiol. The remaining 95% contribution to FEP were mainly from electricity consumption and metal emissions in effluent. Results from both studies indicate that the number of PPCPs considered is not critical, but the prioritised (i.e. top ranked) PPCPs such as 17α-ethinylestradiol and 17β-estradiol are more important in the toxicity study involving micropollutants/PPCPs. Thus, more research is needed for the identification of PPCPs with higher toxicity to guide LCA toxicity assessment. In addition, with the current knowledge, the main concern regarding FEP is still from heavy metals although the inclusion of PPCPs can lead to a more accurate FEP assessment.

On the other hand, the inaccuracy of characterisation factors of PPCPs and metals modelled might be another concern, which needs further investigation. The inaccuracy of CFs of PPCPs and metals is from inconsistent input variables (e.g. physicochemical properties, degradation rates, human exposure and ecotoxicity rate of pollutants), which determines the CF value of each substance together with the model itself. Another challenging part for LCA toxicity assessment is that the characterisation factors of each PPCPs provided in the existing models are not fully available. In this situation, researchers have to look for the above-mentioned input data from literature and put these data into toxicity models for CF calculation. In this way, different researchers might get different CF values even with the same model. For different models, CFs of the same PPCPs might be different due to different modelling method and different input parameters for CFs calculation. For example, melting temperature of chemicals/PPCPs is not required in USEtox but required for USES-LCA (CML-IA) which leads to different magnitudes of CF values of each chemical by different models. Furthermore, it has been suggested that CFs should be regionalised to represent the real toxicity impact (Yang et al., 2017). Santos et al. (2018) highlighted that the regionalised CFs with consideration of local specific criteria (e.g. soil properties and climate) could reduce the uncertainty of terrestrial ecotoxicity impact result corresponding to the spatial variability. All above-mentioned uncertainty makes toxicity assessment challenging. Thus, this study aims to evaluate the importance of including metals and PPCPs with current knowledge and models. From this study, it can be seen that considering the large contribution of metals and non-negligible contribution of PPCPs to FEP, both metals and PPCPs should not be ignored in FEP assessment. Further studies in particular on PPCPs removal efficiency in conventional biological treatment plants and model improvement for computing CFs of PPCPs are needed for more accurate toxicity results.

Direct emissions to the soil are the main contributors to TEP. Because only metals in sludge were considered, heavy metals such as nickel, zinc, mercury, copper, and lead contributed 88.2% to TEP. This result is comparable to that reported by Niero et al. (2014) who found that six metals and one pharmaceutical in soil contributed more than 90% to TEP, with mostly copper and zinc. Thus, further treatment such as chemical immobilisation is required to reduce metal concentration in sludge before sludge disposal or land application (Suh & Rousseaux, 2002). As shown in **Figure 14a**, TEP was found to be less significant impact category compared with FEP (97% less than FEP) and HTP for WWTPs with sludge landfilled. It is because sanitary landfills are engineered to minimise the environmental impact on the soil by leachate. If sludge is used in agriculture land, TEP results could be very different because CF values of metals in agricultural land are higher than those to landfills.

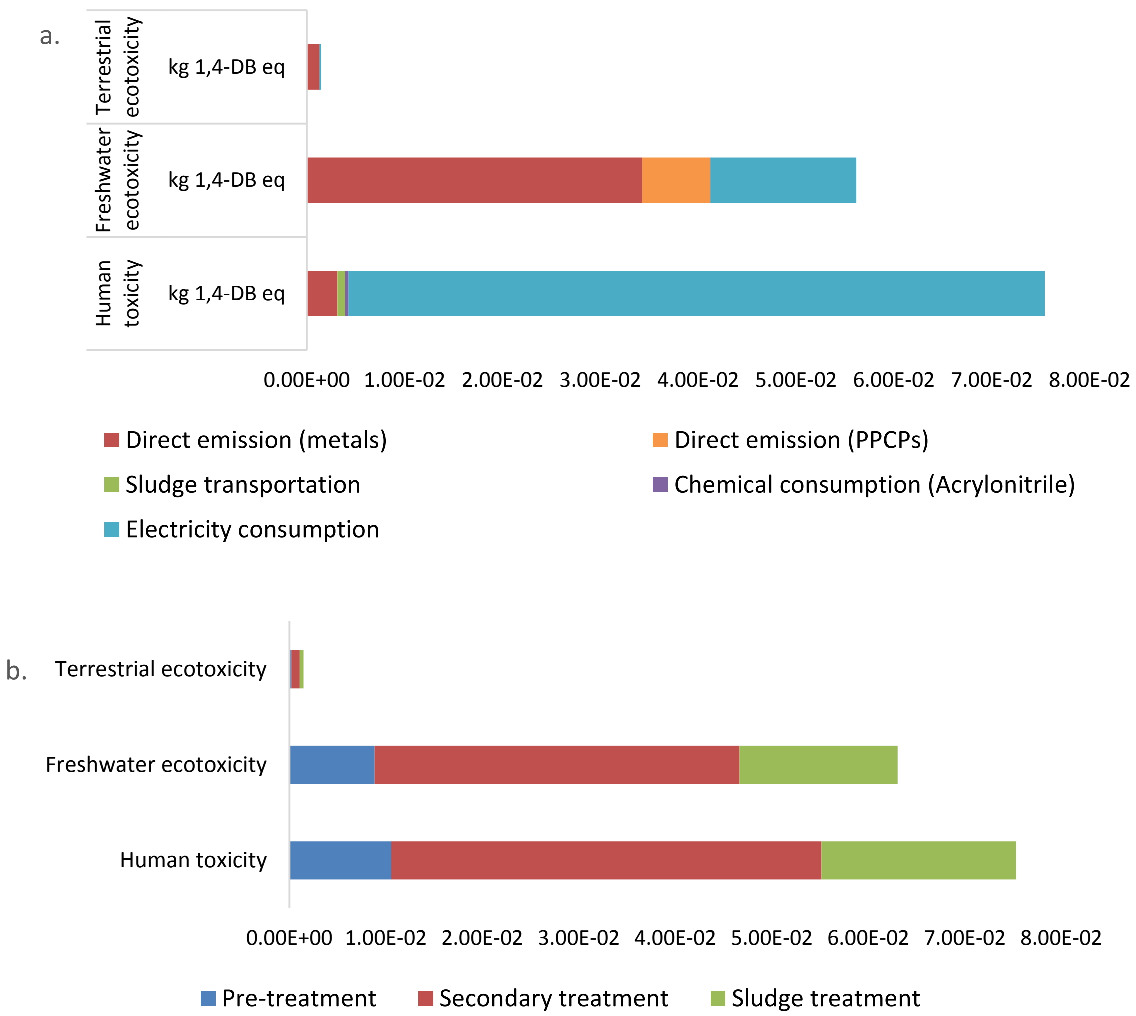


Figure 14. Contributions of different inputs to human toxicity, freshwater ecotoxicity and terrestrial ecotoxicity potentials from the Malaysian STP (FU: 1m^3 of treated wastewater) assessed by CML-IA with: a) contributions from direct and indirect emissions; b) contributions of different processes to each toxicity impact potential.

Table 23. Contribution of dominant metals and PPCPs in effluent of Malaysian STP, metals in excess sludge, and metals released from electricity production to freshwater ecotoxicity potential (FEP)

Source of emission	FEP (%)
Direct emission (metals)	
nickel (effluent)	34.0
zinc (effluent)	14.2
nickel (sludge)	5.7
copper (sludge)	4.2
copper (effluent)	3.3
zinc (sludge)	2.9
cadmium (effluent)	0.2
others	0.8
Sub-total	65.3
Direct emission (PPCPs)	
17β-estradiol (effluent)	7.0
Triclosan (effluent)	2.8
17α-ethinylestradiol (effluent)	0.5
others	0.7
Sub-total	11.0
Indirect emission (electricity)	
barium (water)	22.7
nickel (air)	0.3
beryllium (water)	0.2
others	0.5
Sub-total	23.7
Total	100

4.3.4 Comparison of CML-IA and USEtox methods for toxicity assessment with the inclusion of direct emissions of both metals and PPCPs

Figure 15 shows the comparison of three toxicity impacts including metals and PPCPs assessed by CML-IA and USEtox methods. The results from the USEtox model suggest the dominant impact of direct toxic pollutants on human toxicity while electricity consumption was the main contributor to HTP using the CML-IA method, accounting for 94.4%. With USEtox, direct emissions by metals and PPCPs in effluent and sludge contributed 74.0% and 91.8%, to HTPcancer and HTPnon-cancer, respectively. **Table 24** shows the detailed contributions of different sources in percentages to each toxicity impact by using two LCIA methods, respectively. PPCPs in the effluent of the Malaysian STP contributed less than 0.23% to HTP with any of LCIA methods, indicating a more important role that metals contribute to HTP.

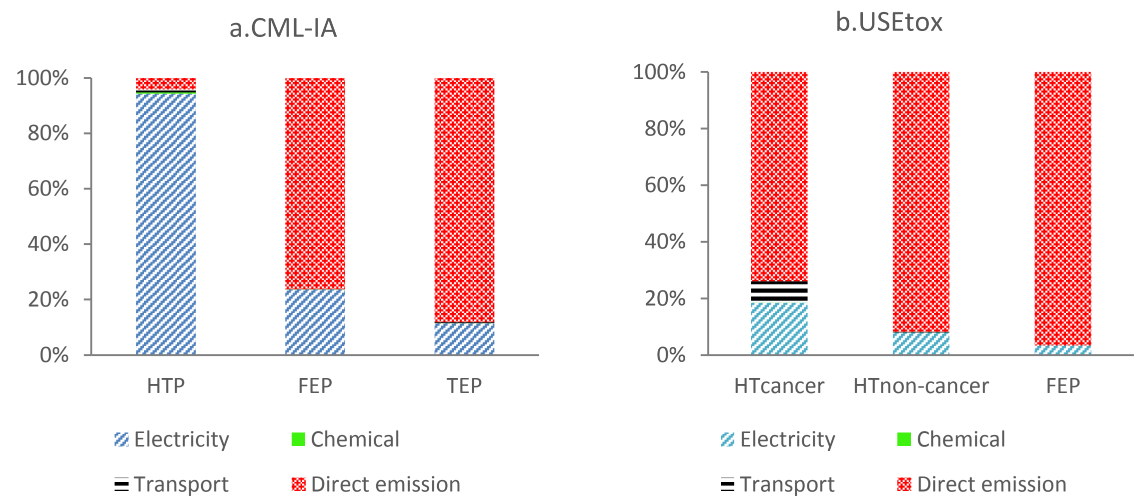


Figure 15. Contribution of metals and PPCPs to each toxicity impact category by using CML-IA and USEtox methods, respectively. Direct emissions include metals and PPCPs in effluent and sludge of Malaysian STP.

Note: Direct emission includes metals and PPCPs in effluent and sludge of MSTP.

Table 24. Contribution of different sources to toxicity potentials assessed by CML-IA and USEtox, respectively

		CML-IA			USEtox		
		HTP	FEP	TEP	HTcancer	HTnon-cancer	FEP
		%	%	%	%	%	%
1	Electricity	94.40	23.50	11.40	18.40	7.90	3.30
2	Chemical	0.40	0.10	0.00	0.00	0.00	0.00
3	Transport	1.00	0.10	0.40	7.60	0.30	0.10
4a	Direct emission (PPCPs)	0.03	10.99	0.00	0.23	0.08	2.47
4b	Direct emission (metals)	4.17	65.26	88.17	73.73	91.72	94.13
	Total	100	100	100	100	100	100

For FEP, direct emissions including metals and PPCPs contributed 76.3% by CML-IA and 96.6%, by USEtox, with a less than 23.7% contribution from electricity consumption. As shown in **Table 24**, although specific percentages from each source to FEP were different by each LCIA method, the general trends by using two LCIA methods were similar, i.e. the largest contribution were from direct emissions of metals followed by electricity and PPCPs. The difference of PPCPs contributions to FEP are mainly due to the differences in CF value between the two models as shown in **Table 15**. Among PPCP substances, 17β-estradiol is the pollutant which contributes most to FEP in both models (**Table 25)**. Although 97.6% of influent 17β-estradiol in Malaysian STP was removed in wastewater treatment process with an effluent concentration as low as 0.001 µg/L, but the high CF value of this substance (representing high toxicity) led to the high contribution of this substance to FEP. From this aspect, LCA analysis is an efficient tool to help identify the toxicity contribution of a specific type of pollutant to guide the setting of WWTP discharge consents. Meanwhile, metals in effluent and sludge contributed 65% by the CML-IA and 94% by USEtox method, respectively, suggesting similar trends by two methods. In addition, sludge disposal is important for LCA in WWTPs in terms of terrestrial ecotoxicity potential (TEP) assessment, but USEtox cannot do TEP assessment. Although USEtox was recommended as a scientific consensus for toxicity assessment in general areas. Thirdly, metals, dissociating and amphiphilic substances are dominant chemicals as CECs in WWTPs, but only interim CFs of these chemicals are provided so far. Thus, for wastewater treatment plants, CML-IA method is more suitable for life cycle toxicity assessment especially for FEP and TEP from the practical perspective compared with USEtox although special attention needs to be paid on human toxicity potential because two methods produce very different human toxicity results. Alfonsin et al. (2014) compared USEtox and CML methods by recalculating 13 new CF values of human toxicity and freshwater ecotoxicity for PPCPs, but the assessment only focused on PPCPs without the inclusion of direct emission of metals in effluent. The specific toxicity impact comparison between different LCIA methods (e.g. CML-IA and USEtox) involving both metals and PPCPs in wastewater was not conducted in any previous research. Thus, results in this study can provide useful information to LCA practitioners for the selection of toxicity models for LCA in WWTPs with focus on toxicity assessment involving micropollutants.

Table 25. Contribution percentages of PPCPs to toxicity potentials by CML-IA and USEtox

		CML-IA (%)			**USEtox (%)**		
	Substances	**HTP**	**FEP**	**TEP**	**HTPcancer**	**HTPnon-cancer**	**FEP**
1	17α-ethinylestradiol	75.7	25.6	0.0	27.8	97.8	1.4
2	17β-estradiol	7.1	72.6	0.0	72.2	0.5	98.4
3	Bisphenol-A	0.4	0.0	0.0	0.0	0.0	0.0
4	Carbamazepine	0.1	0.0	0.0	0.0	0.2	0.0
5	Diclofenac	0.1	0.0	0.0	0.0	0.1	0.2
6	Estrone	14.0	0.2	0.1	0.0	1.4	0.0
7	Ibuprofen	0.4	0.1	17.4	0.0	0.0	0.0
8	Nonylphenol	0.0	0.0	0.0	0.0	0.0	0.0
9	Triclosan	2.2	1.5	82.5	0.0	0.0	0.0
10	Trimethoprim	0.0	0.0	0.0	0.0	0.0	0.0

4.3.5 Comparison of different LCIA methods for toxicity assessment with the inclusion of direct metal emissions

Because heavy metals are the main contributors to toxicity potentials and PPCPs are not measured as commonly as metals, the contributions of heavy metals from four different inputs, namely direct emissions from effluent and sludge, electricity, chemicals, and transportation, were further assessed using five different LCIA methods, i.e. CML-IA, Recipe, IMPACT 2002+, EDIP 2003, and USEtox, to investigate how various LCIA methods affect the impact results by considering direct metal emissions. As shown in **Figure 16a**, human toxicity potential results from direct metal emissions (i.e. effluent and sludge) and indirect emissions (i.e. electricity use) were inconsistent between the five models (including cancer and non-cancer categories by IMPACT 2002+ and USEtox). Impact percentages due to direct metal emissions using CML-IA and Recipe were only 2.1% and 6.5%, respectively, suggesting unimportance of expanding inventory data to direct metal emissions from effluent and sludge by using these two methods. Instead, metals such as barium, nickel, and chromium from electricity generation were the main pollutants, contributing 92.1% and 97.3% to HTP, respectively. However, the other models including IMPACT 2002+, EDIP 2003, and USEtox indicate the necessity of including direct metal emissions from the Malaysian STP such as zinc, cadmium, antimony, and arsenic, with the contribution of 31-92% to HTP. Renou et al. (2007) compared five different LCIA methods namely CML2000, Eco indicator99, EDIP96, EPS, and Ecopoints 97 for five impact category assessment of wastewater treatment including, acidification potential, eutrophication potential, global warming potential and human toxicity potential. They found that all impact category results by different methods were almost similar except for the human toxicity potential result. This highlights that human toxicity assessment in WWTP is the most challenging.

Unlike HTP, FEP impact caused by direct pollutants including metals between models ranged between 48% and 97%. If EDIP was not considered, the difference could be as small as 23%. This is due to more consistent CF values of metals for FEP when discharging to water bodies as shown in **Table 26,** although CFs have different units in different LCIA methods. The dominant pollutants from these methods were almost similar, which were nickel, zinc, cadmium, and copper. Halleux et al. (2006) reported that two methods, CML-IA and IMPACT 2002+ produced almost similar results in terms of freshwater ecotoxicity potential, with nickel and zinc as the main pollutants. Additionally, a study by Lorenzo-Toja et al. (2016) indicated that copper, zinc, and nickel were the main contributors in a Spanish WWTP treating high strength wastewater. The high contribution from direct metal emissions to FEP in all methods indicate that the metals in the effluent of the Malaysian STP have a more significant impact on the water body than indirect emissions, regardless of wastewater strength. Thus, more attention needs to be paid to nickel, zinc, copper and cadmium if sludge will be used in agriculture land or treated water will be reused. Meanwhile, electricity consumption contributes 3-52% to FEP in all methods, indicating that electricity is a moderate contributor to FEP. Similar to HTP, chemicals consumption and transportation in this study had a negligible impact on FEP. It is noted that CFs from different LCIA methods for all metals are inconsistent. This means the difference of toxicity impact were also caused by different CFs of pollutant in each method. For example, CF of nickel (3.24E+03) is 2 order of magnitude higher than mercury

(1.72E+03) in CML-IA method. While, CF of nickel (9.84E+01) is one order of magnitude higher than mercury (9.32E+01) in Recipe. By contrast, CF of mercury is 12 times higher than that of nickel in IMPACT2002+ (**Table 26**). This highlights that CFs from these three methods is not consistent and varies in the rankings of metals although the overall contribution is quite similar. Overall, except EDIP, this study found that CML-IA, Recipe, IMPACT2002+, and USEtox produce consistent FEP results, thus, any of them could be selected for future LCA studies when including direct metal emissions in the inventory for FEP assessment.

As shown in **Figure 16c**, three models namely CML-IA, Recipe, and IMPACT 2002+ demonstrated less than <13.5% difference. The main contributors to TEP were copper, zinc, nickel, and cadmium. By contrast, the EDIP model in this study showed that 81% of TEP was from electricity consumption, followed by 17% from chemical consumption. Considering that FEP and TEP results by EDIP have a very obvious difference compared with other methods in this study, EDIP is not recommended for toxicity study in WWTPs. In overall, this study shows that CML-IA, Recipe, and IMPACT 2002+ produce consistent results of HTP, FEP, and TEP when including direct metal emissions, whereas EDIP and USEtox provide different results. This model comparison provides useful information and guideline for future LCA practice on the selection of LCIA methods according to specific aims.

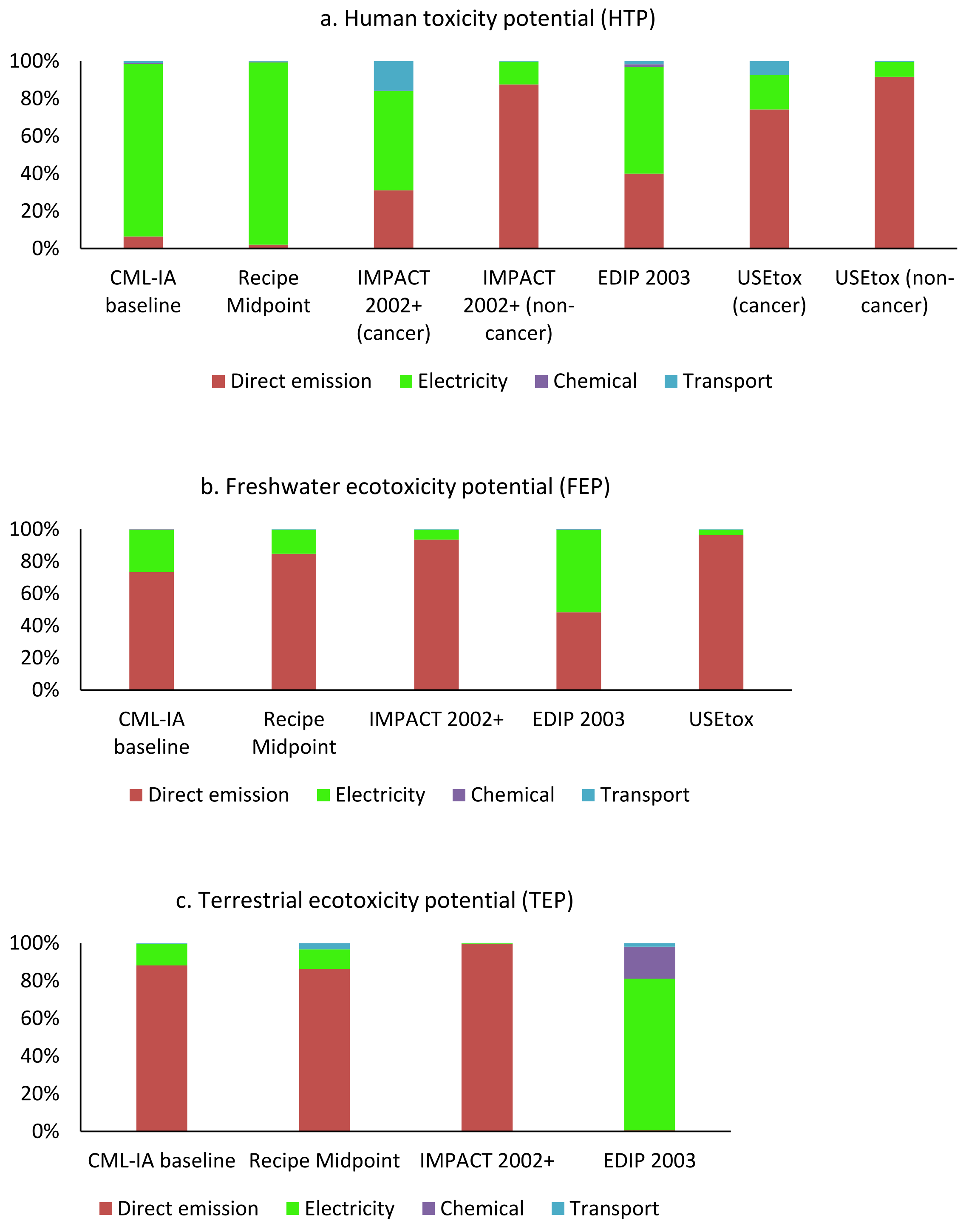


Figure 16. The comparison of metal contributions to each toxicity category from four different sources in Malaysian STP, i.e., direct emissions from effluent and sludge, electricity consumed for wastewater treatment, chemical consumed in the plant and transportation for sludge disposal, between different LCIA toxicity methods

Table 26. Characterisation factors (CFs) for 9 metals by CML-IA, Recipe, IMPACT 2002+, EDIP 2003 and USEtox. Emission compartment: freshwater (water)

		CML-IA	Recipe	IMPACT2002+	EDIP 2003	USEtox
No	Substances	kg 1,4-DB eq	kg 1,4-DB eq	kg TEG water	m^3	CTUe
1	Antimony, Sb	1.97E+01	1.54E+01	2.10E+06	1.72E+06	1.22E+03
2	Arsenic, As	2.07E+02	1.56E+01	3.88E+05	1.75E+06	2.78E+04
3	Cadmium, Cd	1.52E+03	9.05E+01	2.92E+06	1.07E+08	9.71E+03
4	Chromium VI, Cr VI	2.77E+01	9.02E-01	4.53E+05	6.07E+05	1.05E+05
5	Copper, Cu	1.16E+03	1.18E+02	2.06E+07	1.14E+07	5.52E+04
6	Lead, Pb	9.62E+01	4.14E-01	2.63E+05	1.82E+06	375E+00
7	Mercury, Hg	1.72E+03	9.32E+01	1.58E+07	3.64E+06	2.21E+04
8	Nickel, Ni	3.24E+03	9.84E+01	1.27E+06	6.07E+05	1.49E+04
9	Zinc, Zn	9.17E+01	7.52E+00	1.40E+06	9.10E+05	3.86E+04

Reference: CML-IA (http://cml.leiden.edu/software/data-cmlia.html); Recipe (www.lcia-ReCiPe.net); IMPACT2002+ (http://www.impactmodeling.org); EDIP2003 (http://www.lca-center.dk/cms/site.aspx?p=4441); USEtox (www.usetox.org)

4.3.6 Sensitivity analysis

Toxicity impact assessment results in Malaysian STP highlight that metals in sludge, metals and PPCPs in influent/effluent, and electricity consumption are the key factors that affect toxicity impact results. Table 5 shows how toxicity and ecotoxicity impacts from Malaysian STP were affected by changing ±10% of main inventory values from scenario 3, by using CML-IA method. The result shows that HTP, FEP and TEP change with ±9.89%, ±3.95% and ±2.75% respectively from ±10% variation in electricity consumption. According to Piao et al. (2016), electricity consumption is the most sensitive factors to human toxicity potential. Metals in wastewater (i.e. nickel, zinc and copper) mainly affecting FEP result with nickel is the most affected pollutants at ±4.40%, while only affecting HTP with <±1.0%. PPCPs in wastewater only affecting FEP result with the highest is from 17β-estradiol at ±1.22%. Finally, metals in sludge show more effects on TEP by nickel and zinc, and there is negligible change to HTP and FEP result. In general, 10% variation in concentration of metals in sludge, concentration of metals and PPCPs in wastewater, and electricity consumption do not cause major environmental impact change.

Table 27. Sensitivity analysis results to human toxicity and ecotoxicity impacts by changing each inventory component by ±10% according to 1 m^3 of treated wastewater by CML-IA method

		Impact categories		
		Human toxicity potential (HTP) (%)	Freshwater ecotoxicity potential (FEP) (%)	Terrestrial ecotoxicity potential (TEP) (%)
	Inventory components (±10%)			
1	Electricity consumption	±9.89	±3.95	±2.75
	Metals in wastewater:			
2	Nickel	±0.28	±4.40	±1.65
3	Zinc	±0.15	±2.32	±0.84
4	Copper	±0.02	±1.26	±0.03
	PPCPs in wastewater:			
5	17β-estradiol	±0.01	±1.22	±0.00
6	Triclosan	±0.00	±0.90	±0.00
7	17α-ethinylestradiol	±0.00	±0.14	±0.00
	Metals in sludge:			
8	Nickel	±0.29	±1.10	±4.43
9	Zinc	±0.08	±0.95	±3.91
10	Copper	±0.04	±1.34	±1.20

4.4 Conclusion

To understand the toxicity impacts of site-specific pollutants such as metals and PPCPs in the effluent and sludge of WWTPs, sampling work and detailed analysis were conducted to identify their occurrence and contribution to the environment. LCA was used to investigate the contribution of these direct pollutant emissions to toxicity and their comparison with other sources such as electricity, transportation and chemical consumption to guide future life cycle toxicity studies. The main findings from the assessment in this study are summarised below.

- Unlike general pollutants in wastewater such as COD, BOD and nutrients, concentrations of PPCPs and metals in wastewater and their removal efficiencies in treatment processes in developing and developed countries with high and low strength wastewater showed no any clear patterns. Thus, sampling work is recommended for the acquisition of PPCPs and metals data to ensure accuracy and reliability of toxicity assessment.
- The inclusion of PPCPs and heavy metals from effluent and sludge caused a 76% increase in FEP and an 88% increase in TEP, respectively, highlighting the importance of their direct emissions to the environment. Therefore, PPCPs and heavy metals should not be omitted from toxicity studies particularly on freshwater and terrestrial ecotoxicity.
- The contribution of PPCPs in the effluent to FEP by the CML-IA method is only 11%, which is mainly from 17β-estradiol, triclosan, and 17α-ethinylestradiol, while there is negligible contribution from other PPCPs. Thus, the inclusion of prioritised PPCPs is far more important than a large number of PPCP types for freshwater ecotoxicity studies. In addition, the contribution from direct metal emissions to FEP is 65.3%, indicating metals play more negative role to FEP than PPCPs.
- Direct emissions of heavy metals from the effluent and sludge, and indirect emissions by electricity generation from fossil fuels such as coal and oil, were the main contributors to the three toxicity impact categories, i.e. HTP, FEP and TFP. Thus, the reduction in toxicity relies on the shift in energy sources from fossil fuels to renewable energy, the reduction in energy consumption for wastewater treatment, and the removal of key heavy metals from wastewater and sludge.
- For ecotoxicity assessment in WWTPs with the inclusion of direct metal and PPCP emissions, CML-IA and USEtox methods can get similar results.
- When including direct metal emissions, CML-IA, Recipe, and IMPACT 2002+ produced consistent results of human toxicity, freshwater ecotoxicity, and terrestrial ecotoxicity, whereas EDIP and USEtox

provided different results. CML-IA, Recipe, and IMPACT 2002+ are thus recommended for toxicity assessment in WWTPs.

- In the sensitivity analysis, HTP, FEP and TEP change with ±9.89%, ±3.95% and ±2.75% respectively from ±10% variation in electricity consumption. Meanwhile, the ±10%, variation in concentration of top 3 metals in sludge, and top 3 metals and PPCPs in wastewater only affecting less than ±5% to human toxicity and ecotoxicity impacts.

This work identifies the importance of considering local toxic pollutants such as metals and PPCPs in effluent and sludge for toxicity assessment in WWTPs and sampling for data reliability. LCIA methods were recommended for ecotoxicity assessment.

5 Upgrading a large and centralised municipal wastewater treatment plant with sequencing batch reactor technology for integrated nutrient removal and phosphorus recovery: environmental and economic life cycle performance

5.1 Introduction

With more concerns on eutrophication in natural water bodies, wastewater treatment plants (WWTPs) built for removals of only organic matter and suspended solids cannot meet the demand of environmental protection, particularly in nutrient-sensitive areas. Upgrading wastewater treatment plants for nutrient removal has been imperative in many areas. Nutrient removal, however, requires larger reactor volume, more energy and chemical consumption, and produces probably more chemically enriched sludge for disposal (Meneses et al., 2015). This might lead to a transfer of environmental impact caused by the nutrient in water to other environmental compartments such as air and soil. A holistic environmental assessment is, therefore, necessary for a selection of available technologies for upgrading and an evaluation of the overall environmental impact of the upgraded plants. Meanwhile, an economic assessment can provide information about affordability and price for environmental benefits (Garcia & Pargament., 2015). This is extremely important in developing countries with limited resource allocation for environment protection.

Eutrophication in Malaysia has reached a point where it cannot be ignored. According to Huang et al. (2015), 72% of rivers and lakes in Malaysia were in serious eutrophic conditions. But almost all existing WWTPs in Malaysia were not designed for nutrient removal. Therefore, upgrading WWTPs for nutrient removal in Malaysia has appeared inevitable in the future, just like what developed countries and some developing countries such as China have been doing. Malaysia is located in a tropical region with highly diluted municipal wastewater (Rashid & Liu, 2020) and many WWTPs, especially in large cities being operated with large capacity such as 500,000 population equivalent. Besides, a sequencing batch reactor (SBR) technology is widely adopted. With these features together, upgrading WWTPs and the environmental benefits and burdens it thus brings could be different from those in other regions. It should be pointed out that the selection of upgrading technology for nutrient removal should be based on the existing technology used for wastewater treatment to ensure the feasibility of upgraded technology and effective integration with existing facilities. Most of the previous research on technology selection via techno-economic and environmental assessment did not consider local factors such as wastewater characteristics, which might cause infeasibility of conclusions to a specific region. Furthermore, environmental assessment of WWTPs in developing countries is significantly less than developed countries as reported by Gallego-Schmid & Tarpani, 2019 and Zang et al., 2015, let alone environmental impact from the upgrading of WWTPs. Therefore, this study investigated the upgrading of a large and centralised WWTP with SBR technology and the selection of available technologies for nutrient removal and recovery from environmental and economic perspectives.

Various technologies with great potential to reduce chemical and energy consumption for nitrogen and phosphorus removal from municipal wastewater have been reported such as anammox, denitrifying phosphorus removal (DPR), and reverse/forward osmosis membrane filtration, but almost all of these are still at research stage without application (Third et al., 2005; Haiming Zou et al., 2014; Hube et al., 2020), Anammox is a novel/cost-effective way to reduce nitrogen in ammonium-rich wastewater (Hauck et al., 2016;), but not applicable to diluted municipal wastewater. DPR can remove phosphorus and nitrite/nitrate simultaneously with limited chemical oxygen demand (COD) and reduced aeration, but it demands complicated control without practical application so far (Jin et al., 2017). Some variants of existing mature technologies such as integrated fixed-film activated sludge process (Waqas et al., 2020), and sequencing batch membrane photobioreactor seem more promising for rapid full-scale application (Lau et al., 2019; Sheng et al., 2017). In practice, nitrogen removal from municipal wastewater still relies on biological nitrification and denitrification while phosphorus removal depends on either enhanced biological phosphorus removal (EBPR) or chemical precipitation by aluminium, ferric or calcium salts. EBPR is more environmentally friendly without or with little chemical consumption, but it has relatively lower phosphorus removal efficiency and less performance stability compared with chemical precipitation. Conventional chemical phosphorus removal demands chemicals to precipitate phosphate in wastewater, producing more chemically enriched sludge for disposal. Both technologies are widely used in practice. It is thus not surprising that to upgrade WWTPs for nutrient removal, nitrification/denitrification for nitrogen removal, and EBPR or chemical precipitation (Maurer & Boller, 1999) for phosphorus removal have to be adopted. More recently, a new technology called aerobic granular sludge technology has been reported to have good nutrient removal efficiency due to the co-existence of aerobic, anoxic and anaerobic zones in granules (Piotr & Cydzik-kwiatkowska, 2018; Liu et al., 2010). A full-scale aerobic granular sludge process for sewage treatment has demonstrated that total nitrogen and phosphorus concentrations can be reduced to 7 mg/L and 1 mg/L, respectively, without chemical dose in normal conditions (Pronk et al., 2015). More importantly, aerobic granule technology is based on SBR operation, which enables retrofitting of existing WWTPs with SBR technology to granular sludge SBR relatively easy by changing operational conditions. More than 60 successful full-scale application of aerobic granular sludge-SBR technology allows itself to be one of the feasible options to upgrade especially SBR based plants in Malaysia.

With the concern of phosphorus depletion within the next 50 to 100 years (Cordell et al., 2009), wastewater has been considered as one of the important phosphorus sinks for phosphorus recovery (Egle et al., 2016). Sweden, for example, has required that 60% of phosphorus in municipal wastewater needs to be recovered for phosphorus security (Hultman et al., 2004). Apart from the direct application of stabilised sewage sludge to land for nutrient recovery, some technologies have been developed to recover phosphorus from sludge to make phosphorus products such as slow-releasing fertiliser struvite (Corre et al., 2009) or calcium phosphate (Woods et al., 1999) to supplement rock phosphate. Most successfully commercialised phosphorus recovery technologies are Ostara from Canada, Gifhorn and Airprex from Germany, and Unitika from Japan. Phosphorus recovery cannot only recover phosphorus resource but also alleviate pipe and pump clogging problems caused by uncontrolled struvite crystallisation and deposition in the sludge digestion and downstream treatment processes (Urdalen, 2013). Thus, phosphorus recovery could be an option to WWTPs which need upgrading. Although upgrading WWTPs seems still too costly in developing countries, it would be beneficial to see how much environmental and economic benefits or burdens it could bring when upgrading WWTPs for nutrient removal and recovery.

Currently, environmental assessment using life cycle assessment is believed as a useful analytical tool to develop a metric with which to compare, and evaluate processes and products with regards to their potential environmental effects from the cradle to the grave (Hauschild et al., 2013). Thus it could be used effectively to guide technology and process selection. The economic cost, another important factor to consider for WWTP upgrading, has been increasingly conducted using life cycle costing assessment to select wastewater treatment solutions or processes (Rawal & Duggal, 2016; Hernandez-Sancho et al., 2010). Both environmental impact assessment and economic analysis can provide comprehensive information as guidelines to decision-makers for upgrading WWTPs from both financial and environmental perspectives.

This study thus aims to design upgrading processes based on an existing Malaysian centralised wastewater treatment plant with SBR technology for nutrient removal and resource recovery, and to assess economic burdens and environmental benefits or burdens of upgraded processes with life cycle assessment. All selected technologies for upgrading are commercially successful in ensuring the practical feasibility of upgraded processes. Phosphorus recovery as a possible option in the future was also considered to investigate net environmental benefit. The ultimate goals of this study include the development of general guidelines for upgrading WWTPs for nutrient removal or phosphorus recovery and the provision of comprehensive information to decision makers for upgrading.

5.2 Materials and method

5.2.1 The selection and description of case study

A large and centralised municipal Malaysian sewage treatment plant (STP) in Penang, Malaysia, was selected for upgrading to remove nutrients and recover phosphorus to improve the local environmental status and phosphorus security. Malaysian STP treated an average flow rate of 148,950 m^3/d domestic wastewater from a separate sewer system to serve 662,002 population equivalent in 2017. The existing Malaysian STP mainly consists of grit and grease screening as primary treatment, 4 SBRs for a combined primary sludge settling and chemical oxygen demand (COD) removal, gravity belt thickener for sludge thickening, anaerobic digester for sludge volume reduction, and biosolids dewatering for final sludge landfilling (**Figure 17**). Three high-strength streams produced from sludge pre-holding tank, sludge thickening tank and centrifuge decanter for dewatering are returned to SBRs for treatment. The treated water is discharged into the river nearby, while the sludge produced is sent to a landfill located 47 km away. This type of SBR based WWTPs is widely used in Malaysia and is considered as a typical wastewater treatment plant. The infrastructure of Malaysian STP was built in 2007 and is expected to have a 40 to 50-year lifetime as suggested by (Ruhland et al., 2006).

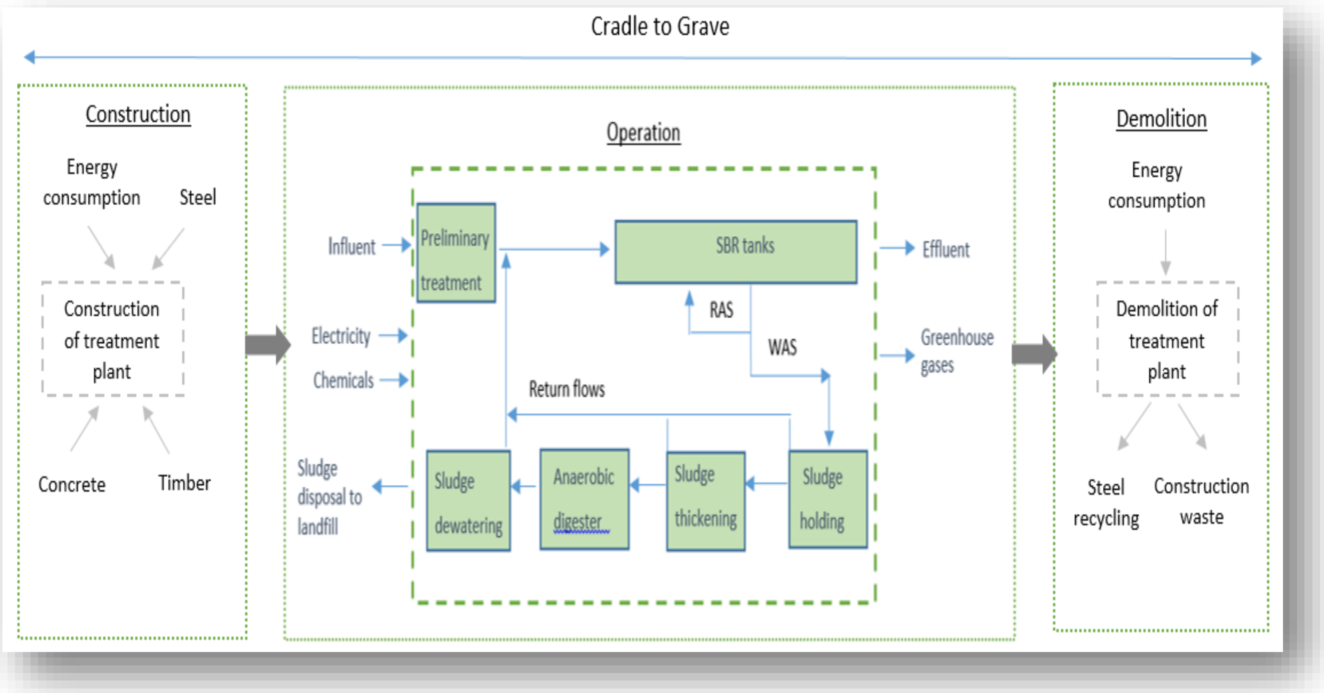


Figure 17. Schematic diagram of the existing Malaysian STP in the system boundary of this study with 40-year operation.

Note: (SBR = sequencing batch reactor; RAS = return activated sludge, WAS = waste activated sludge)

5.2.2 Sampling and analytical method

The data of process, operation, and quality of influent and effluent in Malaysian STP were provided by the plant manager. To supplement any necessary data for this study especially on the mass balance calculation, additional samples at 4 different points, i.e. the influent to the treatment plant, the immediate inlet to SBR after mixing with side stream from sludge treatment units, the inlet to the sludge treatment units and the effluent to the environment, were taken and analysed in August 2017. Glass bottles were used to collect and store the samples from all four sampling points. All samples were labelled and kept cold inside iceboxes at 4 ^{o}C during collection, and then transported to the laboratories for analytical determination by the analytical team from the National University of Malaysia (UKM). Total biochemical oxygen demand ($TBOD_5$), total chemical oxygen demand (TCOD), total suspended solids (TSS), total nitrogen (TN) and total phosphorus (TP) were analysed following standard methods by American Public Health Association (APHA). While nitrate and sulfate were measured using HACH method (i.e. HACH 8171) (**Table 28**). The data from these 4 sampling points can be used not only to validate the methods used for mass balance in the existing STP, but also to conduct mass balance in the three upgraded processes.

Table 28. Lists of pollutants from wastewater streams and relevant analytical methods used

No	Substances	Method	Unit	Location of sampling
1	Total biochemical oxygen demand, $TBOD_5$	Standard methods-5210 B electrode method ysi meter	mg/L	Influent, inlet to the aeration tank, effluent and inlet to sludge treatment.
2	Total chemical oxygen demand, TCOD	Hach-method 8000-Reactor Digestion method	mg/L	
3	Total suspended solids, TSS	Standard methods – 2540D	mg/L	
4	Total nitrogen, TN	Kjehdahl's method	mg/L	
5	Total phosphorus, TP	Microwave digestion method HPR-EN-11 (ICP-MS)	mg/L	
6	Nitrate	HACH method 8171 – Cadmium reduction method	mg/L	
7	Sulfate	HACH method 8171 – Cadmium reduction method	mg/L	

5.2.3 Design of the three upgraded processes

The existing wastewater treatment process is shown in **Figure 17**. SBRs play dual roles for both primary sludge settling and biological COD oxidation. Apart from 1-hr filling, 1-hr settling and 1-hr decanting, only 1-hr aeration is used to oxidise COD, making the total cycle time as 4 hours. 4 SBRs with each reactor working volume of 6206 m^3 are being operated alternatingly to deal with 148,950 m^3 municipal wastewater per day continuously. The calculation of the number of reactors required in 3 upgraded processes are attached in **Appendix C**. The characteristics of municipal wastewater to this plant are shown in **Table 29.** Based on local wastewater characteristics and SBR technology adopted in the existing Malaysian STP, three new processes denoted as Process A, Process B and Process C, respectively, were designed by adopting commercially available technologies to upgrade the existing plant for nutrient removal and phosphorus recovery. One important criterion for the upgrading design is to minimise the retrofitting requirement and meanwhile to make the best use of the existing facilities to maximise the integration. Processes A and B adopted nitrification and denitrification for nitrogen (N) removal with extended cycle time of SBRs. Process A relied on enhanced biological phosphorus (P) removal (EBPR), while Process B used ferric precipitation to remove phosphorus. Process C adopted aerobic granular sludge technology for simultaneous biological N and P removal.

Commercial technologies, i.e. AirPrex and Gifhorn, were chosen for phosphorus recovery with AirPrex for P recovery in Process A and Gifhorn in Processes B and C. It is assumed that 22% and 40% of TP with respect to sludge input was recovered by Airprex (Kabbe, 2015) and Gifhorn (Egle et al., 2016), respectively. Airprex forms struvite by stripping out CO_2 and adds $MgCl_2$, and installed between the anaerobic digester and dewatering equipment. The process converts orthophosphate into struvite crystals which are harvested from the bottom of the reactor, i.e. sand washer (Niewersch & Stemann, 2014; P-Rex Factsheet, 2015). In Gifhorn, phosphorus bound in the biomass is extracted from the solid phase of digested sewage sludge by the addition of sulfuric acid (H_2SO_4). In a second step, the dissolved heavy metals are precipitated as sulfides (dosing of Na_2S) by adjusting pH with NaOH, to minimise the co-precipitation of heavy metals with fertiliser products in the subsequent step for phosphate precipitation. After solid/liquid separation with a decanter, dosing of $Mg(OH)_2$ initiates precipitation of phosphorus as a mix of struvite/calcium phosphate (adjusted with NaOH). The P product is harvested by a second solid/liquid separation (P-Rex Factsheet, 2015).

Table 29. Water quality parameters of the existing process of MSTP

Parameters	Unit	Influent	Inlet to aeration tank	Effluent	Inlet to sludge treatment
Population equivalent	PE	662,002.0			
Total suspended solids (TSS)	mg/L	174.0 ±10.5	150.0 ±10.4	14.6 ±5.8	5980.0 ±330.0
Total biochemical oxygen demand ($TBOD_5$)	mg/L	126.0 ±11.0	66.4 ±10.5	10.1 ±1.6	765.0 ±21.2
Total chemical oxygen demand (TCOD)	mg/L	433.0 ±21.3	304.0 ±19.5	44.7 ±3.1	5109.0 ±250.5
Total nitrogen (TN)	mg/L	28.0 ±2.2	27.0 ±2.1	15.0 ±1.5	210.0 ±11.3
Total phosphorus (TP)	mg/L	2.6 ±0.07	2.5 ±0.04	1.1 ±0.03	18.0 ±1.60
COD:N:P ratio	-	167:11:1	122:11:1	41:14:1	284:12:1

5.2.4 Mass and energy balances

The mass balance of the existing and the three upgraded processes was calculated based on flowrate, TCOD, TN, TP, and suspended and volatile suspended solids (SS and VSS) to generate balancing inventory data in water and sludge streams, respectively. The plant-wide mass balance started from influent to WWTP and ended with effluent to rivers, and sludge to landfill. An iterative procedure was developed in an excel spreadsheet for the main interrelated parameters to carry out the mass balance calculation. The first iteration from the initial flow rate determined return flow rates, which affected the flow rate of the stream into SBRs. From here, the second iteration started. Iteration was stopped until the incremental changes in flow quantities of carbon and nutrients in return flows were less than 5%. The validity of the iterative procedure developed was verified with the sampled data from the existing process.

The calculation of energy balance for all upgraded processes was carried out based on the energy consumption in the existing process and the additional energy consumption for nutrient removal, phosphorus recovery and electricity recovery (**Table 37**). The electricity consumption for blowers and P recovery is shown in **Table 37**.

Electricity consumption for P recovery process was calculated based on the average total electricity demand suggested by P-Rex Factsheet, 2015. It is assumed that Airprex and Gifhorn require 10.3 kWh/1 kg P recovered and 6.9 kWh/1 kg P recovered, respectively (P-Rex Factsheet, 2015). Electricity production from the sludge anaerobic digestion and CHP were calculated in the three upgraded processes with the assumption of 40% electricity recovery efficiency from CHP.

5.2.5 Environmental assessment

The environmental assessment was performed by a life cycle assessment (LCA) using SimaPro v9.0. International standards and recommendations by ISO, 2006 were followed.

5.2.5.1 Goal and scope

The goal was to carry out a comparative assessment of LCA to evaluate the environmental benefits/burdens of the three newly upgraded processes, which can be used as options for the upgrading of WWTP to remove nutrients and recover phosphorus. 'Cradle to grave' analysis was adopted which began with the construction and ended at the demolition stage. In the operation stage, wastewater flowrate, pollution loads, transportation of chemicals and sludge, energy consumption and chemical consumption were considered. In the construction stage, materials (e.g. steel, concrete and timber) and energy used for the construction of all operation units were considered. While in the demolition of operation units and buildings, steel recycled and energy used were considered (Hao et al., 2019). Also, the avoided products such as struvite and electricity recovered from the upgraded processes of Malaysian STP were included, but the impact from struvite application as fertiliser was not considered. The illustrated system boundary for this LCA - WWTP study is shown in **Figure 17.**

5.2.5.2 Functional units

1 m^3 of treated wastewater was used as functional unit 1 (FU1) to compare the environmental impacts between the upgraded processes and the existing process. FU1 was widely adopted for LCA in WWTPs with/without nutrient removal and recovery (Piao et al., 2015; Lorenzo-Toja et al., 2016; Hauck et al., 2016; Coats et al., 2011; Niero et al., 2014). The results based on FU1 could thus be easily compared with LCA studies from the literature. Using per m^3 of treated wastewater as a functional unit, however, is believed not to be able to well reflect wastewater treatment performance especially for nutrient removal and recovery (Pradel et al., 2016). The primary functions to achieve in the upgraded processes in this study are to remove nutrients and to recover phosphate as fertiliser (struvite) from sludge. Therefore, the functional unit defined as 1 kg of struvite recovered ($NH_4MgPO_4.6H_2O$) was used as FU2 as well (Amann et al., 2018; Pradel & Aissani., 2019) to

assess the environmental efficiency of per kg struvite recovered between the three upgraded processes. In this way, we could estimate how much environmental benefits/burdens were generated for per kg struvite recovered. Functional unit 3 (FU3) defined as 1 kgPO_4^{3}-eq removed was used for comparison with the result from functional unit 1 and functional unit 2. FU3 was also used by (Rodriguez-Garcia et al., 2011) and (Comas Matas, 2012). The mass load of eutrophying substances removal, i.e. chemical oxygen demand (COD), total nitrogen (TN), and total phosphorus (TP) in wastewater were converted to kgPO_4^{3}-eq using the characterisation factor from eutrophication potential impact category as defined in the CML-IA baseline v3.04 methodology.

5.2.5.3 Life cycle inventory (LCI)

The operation data provided by the plant managers and the data from sampling in August 2017 in Malaysian STP were used as the basic inventory data. Specifically, the life cycle inventory (LCI) consists of the following parameters:

1) The indirect inputs of resources including electricity consumption for aeration, pumping, stirring; transportation for sludge disposal; and chemical consumption for wastewater treatment and sludge treatment. Background data/indirect emissions were obtained from the Ecoinvent v3.3 database as described below:

   a. Electricity production in Malaysia was selected from the Ecoinvent v3.3 database.
   b. Chemical production: Data on the production of chemicals (e.g. methanol, iron chloride, magnesium chloride, sulphuric acid, sodium sulfide, sodium hydroxide, magnesium hydroxide, and polymers), were selected from the European life cycle database (ELCD) and Ecoinvent v3.3 database. For polyelectrolyte for sludge dewatering, a similar production process for acrylonitrile was taken from the Ecoinvent v3.3 as proposed by Rodriguez-Garcia et al., 2011.
   c. Lorries with a capacity of 3.5-7.5 metric ton were selected as transport vehicles for the disposal of sludge produced from Malaysian STP, as well as for the chemical transportation to the site.
   d. Inputs for construction: Data of the resources such as steel, timber and concrete were selected from the Ecoinvent v3.3 database.

2) The direct pollutants emissions in influent and effluent including TCOD, TN and TP.

3) The direct emissions of gases such as carbon dioxide (CO_2), methane (CH_4) and dinitrogen monoxide (N_2O) from the operation of the plant as outputs. Direct N_2O was mainly generated from biological nitrogen removal process and CH_4 was from anaerobic wastewater and/or sludge treatment (Masuda et al., 2015); gas emissions were calculated according to the Intergovernmental Panel on Climate Change guidelines (IPCC, 2006) based on the 100-year time horizon.

4) The construction inputs such as steel, timber, concrete and energy consumption; and the demolition inputs after lifespan such as energy consumption. Data for the construction and demolition process were calculated by using the method provided by Hao et al., 2019.

5) The avoided products including electricity and struvite recovered in the operation phase, and steel recovered in the demolition phase. All inventory data are provided in **Table 30** for FU1**, Table 31** for FU2 and **Table 32** for FU3.

Table 30. Life cycle inventories of the existing and three upgraded processes. Values are presented based on per m^3 of treated wastewater as functional unit 1 (FU 1)

Parameters	**Unit**	**Existing**	**Process A[a]**	**Process B[b]**	**Process C[c]**
Transport for;					
1.Solid waste disposal	t.km/m^3	1.81E-05	1.81E-05	1.81E-05	1.81E-05
2.Sludge disposal	t.km/m^3	6.46E-03	4.78E-03	5.04E-03	4.81E-03
3.Chemicals	t.km/m^3	2.58E-05	5.15E-05	7.73E-05	6.44E-05
Sub-total	t.km/m^3	*6.50E-03*	*4.85E-03*	*5.13E-03*	*4.89E-03*
Chemical consumption;					
4.Polyelectrolyte	kg/m^3	5.15E-04	5.15E-04	5.15E-04	5.15E-04
For nutrients removal;					
5.Methanol (CH_3OH)	kg/m^3	-	2.40E-02	2.62E-02	-
6.Iron chloride III ($FeCL_3$)	kg/m^3	-	-	1.53E-02	-
For Phosphorus recovery;					
7.Magnesium chloride ($MgCl_2$)	kg/m^3	-	8.77E-03	-	-
8.98% sulfuric acid (H_2SO_4)	kg/m^3	-	-	7.99E-03	8.47E-03
9.Sodium sulfide (Na_2S)	kg/m^3	-	-	7.79E-04	8.26E-04
10.Sodium hydroxide $Na(OH)_2$	kg/m^3	-	-	2.82E-03	2.99E-03
11.Magnesium hydroxide $Mg(OH)_2$	kg/m^3	-	-	1.95E-04	2.07E-04
Electricity input;					
12.Electricity consumption	kWh/m^3	2.05E-01	2.69E-01	2.75E-01	2.80E-01
Avoided products;					
13.Electricity generated	kWh/m^3	0.00E+00	7.00E-02	7.64E-02	7.24E-02
14.Struvite ($MgNH_4PO_4$)	kg/m^3	-	4.78E-03	7.70E-03	8.16E-03
15.Phosphate fertilizer	kg/m^3	-	6.05E-04	9.74E-04	1.03E-03
Emission to air;					
16.Carbon dioxide (CO_2) biogenic	kg/m^3	8.93E-02	8.55E-02	8.50E-02	8.62E-02
17.Methane (CH_4)	kg/m^3	1.10E-03	6.82E-04	6.93E-04	7.04E-04
18.Dinitrogen monoxide (N_2O)	kg/m^3	4.60E-04	5.70E-04	5.75E-04	5.80E-04
Effluent to rivers					
19.Chemical oxygen demand (COD)	kg/m^3	4.47E-02	4.47E-02	4.47E-02	4.47E-02
20.Total nitrogen (TN)	kg/m^3	1.50E-02	5.40E-03	4.40E-03	3.15E-03
21.Total phosphorus (TP)	kg/m^3	1.05E-03	2.40E-04	1.90E-04	6.00E-05
Materials for construction					
22.Steel	kg/ m^3	2.13E-03	2.35E-03	2.45E-03	2.45E-03
23.Concrete	m^3/ m^3	1.79E-05	1.97E-05	2.06E-05	2.06E-05
24.Timber	kg/ m^3	1.20E-06	1.32E-06	1.38E-06	1.38E-06
25.Energy consumption	kWh/ m^3	4.69E-03	5.16E-03	5.39E-03	5.39E-03
Demolition					
26.Energy consumption	kWh/ m^3	3.79E-03	4.17E-03	4.36E-03	4.36E-03
27.Construction waste	kg/ m^3	3.45E-02	3.79E-02	3.96E-02	3.96E-02
28.Steel recycling	kg/ m^3	2.18E-03	2.40E-03	2.50E-03	2.50E-03

[a] Nutrient removal by EBPR, nitrification and denitrification (AOA) and P recovery by Airprex technology;
[b] Nutrient removal by ferric precipitation, post anoxic denitrification, and P recovery by Gifhorn technology;

[c] Nutrient removal by AGS and P recovery by Gifhorn technology;
(EBPR= enhanced biological phosphorus removal; AOA = anaerobic, aerobic anoxic; AGS = aerobic granular sludge)

Table 31. Life cycle inventories of the three upgraded processes. Values are presented based on per kg of struvite recovered from wastewater as functional unit 2 (FU2)

Parameters	Unit	Process A	Process B	Process C
<u>*Transport for;*</u>				
1.Solid waste	t.km/kg struvite	3.79E-03	2.35E-03	2.22E-03
2.Sludge	t.km/kg struvite	1.00E+00	6.54E-01	5.89E-01
3.Chemicals	t.km/kg struvite	1.08E-02	1.00E-02	7.89E-03
Sub-total	t.km/kg struvite	**1.01E+00**	**6.67E-01**	**6.00E-01**
<u>*Chemicals consumption;*</u>				
4.Polyelectrolyte	kg/kg struvite	1.08E-01	6.69E-02	6.31E-02
For nutrients removal;				
5.Methanol (CH_3OH)	kg/kg struvite	5.01E+00	3.41E+00	-
6.Iron chloride III ($FeCL_3$)	kg/kg struvite	-	1.99E+00	-
For Phosphorus recovery:				
7.Magnesium chloride ($MgCl_2$)	kg/kg struvite	1.83E+00	-	-
8.98% sulfuric acid (H_2SO_4)	kg/kg struvite	-	1.04E+00	1.04E+00
9. Sodium sulfide (Na_2S)	kg/kg struvite	-	1.01E-01	1.01E-01
10. Sodium hydroxide ($Na(OH)_2$)	kg/kg struvite	-	3.67E-01	3.67E-01
11.Magnesium hydroxide ($Mg(OH)_2$)	kg/kg struvite	-	2.53E-02	2.53E-02
<u>*Electricity input;*</u>				
12.Electricity consumption	kWh/kg struvite	5.63E+01	3.57E+01	3.43E+01
<u>*Avoided products;*</u>				
13.Electricity generated	kWh/kg struvite	1.46E+01	9.93E+00	8.87E+00
14.Struvite ($MgNH_4PO_4$)	kg/kg struvite	1.00E+00	1.00E+00	1.00E+00
15.Phosphate fertilizer	kg/kg struvite	1.27E-01	1.27E-01	1.27E-01
<u>*Emission to air;*</u>				
16.Carbon dioxide (CO_2-biogenic)	kg/kg struvite	1.24E-01	1.20E-01	1.35E-01
17.Methane (CH_4)	kg/kg struvite	2.30E-01	1.43E-01	1.35E-01
18.Dinitrogen monoxide (N_2O)	kg/kg struvite	3.46E-02	1.73E-02	1.24E-02
<u>Effluent to river</u>				
19.Chemical oxygen demand (COD)	kg/kg struvite	9.35E+00	5.81E+00	5.48E+00
20.Total nitrogen (TN)	kg/kg struvite	1.13E+00	5.72E-01	3.86E-01
21.Total phosphorus (TP)	kg/kg struvite	5.02E-02	2.47E-02	7.35E-03
<u>*Materials for construction;*</u>				
22.Steel	kg/kg struvite	4.91E-01	3.19E-01	3.00E-01
23.Concrete	m^3/kg struvite	4.13E-03	2.68E-03	2.53E-03
24.Timber	kg/kg struvite	2.75E-04	1.79E-04	1.69E-04
25.Energy consumption	kWh/kg struvite	1.08E+00	7.01E-01	6.61E-01
<u>*Demolition;*</u>				
26.Energy consumption	kWh/kg struvite	8.72E-01	5.66E-01	5.34E-01
27.Construction waste	kg/kg struvite	7.93E+00	5.15E+00	4.86E+00
28.Steel recycling	kg/kg struvite	5.01E-01	3.25E-01	3.07E-01

[a] Nutrient removal by EBPR, nitrification and denitrification (AOA) and P recovery by Airprex technology;
[b] Nutrient removal by ferric precipitation, post anoxic denitrification, and P recovery by Gifhorn technology;
[c] Nutrient removal by AGS and P recovery by Gifhorn technology;
(EBPR= enhanced biological phosphorus removal; AOA = anaerobic, aerobic anoxic; AGS = aerobic granular sludge)

Table 32. Life cycle inventories of the three upgraded processes. Values are presented based on per kg of eutrophication reduction as functional unit 3 (FU3)

Parameters	Unit	Process A	Process B	Process C
Transport for:				
1.Solid waste	t.km/kg PO_4^{3-}eq	7.17E-04	7.02E-04	6.78E-04
2.Sludge	t.km/kg PO_4^{3-}eq	1.89E-01	1.95E-01	1.80E-01
3.Chemicals	t.km/kg PO_4^{3-}eq	2.04E-03	2.99E-03	2.41E-03
Sub-total	t.km/kg PO_4^{3-}eq	**1.92E-01**	**1.99E-01**	**1.83E-01**
Chemicals consumption:				
4.Polyelectrolyte	kg/kg PO_4^{3-}eq	2.04E-02	2.00E-02	1.93E-02
For nutrients removal;				
5.Methanol (CH_3OH)	kg/kg PO_4^{3-}eq	9.48E-01	1.02E+00	-
6.Iron chloride III ($FeCL_3$)	kg/kg PO_4^{3-}eq	-	5.93E-01	-
For phosphorus recovery:				
7.Magnesium chloride ($MgCl_2$)	kg/kg PO_4^{3-}eq	3.47E-01	-	-
8.98% Sulfuric acid (H_2SO_4)	kg/kg PO_4^{3-}eq	-	3.09E-01	3.16E-01
9. Sodium sulfide (Na_2S)	kg/kg PO_4^{3-}eq	-	3.02E-02	3.09E-02
10. Sodium hydroxide ($Na(OH)_2$)	kg/kg PO_4^{3-}eq	-	1.09E-01	1.12E-01
11. Magnesium hydroxide ($Mg(OH)_2$)	kg/kg PO_4^{3-}eq	-	7.54E-03	7.73E-03
Electricity input:				
12.Electricity consumption	kWh/kg PO_4^{3-}eq	1.06E+01	1.06E+01	1.04E+01
Avoided products:				
13.Electricity generated	kWh/kg PO_4^{3-}eq	2.77E+00	2.96E+00	2.71E+00
14.Struvite ($MgNH_4PO_4$)	kg/kg PO_4^{3-}eq	1.89E-01	2.98E-01	3.05E-01
15.Phosphate fertilizer	kg/kg PO_4^{3-}eq	2.39E-02	3.77E-02	3.86E-02
Emission to air:				
16.Carbon dioxide (CO_2)-biogenic	kg/kg PO_4^{3-}eq	1.24E-01	1.20E-01	1.35E-01
17.Methane (CH_4)	kg/kg PO_4^{3-}eq	2.30E-01	1.43E-01	1.35E-01
18.Dinitrogen monoxide (N_2O)	kg/kg PO_4^{3-}eq	3.46E-02	1.73E-02	1.24E-02
Effluent to river:				
19.Chemical oxygen demand (COD)	kg/kg PO_4^{3-}eq	3.38E+00	3.29E+00	3.22E+00
20.Total nitrogen (TN)	kg/kg PO_4^{3-}eq	4.35E-02	4.26E-02	4.11E-02
21.Total phosphorus (TP)	kg/kg PO_4^{3-}eq	6.55E-03	5.17E-03	3.78E-03
Materials for construction:				
22.Steel	kg/kg PO_4^{3-}eq	9.28E-02	9.50E-02	9.16E-02
23.Concrete	m^3/kg PO_4^{3-}eq	7.80E-04	7.99E-04	7.71E-04
24.Timber	kg/kg PO_4^{3-}eq	5.21E-05	5.33E-05	5.14E-05
25.Energy consumption	kWh/kg PO_4^{3-}eq	2.04E-01	2.09E-01	2.02E-01
Demolition:				
26.Energy consumption	kWh/kg PO_4^{3-}eq	1.65E-01	1.69E-01	1.63E-01
27.Construction waste	kg/kg PO_4^{3-}eq	1.50E+00	1.54E+00	1.48E+00
28.Steel recycling	kg/kg PO_4^{3-}eq	9.48E-02	9.70E-02	9.36E-02

[a] Nutrient removal by EBPR & nitrification and denitrification (AOA), and P recovery by Airprex technology;
[b] Nutrient removal by ferric precipitation & post anoxic denitrification, and P recovery by Gifhorn technology;
[c] Nutrient removal by AGS, and P recovery by Gifhorn technology;
(EBPR= enhanced biological phosphorus removal; AOA = anaerobic:aerobic:anoxic; AGS = aerobic granular sludge)

5.2.5.4 Life cycle impact assessment (LCIA) and interpretation

Life cycle impact assessment (LCIA) was conducted with the characterisation factors from CML-IA (baseline v3.04) methodology (Mbaya et al., 2017; Ruhland et al., 2006) to compare the environmental footprints of the existing and the three upgraded processes. As wastewater treatment plants mainly generate climate change-related impacts and environmental quality issues (Renou et al., 2007), six midpoint characterisation impact categories such as eutrophication potential (EP), freshwater ecotoxicity potential (FEP), human toxicity potential (HTP), global warming potential (GWP), abiotic depletion (fossil fuel) potential (ADFP) and acidification potential (AP) were chosen as the main assessment categories. Since concentrations of heavy metals in sludge were not available in the three new processes, terrestrial ecotoxicity impact was not included in the assessment. The LCA results were finally interpreted to assess the contribution of each component in the inventory of each environmental impact category. Besides, the normalisation factors from World 2000 in CML-IA method were used for the normalisation of the environmental impact categories at the midpoints based on per person per year. .

Upgraded Malaysian STP is expected to have a 40-year lifetime. In the next 40 years of operation of Malaysian STP, there will be variation in the mass load of pollutants which could affect the electricity consumption, chemicals consumption and nutrient concentration in the effluent. Sensitivity analysis was thus conducted to evaluate how the variations in inventory data such as electricity consumption, nutrient concentrations in effluent and chemical consumption affect LCA impact category results with 20 and 40 years of design life. ±10% variation of inventory data for 20 years was selected to measure the variability of environmental impact results in half-life of the upgraded STP. While ±20% variation of inventory data for 40 years was selected to measure higher variability of environmental impact results in the whole lifetime of the upgraded STP. In this way, the effects of the accuracy of inventory data of wastewater treatment plant with a long design life were evaluated. FU1, i.e. per m^3 treated wastewater was selected for this analysis to facilitate the comparison with the results from other studies. However, the effect from construction and demolition were not included in the sensitivity analysis because their inventory data are the same throughout the whole lifetime of upgraded Malaysian STP.

5.2.6 Hotspot analysis of electricity consumption

Energy consumption was used to identify hotspots because it is the main contributor during the operation of WWTP to many environmental impact categories (i.e. GWP, ADFP, HTP and AP). Hotspot analysis was conducted to check how much that nutrient removal and phosphorus recovery in the existing and upgraded processes could contribute to the total electricity consumption. Power consumption data from each electric device in water and sludge line of the existing plant (i.e. pumping, bar screen, aeration and mechanical dewatering) was provided by the plant. The data of energy required in the newly designed processes such as for struvite recovery and the nutrient removal in SBRs were obtained from the energy balance.

5.2.7 Economic assessment

The economic cost of different processes during construction and operation periods was assessed. Life cycle cost (LCC) based on per population equivalent (PE) per day was calculated by **Equation 5.2.7-1** according to Awad et al., 2019. Per PE was used as a functional unit in LCC assessment as WWTPs are designed, constructed and operated based on PE. The prices of materials (i.e. steel, concrete or timber), transport, disposal fee and electricity were obtained from the current Malaysian market (2017-2019). For the construction cost, additional items for the upgrading processes were considered, i.e. new reactor cost, CHP generator, extra blower, Airprex reactor and Gifhorn reactors. The operation cost for P recovery process was assumed as 9.0 USD/1kg P recovered for Airprex, and 17.8 USD/1 kg P recovered for Gifhorn (Egle et al., 2016). Prices for chemicals were referred to the literature values by Lorenzo-Toja et al., 2016a; Awad et al., 2019; Bertanza et al., 2014. 1 kWh of electricity in Malaysia costs 0.15 USD (United States dollar). Labour cost varies over time and it also depends on the plant location (Awad et al., 2019). Since the comparative assessment in this study is for the existing process and the three upgraded processes at the same plant during the same operation period, labour cost was not considered in this study. To get the net life cycle costs, the revenues from the recovered products such as electricity and struvite from the operation were deducted.

$$LCC\ (USD / PE\cdot day) = CC + OC + TC - S \qquad \textbf{Equation 5.2.7-1}$$

Where:

LCC = Total life cycle cost

CC = Construction cost

OC = Operation cost (i.e. electricity consumption, chemicals and landfill disposal fee)

TC = Transport cost (for sludge and chemicals)

S = Revenue from the recovered products such as electricity and phosphorus

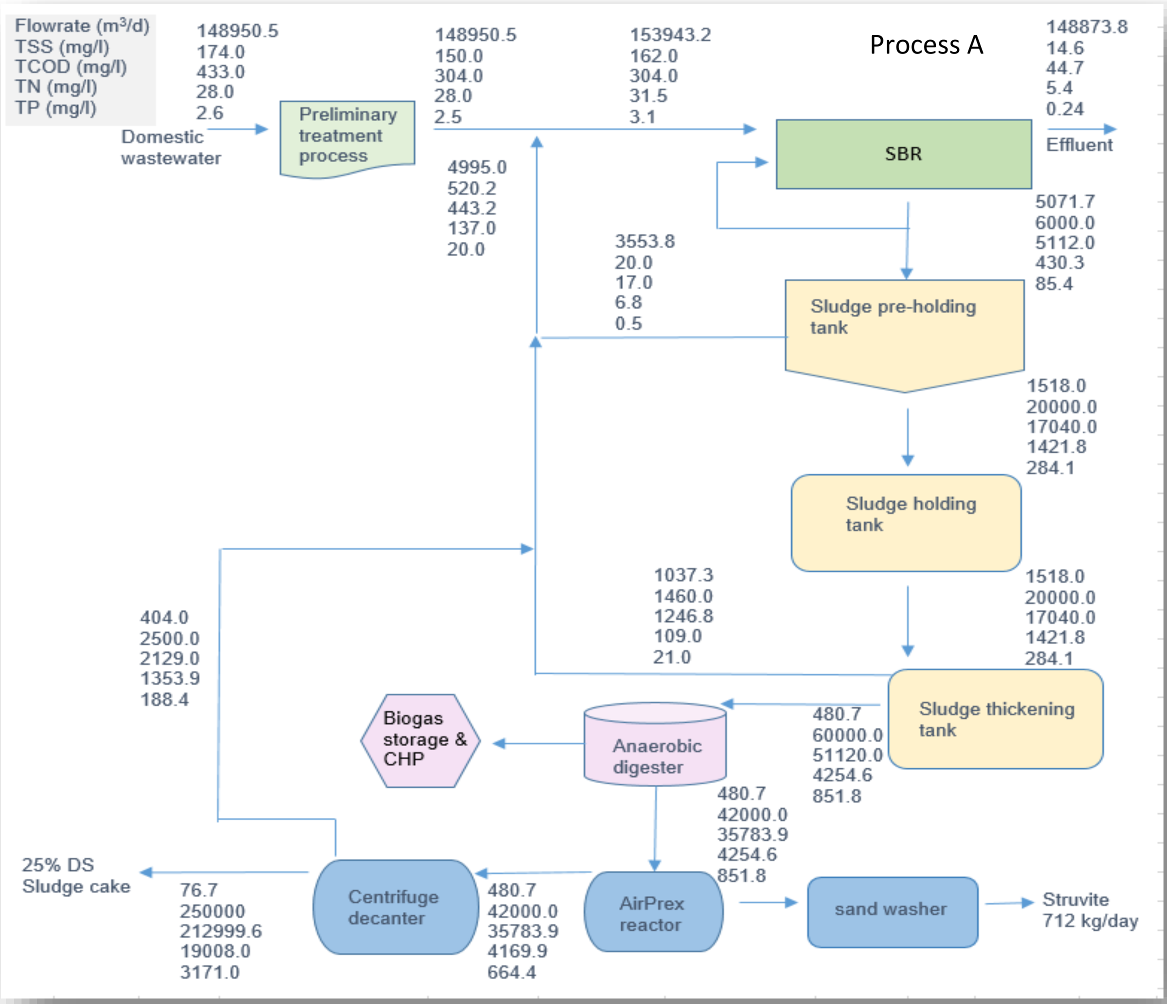
5.3 Results and discussion

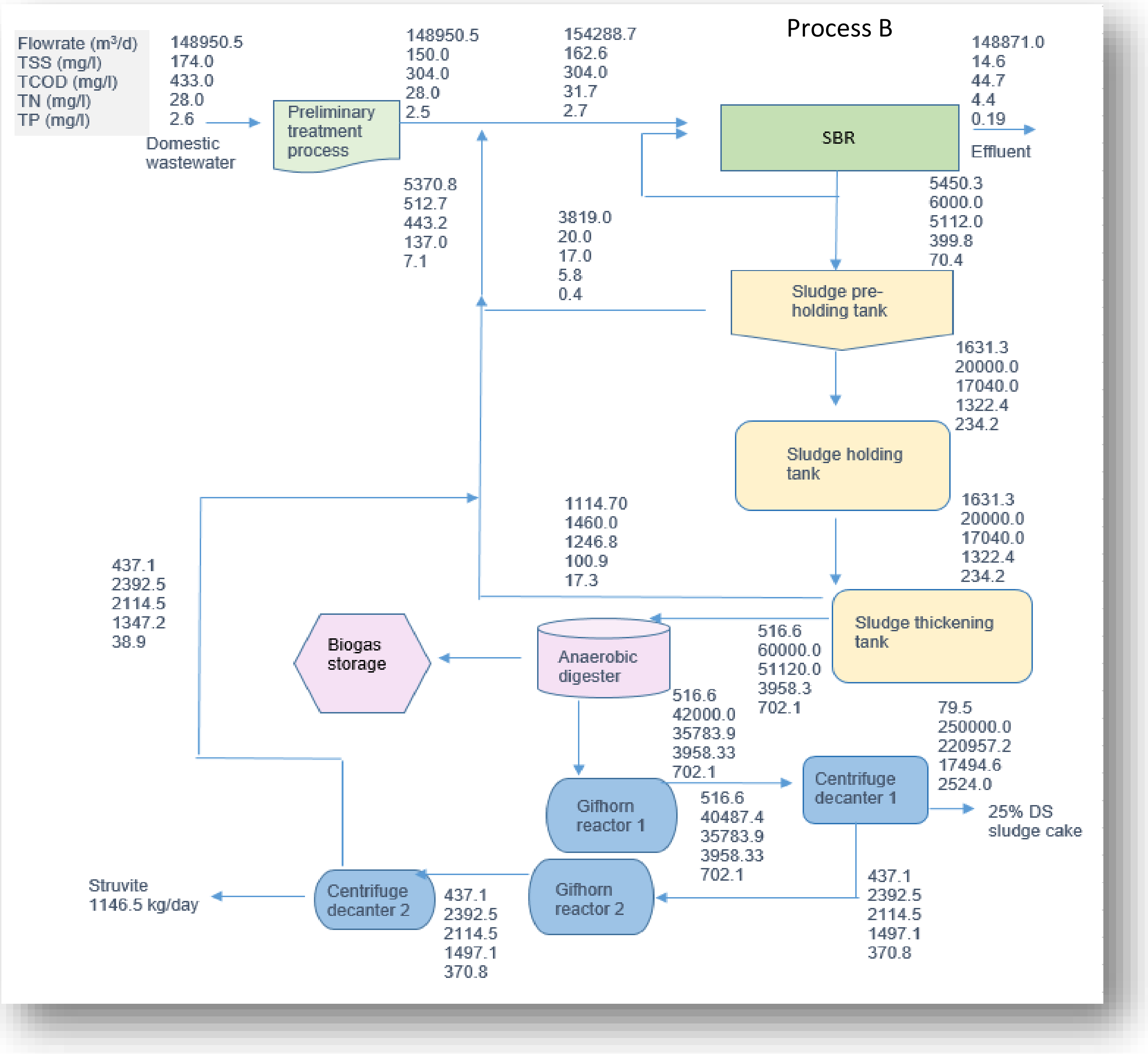
5.3.1 Design of three upgraded processes for nutrient removal and phosphorus recovery

5.3.1.1 Design of three upgraded processes for nitrogen and phosphorus removal

To achieve nitrification, denitrification and EBPR, the cycle time of SBR was extended to 6 hours in upgraded Process A to accommodate anaerobic/aerobic/anoxic (AOA) phases with a ratio of anaerobic: aerobic: anoxic as 1:2:1 (Liu et al., 2013). Soejima et al. (2008) showed that with an insufficient carbon source, nitrogen removal rate in AOA-SBR system was only 34%. Therefore, external carbon sources such as methanol were suggested to be dosed in the anoxic period to improve total nitrogen removal efficiency. Due to the extension of cycle time, the plant's treating capacity was reduced, and 2 more SBRs with the same reactor volume were needed to deal with the same treating capacity of the plant after upgrading. Meanwhile, extra aeration is needed for nitrification on the top of COD oxidation.

Chemical precipitation is widely used in WWTPs for phosphorus removal, thus, in the upgraded Process B, biological nitrogen removal and ferric precipitation were adopted. A typical phase ratio of aerobic to anoxic as 2:1 was selected (Liu et al., 2013). The aeration phase was extended for nitrification. To make the continuous operation of SBRs in the upgraded Process B easier, the cycle time was kept at 6 hours with additional 2 SBRs to deal with the same plant treating capacity. Similar to Process A, methanol was dosed in the anoxic period for denitrification. Aerobic granular sludge technology was adopted in the upgraded Process C with the aeration phase extended from 1 hour to 2 hours for simultaneous nitrogen and phosphorus removal, resulting in 5-hr total cycle time. Thus 1 more SBR in the upgraded Process C was added. The cyclic operation of SBRs in the three upgraded processes with nutrient removal is shown in **Figure 19**. Parameters of SBRs in the three upgraded processes and the existing process are shown in **Table 33**.


Flowrate (m3/d)
TSS (mg/l)
TCOD (mg/l)
TN (mg/l)
TP (mg/l)
Process A
148950.5 174.0 433.0 28.0 2.6
Domestic wastewater
Preliminary treatment process
148950.5 150.0 304.0 28.0 2.5
153943.2 162.0 304.0 31.5 3.1
SBR
148873.8 14.6 44.7 5.4 0.24
Effluent
4995.0 520.2 443.2 137.0 20.0
3553.8 20.0 17.0 6.8 0.5
5071.7 6000.0 5112.0 430.3 85.4
Sludge pre-holding tank
1518.0 20000.0 17040.0 1421.8 284.1
Sludge holding tank
1518.0 20000.0 17040.0 1421.8 284.1
1037.3 1460.0 1246.8 109.0 21.0
404.0 2500.0 2129.0 1353.9 188.4
Biogas storage & CHP
Anaerobic digester
480.7 60000.0 51120.0 4254.6 851.8
Sludge thickening tank
480.7 42000.0 35783.9 4254.6 851.8
25% DS Sludge cake
76.7 250000 212999.6 19008.0 3171.0
Centrifuge decanter
480.7 42000.0 35783.9 4169.9 664.4
AirPrex reactor
sand washer
Struvite 712 kg/day



Process B
Flowrate (m³/d)
TSS (mg/l)
TCOD (mg/l)
TN (mg/l)
TP (mg/l)
148950.5
174.0
433.0
28.0
2.6
Domestic wastewater
Preliminary treatment process
148950.5
150.0
304.0
28.0
2.5
154288.7
162.6
304.0
31.7
2.7
SBR
148871.0
14.6
44.7
4.4
0.19
Effluent
5370.8
512.7
443.2
137.0
7.1
3819.0
20.0
17.0
5.8
0.4
5450.3
6000.0
5112.0
399.8
70.4
Sludge pre-holding tank
1631.3
20000.0
17040.0
1322.4
234.2
Sludge holding tank
1631.3
20000.0
17040.0
1322.4
234.2
1114.70
1460.0
1246.8
100.9
17.3
437.1
2392.5
2114.5
1347.2
38.9
Sludge thickening tank
Biogas storage
Anaerobic digester
516.6
60000.0
51120.0
3958.3
702.1
516.6
42000.0
35783.9
3958.33
702.1
79.5
250000.0
220957.2
17494.6
2524.0
Gifhorn reactor 1
Centrifuge decanter 1
516.6
40487.4
35783.9
3958.33
702.1
25% DS sludge cake
437.1
2392.5
2114.5
1497.1
370.8
Struvite 1146.5 kg/day
Centrifuge decanter 2
437.1
2392.5
2114.5
1497.1
370.8
Gifhorn reactor 2

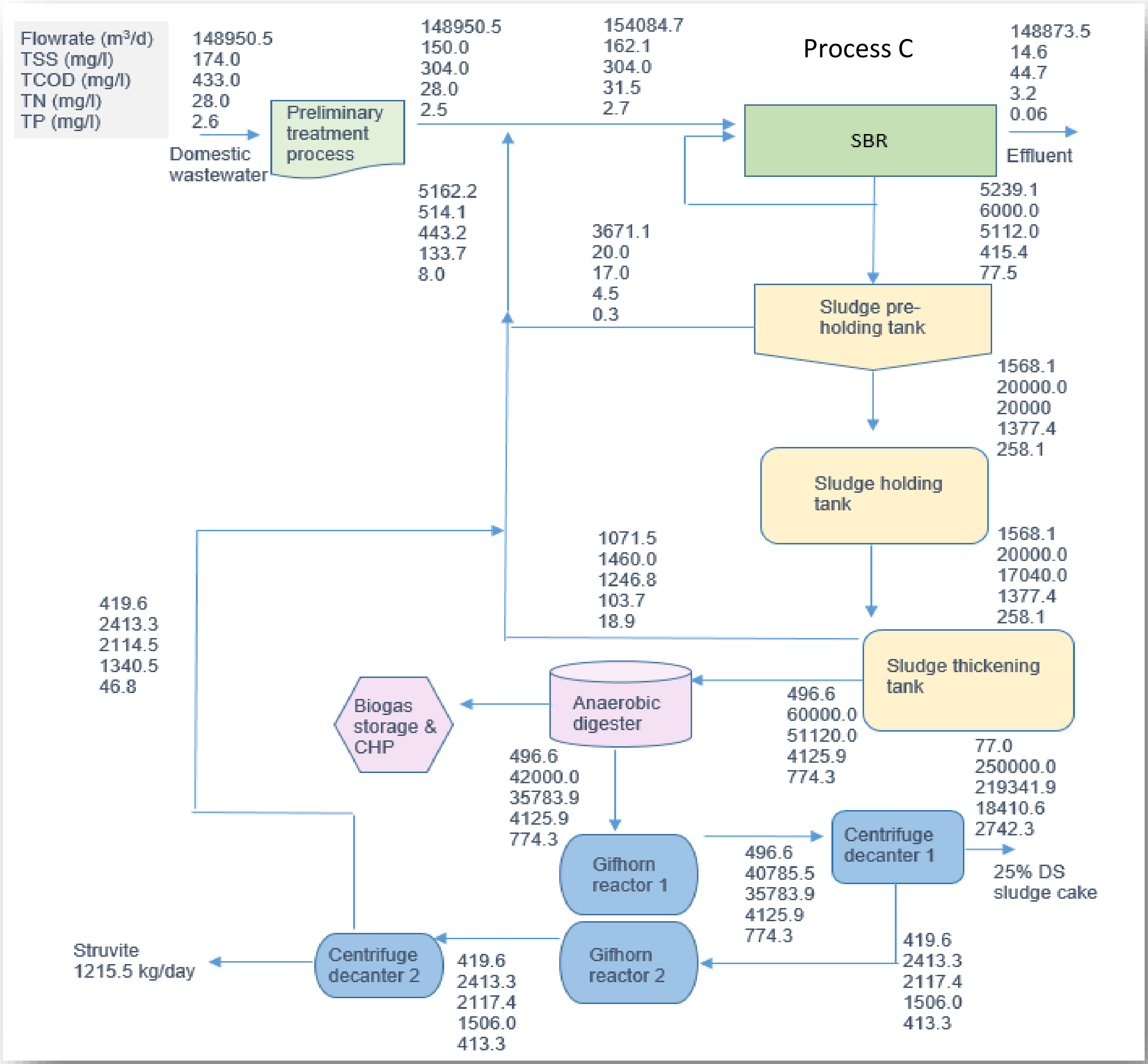


Figure 18. Mass balance for the three upgraded processes

Note: Process A is based on enhanced biological phosphorus removal (EBPR) for phosphorus (P) removal, nitrification and denitrification for nitrogen removal and AirPrex for P recovery. Process B is to use ferric precipitation to removal phosphorus, nitrification and denitrification to remove nitrogen, and Gifhorn to recover P from sludge. Process C is to adopt aerobic granular sludge (AGS) technology to do simultaneous nitrogen and phosphorus removal, and Gifhorn for P recovery.

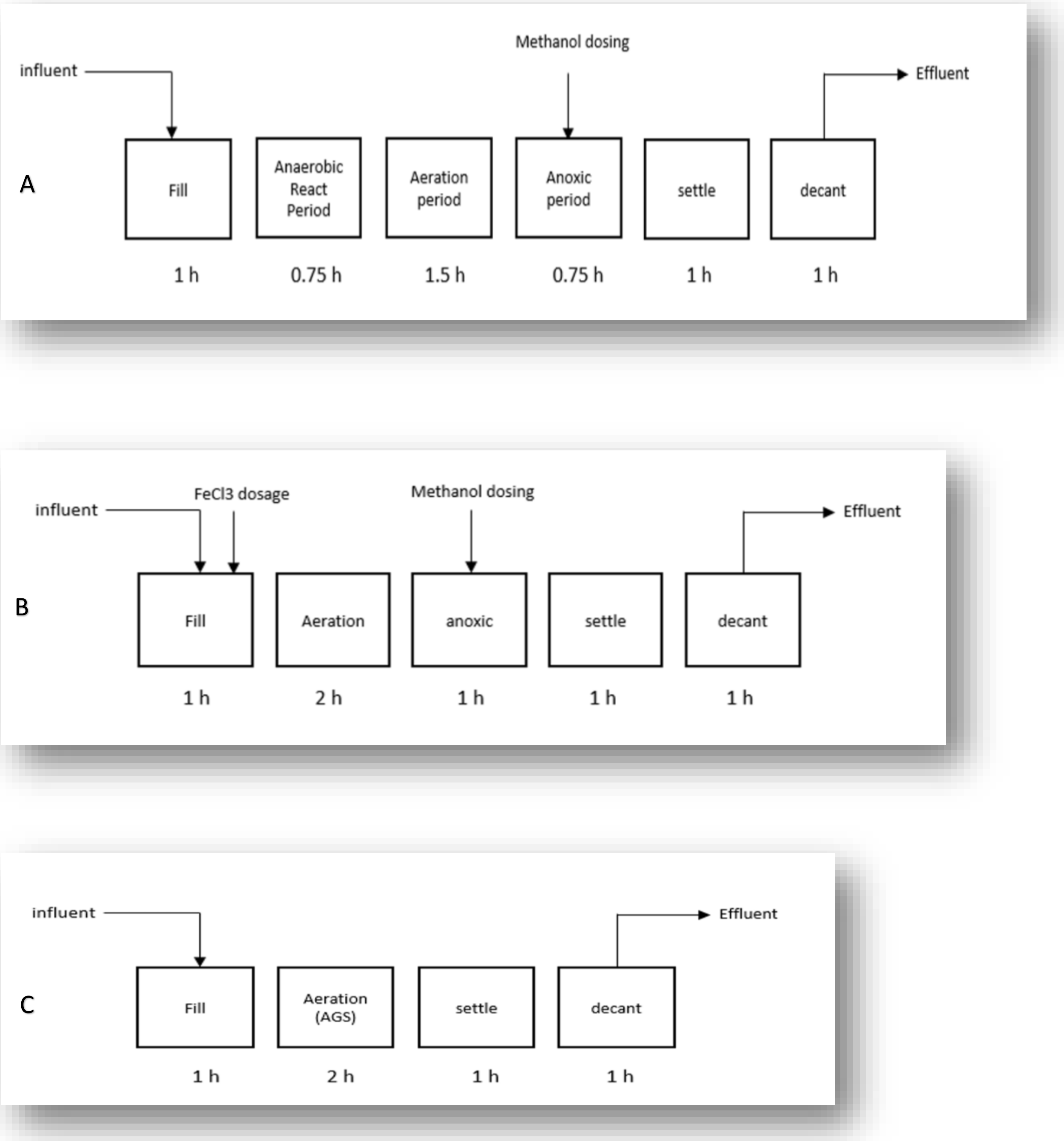


Figure 19. Cyclic operation design for SBRs operation in the three upgraded processes with nutrient removal.

Note: A) Cyclic operation of SBRs in process A with EBPR and nitrification-denitrification (AOA); B) Cyclic operation of SBRs in process B with phosphorus precipitation by ferric and nitrification-denitrification (post-anoxic) and; C) Cyclic operation of SBRs in process C with AGS technology for simultaneous carbon, N and P removal

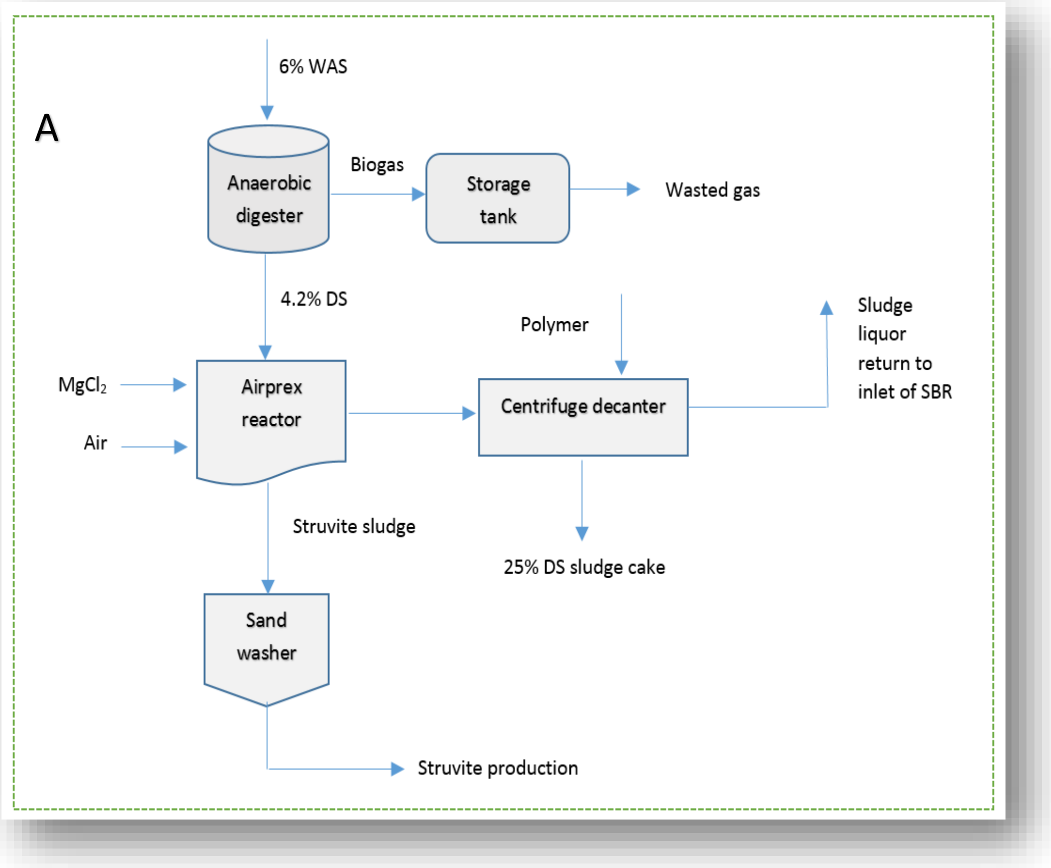
Table 33. Parameters of SBRs in the three upgraded processes and the existing process

Process	Cycle time (hour) per 1 SBR tank	Treating capacity of each reactor (m^3/day)	Number of reactors	Total capacity treated per day (m^3/day)	Operating temperature (^oC)
Existing process	4	37,237.6	4	148,950.0	20
Process A	6	24,825.1	6	148,950.0	20
Process B	6	24,825.1	6	148,950.0	20
Process C	5	29,790.1	5	148,950.0	20

5.3.1.2 Addition of extra units for phosphorus recovery in three upgraded processes

To provide an alternative phosphorus source to the agriculture and to alleviate pipe and pump clogging caused by uncontrolled struvite formation, P recovery from wastewater was integrated into Processes A, B and C. AirPrex technology was selected in the upgraded Process A due to its low chemical demand, low investment cost and applicability to sludge from the biological phosphorus removal process. In Process C, phosphorus could be removed by a combined EBPR and biologically induced phosphorus precipitation in aerobic granules (Manas et al., 2011; Manas et al., 2012). Gifhorn technology was, therefore, chosen due to its applicability to both EBPR and chemically precipitated phosphorus, high technical maturity and P recovery potential (Egle et al., 2016). For Process B, since phosphorus mainly exists in the form of chemical precipitate, Gifhorn technology was used for upgrading.

In the Gifhorn process, sludge from anaerobic digesters was digested first by adding 98% sulfuric acid at a pH of 4.5 to release metals and phosphorus. In the second step, the dissolved heavy metals were precipitated as sulfides by dosing sodium sulfide at pH 5.6 which was adjusted by sodium hydroxide, $Na(OH)_2$. After the solid and liquid separation by a decanter, magnesium hydroxide was dosed into the liquid stream in the second Gifhorn reactor at a pH of 9.0 adjusted by $Na(OH)_2$ for struvite crystallisation. The chemical consumption for each process for nutrient removal and recovery is shown in **Table 34**. Phosphorus recovery with either Airprex or Gifhorn is optional as additional units to integrate with upgraded three processes for nutrient removal. It is worth assessing how much environmental and economic burdens that phosphorus recovery could bring and comparing them with the benefits from it.


A
6% WAS
Anaerobic digester
Biogas
Storage tank
Wasted gas
4.2% DS
Polymer
Sludge liquor return to inlet of SBR
MgCl2
Air
Airprex reactor
Centrifuge decanter
Struvite sludge
25% DS sludge cake
Sand washer
Struvite production

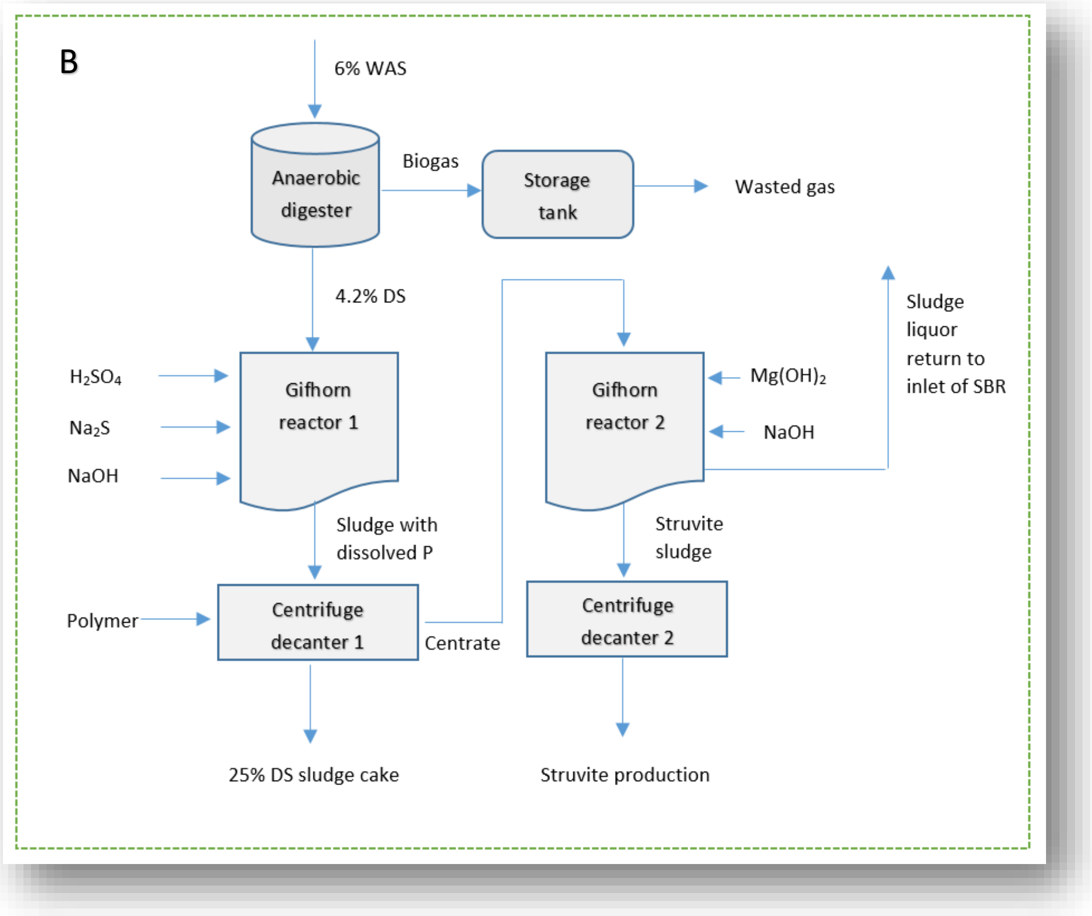


Figure 20. Detail process of P recovery units of; A) Airprex in process A and, B) Gifhorn in processes B and C

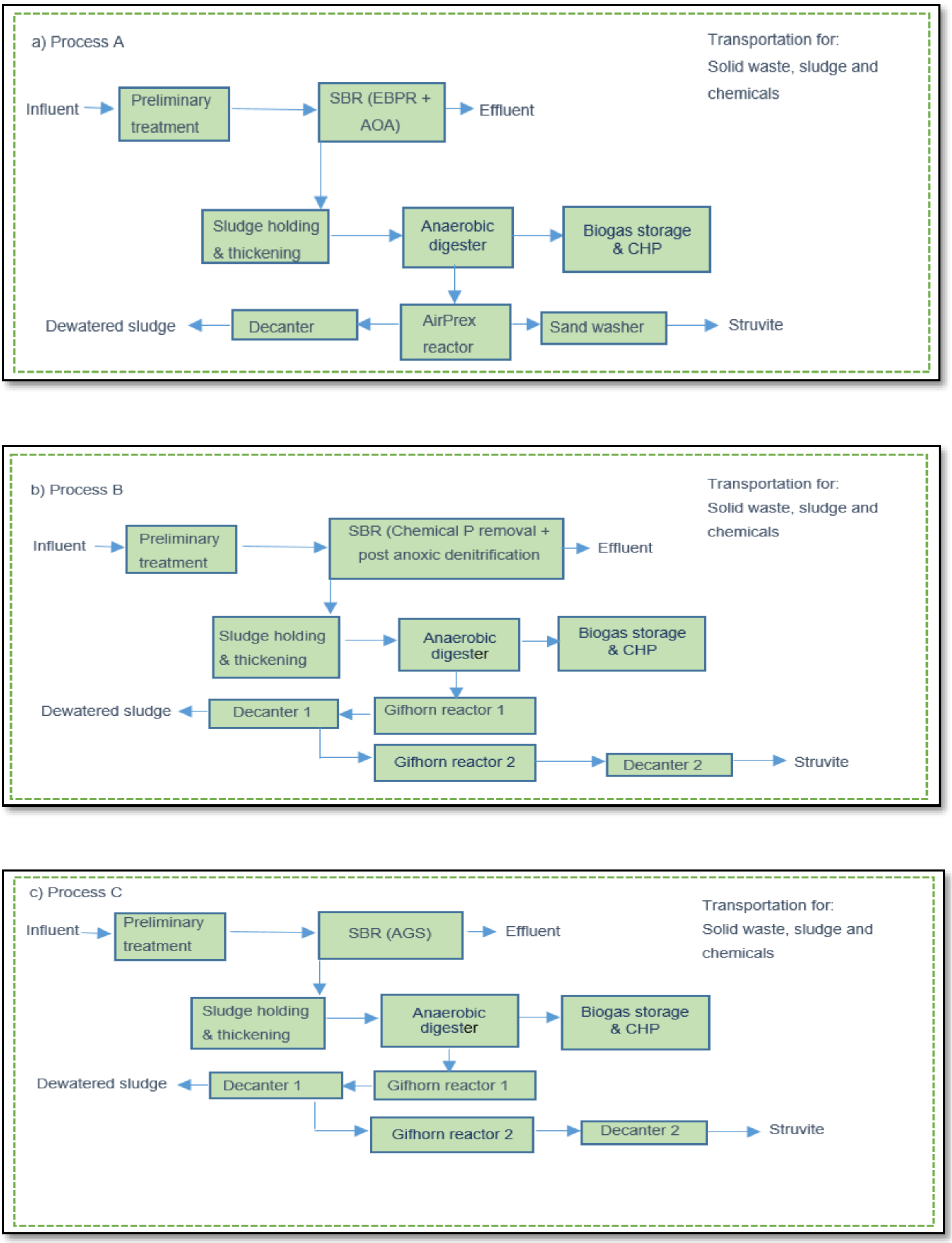


Figure 21. Schematic diagrams of the three upgraded Processes A, B and C which include the existing operating units and additional units after upgrading for nutrient removal in SBR, and phosphorus recovery

Note: (SBR = sequencing batch reactor; EBPR= enhanced biological phosphorus removal, AOA = Anaerobic: aerobic: anoxic, AGS = aerobic granular sludge, AD = anaerobic digestion)

Table 34. Chemical consumption in each upgraded process for nutrient removal and phosphorus recovery

Chemicals consumption	Unit	Process A	Process B	Process C
For nitrogen removal:				
Methanol (CH_3OH)	kg/day	3570.4	3904.2	-
For phosphorus removal:				
Iron chloride III ($FeCL_3$)	kg/day	-	2279.7	-
For phosphorus recovery:				
Magnesium chloride ($MgCl_2$)	kg/day	1306.2	-	-
98% Sulfuric acid (H_2SO_4)	kg/day	-	1189.6	1261.1
Sodium sulfide (Na_2S)	kg/day	-	116.1	123.0
Sodium hydroxide ($Na(OH)_2$)	kg/day	-	420.7	446.0
Magnesium hydroxide ($Mg(OH)_2$)	kg/day	-	29.0	30.8

5.3.2 Mass and energy balance of the existing process and three upgraded processes with nutrient removal and phosphorus recovery

Design parameters of the existing process and three upgraded processes are shown in **Table 35**. To evaluate wastewater treatment performance and to carry out environmental analysis of each unit in the treatment processes, mass balance and energy balance need to be conducted to provide parameters of; 1) each stream from each operation unit and 2) each operation unit in the whole process.

Table 35. Design parameters of the existing process and three upgraded processes

Parameters	Unit	Existing process	Process A	Process B	Process C
Cycle time in SBR	hour	4.0	6.0	6.0	5.0
Total treating capacity	m^3/d	148,950.4	148,950.4	148,950.4	148,950.4
pH range	-	6.8-8.0	7.0-7.5	7.0-8.0	6.5-8.0
Operating temperature	^oC	20.0	20.0	20.0	20.0
Total air supply rate in SBR	m^3/d	259,200.0	475,414.7	496,103.3	515,152.1
Oxygen required for additional nitrification	kg/day	-	8,125.6	8,885.5	9,585.1
Total energy consumption from blowers	kWh/day	8,520.0	17,067.0	17,809.8	18,493.7
Electricity demand of P recovery process	kWh/day	-	927.8	1,001.0	1,061.2
Total electricity consumption per day	kWh/day	30,609.2	40,084.0	40,900.2	41,644.4

5.3.2.1 Mass balance of the existing process and three upgraded processes

Based on the upgraded processes in **Figure 21**, mass balance was conducted and the mass flow of each stream is labelled in **Figure 18** for all three upgraded processes. Water quality parameters are shown in **Table 29**. With the addition of nutrient removal operation, total nitrogen and phosphorus removal efficiencies increased in the upgraded Processes A, B and C by 47% and 37% on average, respectively, compared with the existing process (**Table 36**). Whilst the removal efficiency of TSS and TCOD remain 92% and 90%, respectively, after the upgrading, similar to those in the existing process because the operation for TSS and TCOD removal were not changed by upgrading. In comparison with 15 mg/L TN and 1.05 mg/L TP in the effluent of the existing process, process C with aerobic granular sludge (AGS) achieved the best effluent quality with concentrations of TN and TP at 3.2mg/L and 0.06mg/L, respectively, due to its better treatment performance (Pronk et al., 2015; Chen et al., 2015). Piotr & Cydzik-kwiatkowska. (2018) also reported that the upgraded WWTP based on AGS in Poland achieved 87% of TN and 95% of TP removal efficiencies. Process A with activated sludge has the highest effluent concentrations, i.e. 5.4mg/L of TN and 0.24mg/L of TP. A potential reason could be the P release at anoxic condition, which is not easily resolved (Qiu & Ting, 2014; Zhou et al., 2016). But all three processes achieved satisfactory treatment performance in terms of nutrient removal.

For sludge production, the produced dry-weight sludge decreased by 24% on average in all three upgraded processes compared with the existing process, mainly due to the extended aeration used for nutrient removal as the extended aeration can reduce sludge (**Table 36**). In Process B, the increased sludge from ferric phosphate precipitate is less than the decreased sludge from extended aeration, resulting in a net sludge reduction by 22% compared with the existing process. One of the important reasons for this is that the influent was very low, i.e. 2.6 mg/L, which resulted in little inorganic phosphorus precipitate.

The production of struvite from 3 upgraded processes is shown in **Table 36**. It can be seen that Processes B and C based on Gifhorn produced much more struvite while recovered struvite from Process A with Airprex is only 60% of that from Processes B and C. As pointed out by Amann et al. (2018), P recovery potential of AirPrex process is 10%-22% with respect to WWTP influent, is relatively low compared to that of Gifhorn which can be up to 55%. However, when P recovery cost is considered, AirPrex process is cheaper with lower investment and less chemical demand (Egle et al., 2016). For instance, the cost of 1kg P recovered from Gifhorn process is up to 16€ (≈ 18.3USD) which is almost twice as that from AirPrex process (Egle et al., 2016). Therefore, it is necessary to look into the net costs of both technologies.

Table 36. Comparison of nutrient removal and phosphorus recovery performance between the existing process and the three upgraded processes

Parameters	Unit	Existing process	Process A	Process B	Process C
Total suspended solids removal rate	%	92	92	92	92
Total biochemical oxygen demand removal rate	%	90	90	90	90
Total nitrogen removal rate	%	46	83	86	90
Total phosphorus removal rate	%	60	92	93	98
Dry weight of dewatered sludge cake	kg/day	25,500	18,900	19,900	19,000
Struvite recovered	kg/day	-	712	1,147	1,216
Phosphorus recovered	kg/day	-	90	145	154

5.3.2.2 Energy balance of the existing process and three upgraded processes

The energy consumption in all processes is mainly from aeration, stirring in digesters and pumping fluids between different units. The energy consumption and generation from all processes are shown in **Table 37**. The total electricity consumption for secondary treatment in the upgraded processes increased by 30-34% compared with the existing process (at 18,121kWh/day), which is due to the increased energy consumption for nitrification in SBRs as removal of per g nitrogen demands 4.6 g oxygen. Addition of phosphorus removal units incurred more electricity consumption although P recovery only accounts for 2-3% of the total electricity consumption in the plant. With conventional nitrification/denitrification, it is unavoidable that nutrient removal is achieved at the expense of higher capital and operational costs. This highlights the importance of developing less energy-intensive nitrogen removal technology such as Anammox for mainstream nitrogen removal or high efficient energy recovery technology.

Considering more energy is consumed for nutrient removal, the implementation of sludge digestion and CHP for electricity production is imperative to alleviate carbon emission by reducing net electricity consumption per day in WWTPs. In addition, renewable energy production from sludge can improve the security of energy supply from WWTPs. The net electricity consumption in Process A and B was reduced by 3.1% and 3.6%, respectively, while it increased by 0.8% in Process C compared with the existing process due to its high electricity consumption per day mainly by the secondary treatment. In general, the recovered electricity from sludge just offset the energy used for nutrient removal, allowing equivalent net electricity consumption after upgrading.

Table 37. Comparison of energy consumption and generation in the existing process and the three upgraded processes

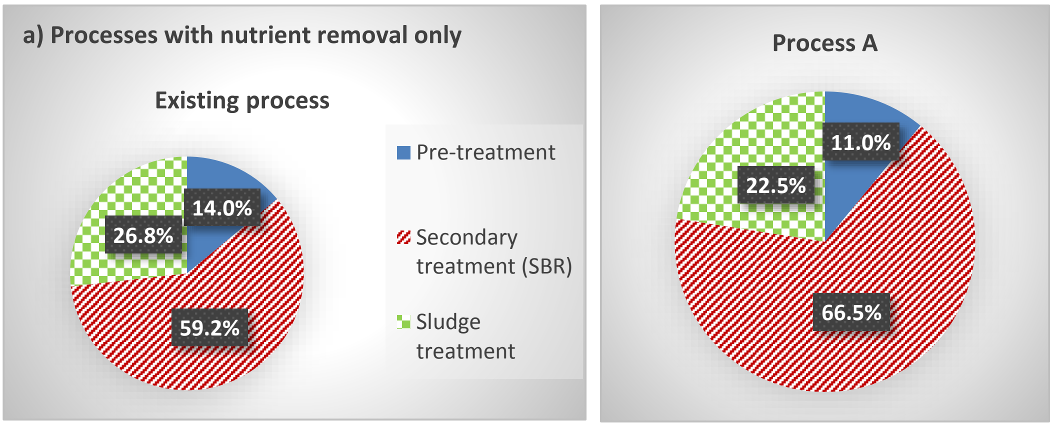
Parameters	Unit	Existing process	Process A	Process B	Process C
Methane gas production	m^3/day	-	2,580	2,773	2,665
Total electricity generated from CHP	kWh/day	-	10,426	11,380	10,784
Total electricity consumption for P recovery	kWh/day	-	928	1,001	1,061
Total electricity consumption for secondary treatment (SBR)	kWh/day	18,121.2	26,044	26,787	27,471
Total electricity consumption per day	kWh/day	30,609	40,084	40,900	41,644
Net electricity consumption per day (total electricity consumption – total electricity generated)	kWh/day	30,609	29,658	29,520	30,860

5.3.2.3 Hotspot analysis of existing process and the three upgraded processes in terms of electricity consumption

Two wastewater treatment scenarios, i.e. scenario 1 with nutrient removal only (**Figure 22a**) and scenario 2 with both nutrient removal and phosphorus recovery (**Figure 22b**), were considered to estimate electricity consumption for wastewater treatment in each process including primary treatment (screening and grid removal), secondary treatment (COD or COD and nutrient removal), sludge treatment and phosphorus recovery. In the existing process, the electricity consumption in the secondary treatment (SBR) process accounted for 59.2% of the total energy consumption due to intensive energy use for aeration in SBRs. This result is within the range in the study by Mininni et al. (2015) who estimated the electricity consumption for aeration in ten case studies mainly with modified ludzack ettinger (MLE) activated sludge process was within 43-60% of total electricity consumption. Gu et al. (2017) also reported in their study that the aeration process contributed to 60% of energy use in conventional activated sludge (CAS) wastewater treatment plant in China. In the three upgraded processes with nutrient removal, the distribution of energy consumption by

SBRs was almost the same, i.e. at around 67% which was higher than the existing process due to nitrification. There was almost no significant difference among the upgraded processes regarding electricity contribution from primary-treatment, secondary treatment, sludge treatment, and with/without P recovery because of similar technologies used for COD and nitrogen removal which are the primary energy consumers.

As shown in **Figure 22a** and **Figure 22b**, the electricity contribution in the secondary treatment increased from 59% in the existing process to 65-68% in the upgraded processes in both scenarios with/without P recovery. Although electricity was recovered in the upgraded processes, electricity still accounted for 2/3 of the total electricity, even higher than the existing process without electricity generation. Although pre-treatment units were the same before and after upgrading, the percentage of electricity consumption for pre-treatment and sludge treatment in the upgraded processes reduced by around 3% and 5%, respectively, compared with the existing process because of more electricity consumed for nutrient removal. In addition, the comparison between two scenarios with and without P recovery showed that introducing P recovery units, either AirPrex or Gifhorn, only contributed less than 2.5% of electricity. These comparisons indicate the importance of developing energy-saving nutrient removal technologies to replace conventional biological nitrification for reduced energy consumption and/or to enhance energy recovery from wastewater to cover more electricity consumed for nutrient removal.



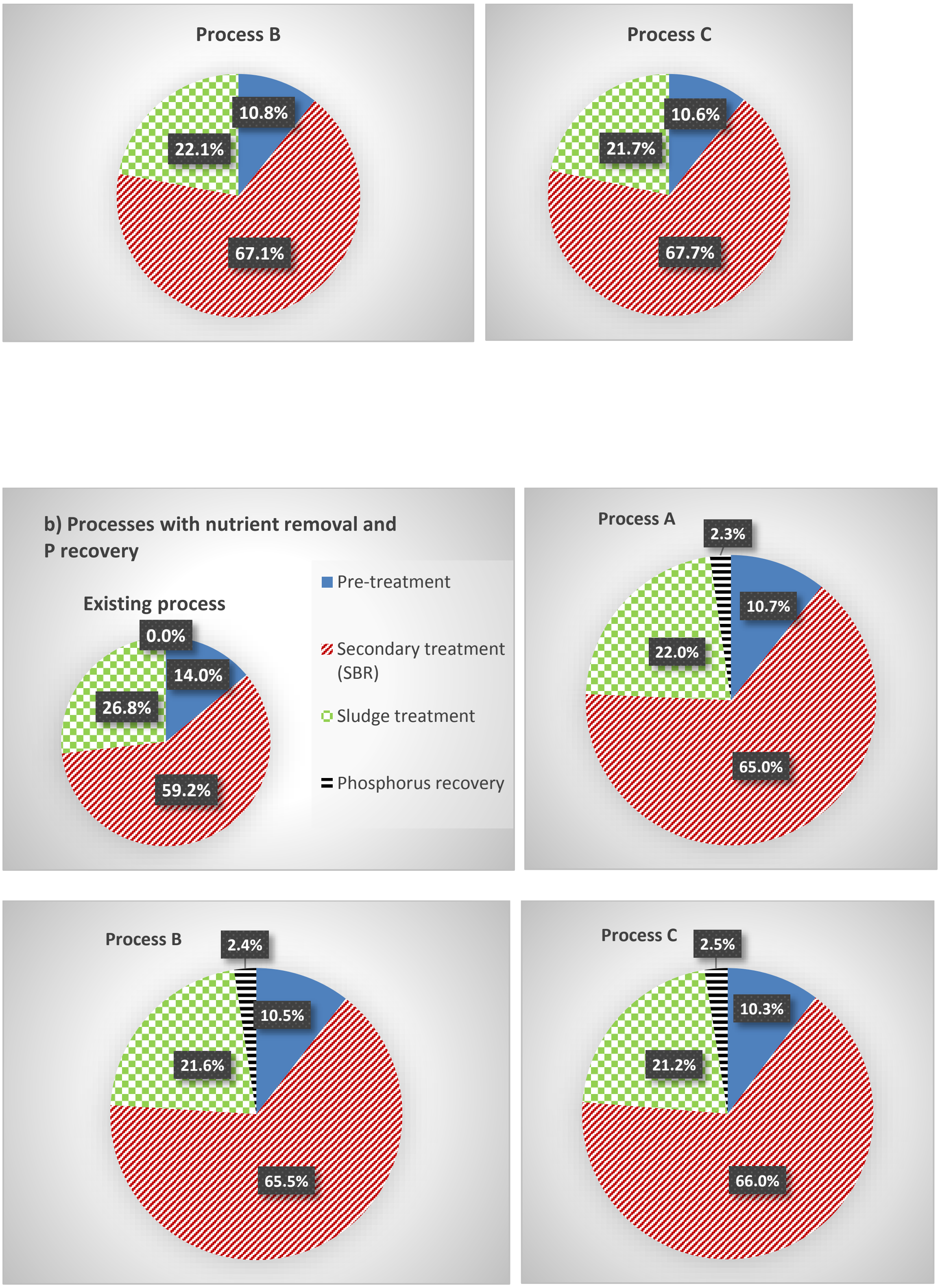


Figure 22. Hotspot analysis of the existing process and the three upgraded processes in terms of electricity consumption with: a) Scenario 1: three upgraded processes with nutrient removal only, and b) Scenario 2: three upgraded processes with both nutrient removal and phosphorus recovery.

5.3.3 Environmental impact analysis of the existing process and three upgraded processes for wastewater treatment

The purpose of upgrading the existing process is to improve effluent quality and recover resources such as energy and phosphorus from wastewater to enhance environmental protection. To assess the holistic environmental benefit, environmental impact analysis with LCA was carried out to guide the selection of newly designed processes. As shown in **Figure 23**, it can be seen that except for eutrophication potential, the environmental impact was largely derived from the operation of treatment plants. Construction and demolition only contribute less than 10% in each impact category for all four processes. A similar finding was reported by Foley et al. (2010) and Hao et al. (2019) that the operation of WWTP contributed more than 90% to environmental impact categories compared with construction and demolition phases. The only environmental benefit in the existing process was from steel recycling in the demolition phase with -7.5% in HTP, and -1.2% in ADFP and GWP, respectively.

In the existing process, electricity consumption contributes 57-95% to five environmental impact categories namely human toxicity potential (HTP), acidification potential (AP), abiotic (fossil fuel) depletion potential (ADFP), freshwater ecotoxicity potential (FEP) and global warming potential (GWP), while eutrophication potential (EP) is mainly from effluent discharge. In the upgraded processes, it is found that electricity recovery benefited the environmental impacts in all six categories particularly in GWP, with an average of 19% reduction. To increase energy recovery, Adsorption-Biological (A-B) process could be used for more capture of carbon (Jonasson, 2007). But for SBRs in this study, it is less likely to upgrade process to A-B process. The upgrading solutions are restricted by the current technology used in the existing plant.

The upgraded processes had additional demand for chemicals for denitrification, phosphorus precipitation and phosphorus recovery to produce struvite, causing additional environmental burdens to all environmental categories except eutrophication. The Process B had the highest chemical consumption due to the consumptions of ferric chloride for phosphorus precipitation, methanol dose for denitrification, as well as sulphuric acid, sodium hydroxide, sodium sulfide and magnesium chloride for P recovery, leading to the highest environmental impact. The chemical contribution to ADFP in Process B reached 28%, the second-highest contributor after electricity. Simultaneous nitrification and denitrification, and the combined EBPR and biologically induced phosphorus precipitation in aerobic granular sludge in Process C required no chemicals for nutrient removal (Piotr & Cydzik-kwiatkowska., 2018; Pronk et al., 2015), making it the most promising technology to upgrade the existing SBR plants for wastewater treatment.

Resource recovery from three upgraded processes created environmental benefits in all six environmental impact categories. For instance, the recovery of electricity and phosphorus from the operation, and steel recycled from demolition contributed up to 19% in GWP. P recovery alone, however, only provided 2-5% environmental benefits by reducing rock phosphate mining. The small net environmental benefit brought by P recovery is also partly due to low P recovery efficiencies accompanied by large amounts of energy and chemical input for nutrient removal (Pradel & Aissani., 2019). The other reason is low influent phosphorus concentration in Malaysian STP at around 2.6 mg/L. The environmental benefit of phosphorus recovery could increase with the increase in influent phosphorus concentration. Therefore, it needs to be careful to consider building phosphorus recovery units in WWTPs with diluted municipal wastewater from an environmental impact perspective. Consequently, more sustainable P recovery technologies with higher P recovery efficiency are needed. It is reasonable to expect that further incremental improvement of the current Airpex and Gifhorn based phosphorus recovery technologies cannot significantly increase environmental benefit from P recovery. Transformative technologies such as separation of black water from other domestic wastewater for P recovery (Verstraete & Vlaeminck., 2011) or more advanced membrane technology for direct phosphorus recovery from municipal wastewater (Qiu & Ting., 2014) might be able to achieve significantly higher environmental benefit. But it needs to point out that the benefit from P recovery should not be limited to positive environmental impact only because P recovery also alleviates the risk of phosphorus depletion within the next 50 to 100 years. This is why even with a small environmental benefit, many countries encourage P recovery in WWTPs. For example, Sweden has regulated that at least 60% P should be recovered from the total wastewater phosphorus (Hultman et al., 2004).

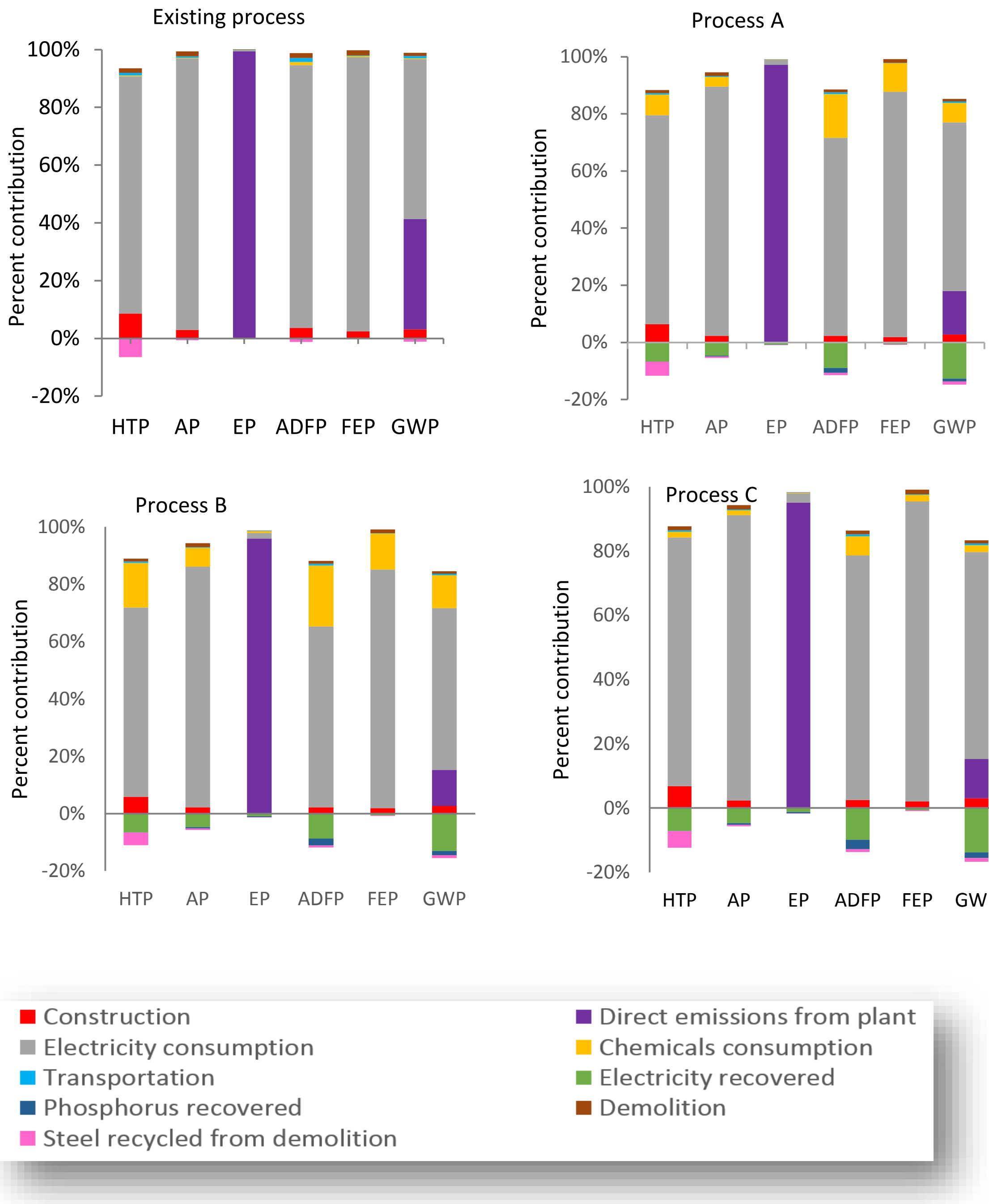


Figure 23. Environmental impact contribution analysis based on different factors in the existing process and the three upgraded processes by using FU1.

Note: (HTP-human toxicity potential, AP-acidification potential, EP- eutrophication potential, ADFP-abiotic depletion (fossil fuel) potential, FEP-freshwater ecotoxicity potential, and GWP-global warming potential).

In terms of total environmental impacts in each category, **Figure 24** shows that the existing process had the lowest impact in HTP, AP, ADFP and FEP categories while the upgraded processes benefit EP and GWP categories. EP reduction in the upgraded processes was mainly due to nutrient removal, while GWP reduction was due to electricity recovery. The comparison between the three upgraded processes indicates that Process C had the lowest impact compared to Processes A and B in all categories (between 5 - 37%) due to less chemical consumption by AGS and a high nutrient removal efficiency. Thus, in terms of total environmental impact without considering economic cost, Process C that integrating nutrient removal by AGS, phosphorus recovery and electricity recovery is the best option for upgrading the Malaysian STP. This result could be the guideline to decision-makers for technology selection when considering technical and environmental impacts.

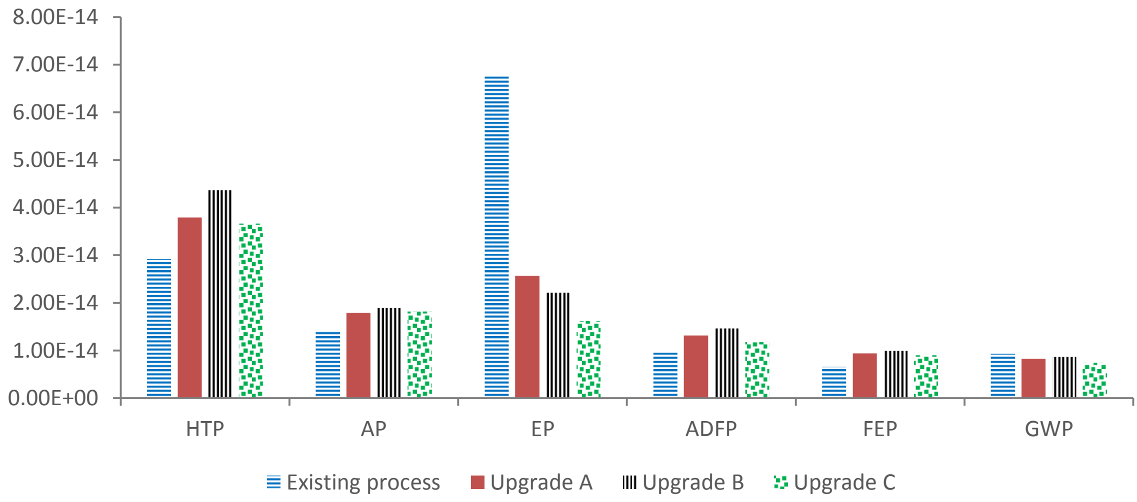


Figure 24. The comparison of environmental impact values between the existing process and the three upgraded Processes A, B and C using FU1 (1 m^3 of treated wastewater).

Note: (HTP-human toxicity potential, AP-acidification potential, EP- eutrophication potential, ADFP abiotic depletion (fossil fuel) potential, FEP-freshwater ecotoxicity potential, and GWP-global warming potential)

5.3.4 LCA of existing process and three upgraded processes with and without P recovery

P recovery from wastewater has been demonstrated at full scale, but it has not been widely adopted due to high cost. As a developing country, Malaysia is less likely to implement P recovery in the near future. It is thus very necessary to compare the environmental impacts of wastewater treatment processes with and without P recovery to provide quantitative data to allow operators, engineers or policymakers to make informed decisions when upgrading the existing wastewater treatment plants. Also, it can provide information to researchers to understand the environmental impact from phosphorus recovery. The comparative results of LCA between the existing process and the three upgraded processes (with and without P recovery) are shown in **Figure 25**. EP is mainly dependent on nitrogen and phosphorus concentrations in the effluent of WWTPs. It can be seen that EP was reduced by 62% in Process A, 67% in Process B and 76% in Process C, respectively, compared with that in the existing process, due to nutrient removal. GWP in upgraded processes was reduced to 7-22% compared to the existing process. It is due to the consequence of electricity generated in the upgraded processes. This reduction highlights the importance of electricity recovery to reduce global warming (Xu et al., 2014). HTP, AP, ADFP and FEP impacts from the upgraded processes were averagely 23% higher compared to the existing process due to increased chemical consumption, especially in Process B. In overall, the upgraded processes with nutrient removal and resource recovery in this study had positive environmental impacts on EP and GWP while there were negative impacts on HTP, AP, ADFP and FEP.

Figure 25 also shows the environmental impact comparison between two scenarios; i. the upgraded processes with nutrient removal only (without P recovery) and; ii. the upgraded processes with both nutrient removal and P recovery. Eutrophication potential impacts in Processes A, B and C were similar in both scenarios because EP was mainly affected by concentrations of pollutants in the effluent (i.e. TCOD, TN and TP). However, other impact categories such as HTP, AP, ADFP, GWP and FEP experienced negligible or small increase ranging from 0.9% to 7.6%, which are the net results from the additional energy and chemical demands for the phosphorus recovery. This indicates that P recovery in this study led to a negligible net impact on the environment. Instead, the substantial environmental loads imposed by the production of mineral fertiliser could be avoided indirectly (Hao et al., 2019).

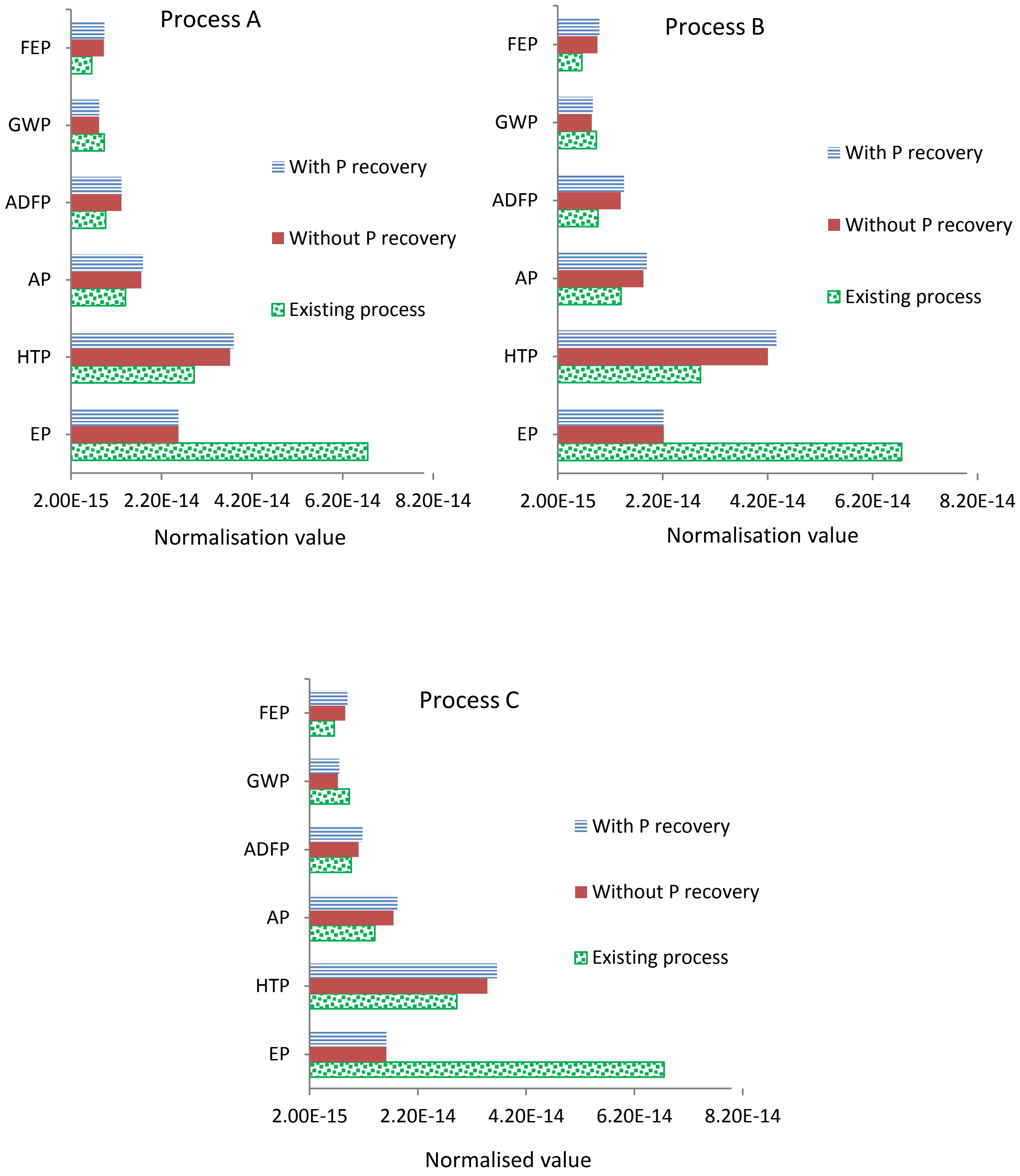


Figure 25. Comparison of environmental impacts between the existing process and the three upgraded processes with nutrient removal only (without P recovery), and the upgraded processes with both nutrient removal and P recovery by using FU1

5.3.5 Effects of the functional units on LCA results for the three upgraded processes with P recovery

The life cycle inventories of FU1, FU2 and FU3 for all three processes are shown in **Table 30, Table 31 and Table 32 respectively**. With per m^3 treated wastewater as FU1 and per kg PO_4 eq. as FU3, Process B had the highest environmental burden in all categories studied except eutrophication, due to its most chemical and energy consumption to treat per m^3 wastewater or per 1 kg eutrophication reduction as shown in **Figure 26a and Figure 26c**. However, by using per kg recovered struvite as FU2, the environmental impact from Process A was averagely 42% higher than those from Processes B and C in all six categories as shown in **Figure 26b** due to high energy and chemical consumption for 1kg recovered struvite. It is mainly due to the low struvite recovery efficiency by Airprex in process A. It is noted that Process A contributed to highest eutrophication potential impact with all three functional units. The inconsistent results from three different functional units suggest the importance of FU selection in LCA, which should be based on different purposes. For the integrated wastewater treatment process with struvite recovery, FU2 is more suitable because it represents the environmental burden from per unit of P/struvite recovered as applied by Amann et al., 2018. FU1 is more suitable for the comparison of processes or technologies for conventional wastewater treatment as conducted by Lorenzo-Toja et al., 2016 and Hauck et al., 2016. While FU3 is more suitable to the advanced wastewater treatment with nutrient removal due to the consideration of pollutants removed as functional unit. However, regardless of the functional unit, Process C always had the lowest impacts of six studied categories among all upgraded processes due to its cleanest effluent and lowest energy and chemical use. Therefore, Process C is recommended as the best technology with the least environmental burden from the aspects of wastewater treatment and struvite recovery. This further indicates the promising prospect of aerobic granular sludge technology for sustainable wastewater treatment. The consideration of more than one functional units in this study suggesting an improvement in LCA-WWTPS methodology which could be used as guideline for future study/research involving advanced treatment of WWTPs. This is because, previous studies regarding advanced treatment technology only focusing on one 1 functional unit without comparison to others as conducted by Fang et al., 2016 and Rahman et al., 2016.

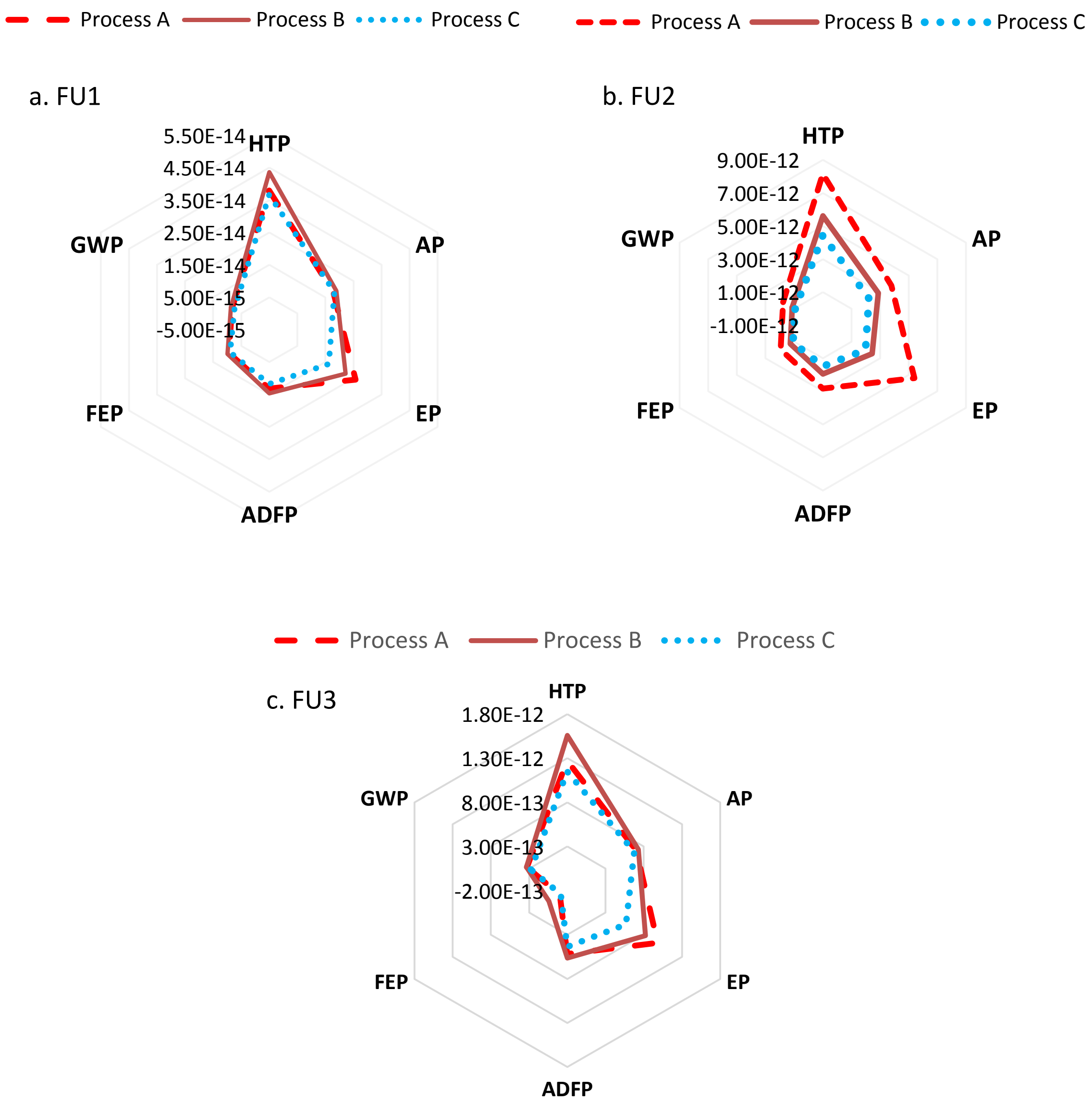


Figure 26. Comparison of environmental impacts from the three upgraded processes by using FU1 (1 m^3 treated wastewater), FU2 (1 kg struvite recovered) and FU3 (1 $kgPO_4^{3-}$-eq.)

5.3.6 Sensitivity analysis

The environmental impact results in this study show that Process C has the least environmental burdens. To further investigate how the variability of inventory data affects environmental impact results, this study used Process C as a case study for a sensitivity analysis. **Table 38** shows the variability of environmental impact results by varying inventory data such as electricity consumption, nutrient concentrations in effluent and chemical consumption by ±10% in 20 years and ±20% of 40 years, respectively. All output variance is within the ranges of input variance. In general output variance corresponding to electricity is the highest while it is the least except eutrophication potential to nutrient concentrations in effluent. For example, environmental impact categories such as HTP, AP, ADFP, FEP and GWP varied from ±7.42% to ±9.98% to respond to the change in electricity consumption by ±10% (in 20 years) and these same impact categories varied from ±14.74% to ±19.92% from the change in electricity consumption by ±20% (in 40 years). EP changed by ±9.1% and ±18.05%, respectively, to respond to ±10% and ±20% changes in TP and TN concentrations in the effluent while the other five impact categories were almost unaffected. Finally, the variance of chemical consumption led to less effects on all outputs compared with electricity. The highest change corresponding to chemical input was FEP, which was ±6.62% in 20 years and ±13.04% in 40 years, respectively, much lower than the input variance. These results are in agreement with those reported by Piao et al. (2015) that variance in electricity consumption caused the most sensitive change to AP and HTP in all WWTPs studied. The fact that the variation in electricity consumption, chemical consumption and nutrient concentrations in the effluent by 10%-20% does not cause an environmental impact output change by more than 20% suggests a less sensitivity of environmental impact results to inventory data in this study. This means the results in this study from the current database are applicable for the WWTP with long design life or for the circumstance with a certain level of variability in the dataset.

Table 38. Sensitivity analysis result by changing selected inventory data by ±10% and ±20% for Process C in 20 years and 40 years respectively, according to 1m^3 of treated wastewater (FU 1)

	Process C in 20 years (±10%)			Process C in 40 years (±20%)		
Inventory components:-	Electricity consumption	TN&TP in effluent	Chemical consumption	Electricity consumption	TN&TP in effluent	Chemical consumption
Potential impacts:						
Human toxicity (HTP)	±9.90	±0.00	±2.90	±19.60	±0.00	±5.72
Acidification (AP)	±9.98	±0.00	±0.65	±19.92	±0.00	±1.29
Eutrophication (EP)	±0.55	±9.10	±0.07	±1.11	±18.05	±0.13
Abiotic depletion (fossil fuels)(ADFP)	±9.83	±0.00	±1.94	±19.53	±0.00	±3.77
Freshwater ecotoxicity (FEP)	±9.96	±0.00	±6.62	±19.91	±0.00	±13.04
Global warming (GWP)	±7.42	±0.00	±2.35	±14.74	±0.00	±4.66

TN and TP = Total nitrogen and total phosphorus

5.3.7 Economic evaluation of the three upgraded processes

The life cycle costs (LCC) of the existing and the three upgraded processes with P recovery based on per population equivalent (PE) per day are shown in **Table 39**. Positive values represent the cost required for treatment/operation while negative values mean the money earned by the plant from the resource recovery. The total LCC of Process A, B and C are averagely 24% higher than that of the existing process (0.0092 USD/PE.day), with Process A having the lowest LCC in three upgraded processes. It is because that additional nutrient removal and resource recovery in upgraded processes increased capital cost and operating cost. Morelli et al. (2019) reported an increase in net life cycle cost of the upgraded process by 17% in a small community wastewater treatment plant with 3,800 m^3/day flowrates after being upgraded for biological nutrient removal with enhanced primary settling and anaerobic digestion (AD). This highlights roughly equivalent additional cost required for the upgrading of WWTPs in both large and small scale plants. Besides, the economic gains from the recovered electricity and phosphorus, and the reduced sludge disposal contributed to the reduction in the net life cycle cost in the three upgraded processes in this study.

Similarly, Xu et al. (2014) reported that 13 sewage sludge treatments in China gained environmental and economic benefits by applying sludge digestion and electricity recovery. Although Process C had the lowest negative environmental impact, it had almost a similar life cycle cost with that of Process B. Processes B and C were 21.3% and 19.5% more expensive than Process A respectively, mainly due to the more chemical consumption in Gifhorn than Airprex process. Thus, from the point of view of economic cost, Process A (i.e. the integration of EBPR and nitrification-denitrification for nutrient removal, Airprex for P recovery and anaerobic digestion for electricity recovery with CHP) is the optimum option among the three upgrading processes.

Table 39. Life cycle cost of the existing and the three upgraded processes in construction and operation phase

Phase	Construction	Operation				Benefit from operation		
Cost (USD/ PE.day)	Capital cost	Electricity consumption	Chemicals consumption	Transport	Disposal fee in landfill	Electricity recovered	Phosphorus recovered	Life cycle cost
Existing process	1.7E-3	6.5E-3	3.9E-4	3.7E-4	2.0E-4	-	-	9.20E-3
Process A	2.5E-3	8.4E-3	1.2E-3	2.7E-4	1.5E-4	-2.1E-3	-1.7E-4	1.03E-2
Process B	4.6E-3	8.6E-3	2.0E-3	2.9E-4	1.5E-4	-2.3E-3	-2.7E-4	1.31E-2
Process C	4.5E-3	8.8E-3	1.6E-3	2.7E-4	1.5E-4	-2.2E-3	-2.9E-4	1.28E-2

5.4 Conclusion

Three processes were designed to upgrade a centralised wastewater treatment plant with SBR technology for nutrient removal and phosphorus recovery. All technologies selected for the upgrading are commercially available to ensure the practical feasibility of the upgraded wastewater treatment, and the meaningful results and conclusions to decision-makers and other researchers. To evaluate the environmental benefits/burdens of the existing and upgraded processes, environmental and economic assessments using LCA were carried out. The main conclusions are summarised as below.

- Upgrading the existing plant for nutrient removal, phosphorus recovery and electricity generation benefits the environment by reducing EP by 62-76%, and GWP by 7-22%. However, these environmental benefits were gained at the cost of increases in HTP, AP, ADFP and FEP by averagely 23%. Therefore, a trade-off between different environmental categories needs to be considered for upgrading especially when protecting the local water eco-system.
- The upgraded Process C is recommended as the best technology with the least environmental burden from the aspects of wastewater treatment and struvite recovery indicating promising prospects of aerobic granular sludge technology for upgrading the existing WWTPs with SBR technology for sustainable wastewater treatment due to its better nutrient removal performance and less chemical consumption.
- The added phosphorus recovery with either Airprex or Gifhorn technology only contributes to 2-5% environmental benefit. This is mainly due to the low influent phosphorus concentration in this study such as around 2.6 mg/L, leading to low P recovery efficiencies. The environmental benefit of phosphorus recovery could rise with an increase in influent phosphorus concentration. Therefore, it needs to be careful to consider adding phosphorus recovery units in WWTPs with diluted municipal wastewater from an environmental impact perspective.
- FU2 (per kg struvite recovered) is more suitable when considering the environmental impact from per kg P recovered from wastewater. FU1 (per m^3 wastewater) is more preferred to evaluate environmental performance for treating per m^3 wastewater. Process A with EBPR and Airprex has the highest environmental burden in terms of per kg P recovered while Process B with chemical P precipitation and Gifhorn shows the highest environmental impact in terms of per m^3 wastewater treated. Process C has the least

environmental impact with either of FU. This provides a guideline for the process selection and highlights the environmental sustainability of aerobic granular sludge technology

- The total life cycle costs of Processes A, B and C were averagely 24% higher than the existing process (0.0092 USD/PE·day) due to increased capital and operating costs. Process C was 19.5% more expensive than Process A mainly due to the more chemical consumption in Gifhorn than Airprex process although Process C had the lowest environmental impact. When phosphorus recovery is needed, more technology combinations such as coupling aerobic granular sludge for nutrient removal with Airprex for phosphorus recovery need to be explored to achieve both minimum environmental impact and economic cost.

This work identified the importance of considering both local wastewater characteristics and the current technology being used in the existing process for selecting technology and relevant process configurations to upgrade an existing WWTP. In addition, technological, economic and environmental assessment is critical to compare different processes to get the best option. The quantitative information from this study could guide decision-making to upgrade existing WWTPs especially in regions with diluted wastewater, which can underpin the transition towards sustainable wastewater treatment.

6 Discussion and conclusions

6.1 General discussion

6.1.1 Summary of key results

This thesis addressed the improvement of Life Cycle Assessment (LCA) methodology by providing comprehensive data inventory and detail analysis to better assess the environmental impact from municipal Wastewater Treatment (WWT), and provide guidance towards sustainability strategy especially to decision makers. The thesis comprised of three result chapters which each contribute towards the fulfilment of the primary research aim which is described below.

Objective 1 (Chapter 3): *To investigate the influence of rainfall on the environmental impacts of WWTPs in two scenarios, i.e. large centralised WWTPs with high strength wastewater (MWTW) and low strength wastewater (MSTP) respectively, but with similar rainfall effects on influent flow rate*.

- Results indicate that for either combined or separate sewer system, rainfall does affect wastewater influent flow rate, wastewater influent and effluent quality, and power consumption which further influence the overall environmental impact from WWTPs.
- The coefficients between monthly rainfall and the influent flow rate to MSTP (i.e. with a separate sewer system and yearly precipitation of 2200mm), and the influent flow rate to MWTW (i.e. with a combined sewer system and yearly precipitation of 870mm), respectively, are similar at around 2500 m^3 influent flow rate/mm precipitation.
- There was a strong correlation, r between rainfall and the influent flow rate in the wastewater treatment plants with either combined sewer system (r=0.66) or separate sewer system (r=0.84).
- In the environmental impact, the rainfall effect is more obvious on the eutrophication potential and global warming potential than on other environmental indicators, while the type of sewer system i.e. combined or separate, seems not important in the two cases studied.
- Both wastewater treatment plants (WWTPs) show a lower environmental burden in the wet season than in the dry season as there are lower pollutant concentrations in the effluent due to the dilution of wastewater in the wet season, by using FU1 (per m^3 treated wastewater).

- However, MWTW shows a more obvious difference between two seasons for all impact categories while MSTP is more or less comparable except for the EP category. It is because that the strength of sewage in MWTW is much higher than that in MSTP, and the dilution during wet season plays a much obvious role in MWTW for reduced electricity consumption as well as reduced pollutant concentrations. Therefore, raw sewage strength is a key factor to lead to different environmental impact in the dry and wet seasons.
- Meanwhile, result by using FU2 (1 $kgPO_4^{3}$-eq removed) shows that MWTW exhibits lower environmental impacts compared to MSTP due to its high nutrient removal efficiency and better effluent quality. For MSTP, each environmental category in dry and wet seasons shows a similar trend when using FU1 and FU2. This suggests that no much difference is caused by adopting different functional units to WWTP with low strength wastewater.
- MWTW, however, demonstrates higher environmental impact in the wet season than the dry season with FU2, which is contrary to that by using FU1 due to the less efficient treatment caused by heavy rainfall during the wet season. Thus, considering pollutant removal efficiency during the wastewater treatment process, using FU2 is more appropriate for an environmental impact assessment. Using FU2 also makes the direct comparison between different WWTPs, or different seasons more meaningful as it is mainly based on pollutant removal by minimising the effect from influent compositions and flows.
- Overall, the results indicate that the environmental burdens particularly eutrophication and global warming caused by WWTPs are dependent on the correlations of rainfall intensity with wastewater quantity and quality, and the selection of functional units. This could be used to guide a stricter control of eutrophication in a more sensitive season (e.g. dry season) or in more vulnerable receiving waters.

Objective 2 (Chapter 4): *To assess toxicity impact of PPCPs and metal emissions from a large centralised wastewater treatment plant in Malaysia with low wastewater strength, and to provide useful information for LCA toxicity assessment practice by identifying the importance and contribution of PPCPs and metals and comparing LCIA models.*

- The removal efficiency of heavy metals from influent to the effluent of Malaysian STP with the SBR system is ranging from 8.8% to 73.0%, while there is good removal of PPCPs ranging from 46.9% to 99.2%.
- However, unlike COD and nutrient removal efficiencies which are usually lower in developing countries, no clear pattern in metals and PPCPs removal exist between developing and developed countries. They are probably closely related to lifestyles, commodities used locally and the acceptance of the use of industrial and commercial wastewater. This highlights the significance of obtaining data from site sampling instead of those from published literature to represent the real effects of heavy metals and PPCPs on human and the environment.
- Terrestrial ecotoxicity impact increased by 88% from the inclusion of metals in sludge due to the pollution by heavy metals through water and soil compartment when sludge is landfilled or applied to the land. Freshwater ecotoxicity potential increased by 76% when comprising metals and PPCPs in the effluent of MSTP. This high impact indicates the importance of considering PPCPs and heavy metals on the toxicity impact regardless of the wastewater strength and the treatment technology used.
- In this type of WWTP in Malaysia, direct emission of heavy metals from effluent and sludge, and indirect emission by electricity generation from fossil fuel such as coal and oil, were the main contributors to the three toxicity impact categories which are human toxicity, freshwater ecotoxicity and terrestrial ecotoxicity. Thus, the reduction in toxicity relies on the shift of energy source from fossil fuel to renewable energy such as solar energy, wind, hydroelectric, etc., or the reduction in energy consumption for wastewater treatment, as well as the removal of key heavy metals from wastewater.
- PPCPs contributed 11% to FEP by using CML-IA method but only contribute 2.5% using USEtox method. This different contribution of PPCPs is mainly due to the difference of CFs value/magnitude provided by the two models. Among PPCPs substances, 17b-estradiol is the pollutant which contributes most to FEP in both methods.
- In the comparative analysis of metals emission to toxicity-related impact categories, CML-IA, Recipe and IMPACT 2002+ produce consistent results of HTP, FEP, and TEP from metal emissions. Whereas EDIP and USEtox provided different results from those three methods. This model comparison provides useful information for future LCA practice on the selection of LCIA methods for toxicity assessment, according to their necessities.
- In the sensitivity analysis, HTP, FEP and TEP change with ±9.89%, ±3.95% and ±2.75% respectively from ±10% variation in electricity consumption. Meanwhile, the ±10%,

variation in concentration of top 3 metals in sludge, and top 3 metals and PPCPs in wastewater only affecting less than ±5% to human toxicity and ecotoxicity impacts.

Objective 3 (Chapter 5): *To design upgrading processes based on an existing Malaysian centralised wastewater treatment plants for nutrient removal and resource recovery by combining local conditions and to assess economic burdens and environmental benefits or burdens of upgraded processes with life cycle assessment*.

- Three upgraded processes were designed based on existing operation of Malaysian STP. Process A is based on enhanced biological phosphorus removal (EBPR) for phosphorus removal, nitrification and denitrification for nitrogen removal and AirPrex for P recovery. Process B is to use ferric precipitation to remove phosphorus, nitrification and denitrification to remove nitrogen, and Gifhorn to recover P from sludge. Process C is to adopt aerobic granular sludge (AGS) technology to do simultaneous nitrogen and phosphorus removal, and Gifhorn for P recovery.
- For all three upgraded processes, the extension of cycle time extends the hydraulic retention time (HRT) which reduces the treatment capacity of the existing 4 operated SBR reactors. Thus, process A and B require additional 2 reactors, and process C requires additional 1 reactor to keep the same treatment capacities.
- Process C based on AGS achieves the best effluent quality indicating high treatment efficiency of AGS. For P recovery, process A based on Airprex only recorded 60% struvite of that from process B and C, indicating that Gifhorn is more efficient for P recovery technology.
- In the hotspot analysis, P recovery only contributes 2-3% of energy consumption in all three processes while SBR with nutrient removal contributes up to 68% of energy consumption. This highlights that effort to reduce energy consumption should be the secondary treatment.
- In terms of environmental impact, process C caused the lowest impact in all categories due to several factors such as less chemical consumption by AGS and high efficiency in nutrient removal.
- With FU1 and FU3, process B had the highest environmental burden due to its more chemical and energy consumed to treat per m^3 wastewater and per kg eutrophication reduction respectively. However, by using FU2 with per kg struvite recovered, the environmental impact from process A was averagely 42% higher than those from

processes B and C in all 6 categories due to high energy and chemical consumption for 1kg recovered struvite. It is mainly due to the low struvite recovery efficiency by Airprex in process A.

- Based on the economic assessment, the total life cycle cost (LCC) of process A, B and C is averagely 24% higher than the existing process (0.0092USD/PE.day). It is because the added nutrient removal and resource recovery in upgraded processes increased capital cost and operating cost. Nevertheless, the economic gain from the recovered electricity and phosphorus, and the reduced sludge disposal facilitated in reducing net life cycle cost in the three upgraded processes in this study.

6.1.2 Significance and implication of the work done

With the main objective of this thesis 'To assess the life cycle impact of large centralised WWTPs based on the extended and comprehensive local databases for the improvement of LCA methodology and sustainable operation of wastewater treatment', the result of this thesis can be very useful for LCA practice and wastewater management. The result and finding from this thesis are interesting and important for further development within the LCA research community and the proposed analysis method can be directly applicable in practice. Stakeholders can benefit from the outcomes of this thesis by the findings from real case application, which demonstrates the usefulness of environmental impact assessment to decision making. The specific contribution to knowledge from each result in Chapter 3, 4 and 5 are described and demonstrated as below;

Chapter 3:

- This research collected and provided complex inventory data of a Malaysian STP for the whole year of 2016, which is the first for LCA of Malaysian municipal WWTP. This will contribute to the understanding of environmental impacts of large WWTPs in developing countries with representative climate pattern and sewer system.
- It was found and reported for the first time that the effect/correlation of rainfall on wastewater inflow to the WWTP with a separate sewer system in Malaysia, is similar to that in the WWTP with a combined sewer system in the UK. To the best of my knowledge, there is no any study on rainfall influence on environmental impacts of WWTPs with separate sewer systems because it is taken for granted that rainfall has a negligible effect on inflow to WWTP with a separate sewer system. This work will cause a paradigm shift for WWTPs with separate

sewer systems.

- For WWTPs with combined sewer systems, the previous environmental impact studies on rainfall influence using LCA are contradictory, indicating some important factors affecting results are missing. This work identified two critical factors which affect LCA assessment and interpretation. They are wastewater strength and functional unit. This identification will improve LCA practice for WWTPs as well as the improvement of WWTPs management to alleviate environmental impacts.

Chapter 4:

- This research provided a comprehensive extended inventory data of Malaysian STP by conducting site sampling and analytical works of pollutants including organic matter, nutrients, heavy metals and pharmaceutical and personal care products (PPCPs). This inclusion of a group of micropollutants is the first in LCA study to Malaysian STP. The result contributes to the understanding of occurrence and removal of these pollutants in developing country situation with low strength wastewater.
- It was reported for the first time a complete LCA study for human toxicity and ecotoxicity impacts that compares metals from electricity production, chemical consumption, transportation, local metals in effluent and sludge, as well as local PPCPs in the effluent. The detail assessment provides guidance for future LCA practice and methodology improvement, as well as for a better result interpretation.
- This work further compared toxicity related impacts by different LCIA methods comprising PPCPs which was not conducted in previous studies. The result from this methods comparison provides useful information for future LCA practice on the selection of LCIA methods regarding metals and PPCPs to suit with their necessities.

Chapter 5:

- The newly design work for nutrient removal and resource recovery from this study have the potential to guide future upgrading process in conventional wastewater treatment especially in the regions with diluted wastewater. This work identifies the importance to consider local wastewater characteristic and the technologies being used in the existing process when selecting technologies and designing process to upgrade WWTPs. The work benefits to the wastewater management as there is increased pressure to improve eutrophication impact in the water body and also for future sustainable strategies.
- It is reported for the first time for the upgrading of Malaysian STP by retrofitting the existing system to potential nutrient removal and resource recovery. By integrating upgrading design with life cycle assessment framework, this approach evaluates the whole system from a holistic perspective for additional nutrient removal and resource recovery process. Furthermore, the evaluation of life cycle assessment to nutrient removal only versus nutrient removal with P recovery has not been applied in the previous study. The result could guide the selection of appropriate technology in terms of additional/reduction of impact by different nutrient removal and P recovery provided.
- Finally, the significance of combining the design to upgrade and LCA assessment could; 1) provide a general direction for upgrading of existing wastewater treatment plant for nutrient removal and P recovery; 2) provide details inventory and newly design data in Malaysian wastewater treatment for guidance in future LCA practice; 3) evaluate both environmental benefits and economic burden of upgrading plants with different technologies for decision makers.

6.2 General conclusions

- The research in Chapter 3 concludes that the consideration of seasonal study in LCA to WWTP determines the difference of environmental impact between dry and wet season, regardless of the separate or combined sewer system. The results also demonstrate that rainfall effects on the environmental impact of WWTPs are more effective in MWTW with higher wastewater strength. The contrasting results of environmental impacts in MWTW during wet and dry seasons by using two different functional units suggest that the selection of functional unit is dependent on the comparison purpose such as the impact of WWTPs effluent to the environment only, or the combined effects from effluent and WWTP treatment efficiency. This work identified the importance of wastewater strength and functional units to the studies of rainfall effects on the environmental profile of WWTPs, which could serve as a basis for further rainfall studies with different coefficients between rainfall intensity and inflow rate, advanced treatment and others.
- In Chapter 4, this study demonstrates that LCA must include toxic pollutants in the data inventory especially metals which is dominant to the toxicity impact. The comparison of concentrations and removal efficiencies of PPCPs and metals in developing and developed countries revealed no apparent patterns due to no pattern of toxic pollutant concentrations in high and low strength wastewater. This result highlights the significance of obtaining data using site sampling instead of from published literature to represent the real effects of heavy metals and PPCPs on humans and the environment. The contribution of PPCPs in the effluent to FEP by the CML-IA method is low (i.e., 11%) suggesting that the negative impact of heavy metals on water bodies warrants more concern. However, only 10 PPCPs were investigated in this study. If more PPCPs were included, the contributions from PPCPs might increase. The quantitative information from this study can provide guidance on considering toxic pollutants in life cycle inventory, especially in regions with diluted wastewater for the reliable result in toxicity impacts. In addition, model comparisons between different available LCIA methods are critical to compare when including toxic pollutants to select the best method.
- In Chapter 5, three new processes were designed to remove nutrients and recover phosphorus based on the characteristics of local wastewater and the existing technology being used. This study undertakes a comprehensive assessment of environmental impacts and economic cost which could guide future LCA practitioner in the assessment of new upgrading and integrated technology. The total electricity consumption per day in the existing process is 23 - 26% lower than those in the upgraded processes for nutrient removal and P recovery. Meanwhile, electricity consumed for P recovery is negligible and average 65% of electricity is

still consumed for aeration. For functional unit, FU2 (per kg struvite recovered) is more suitable when considering the environmental impact from per kg P recovered from wastewater. FU1 (per m^3 wastewater) is more preferred to evaluate environmental performance for treating per m^3 wastewater. The overall results suggest that process A (i.e. the integration of EBPR and nitrification-denitrification for nutrient removal, Airprex for P recovery, and electricity recovery) is the optimum option when low financial impact are considered. In terms of environmental and technical benefits, process C is the best option. This work identifies the importance to consider local wastewater characteristics and the technology being used in the existing process when selecting technology and designing process to upgrade WWTPs. In addition, technological, economic and environmental assessment is critical to compare different processes to get the best option. The quantitative information from this study could provide guidance in decision making on upgrading the existing WWTPs especially in regions with diluted wastewater, which will underpin the transition towards a sustainable wastewater treatment.

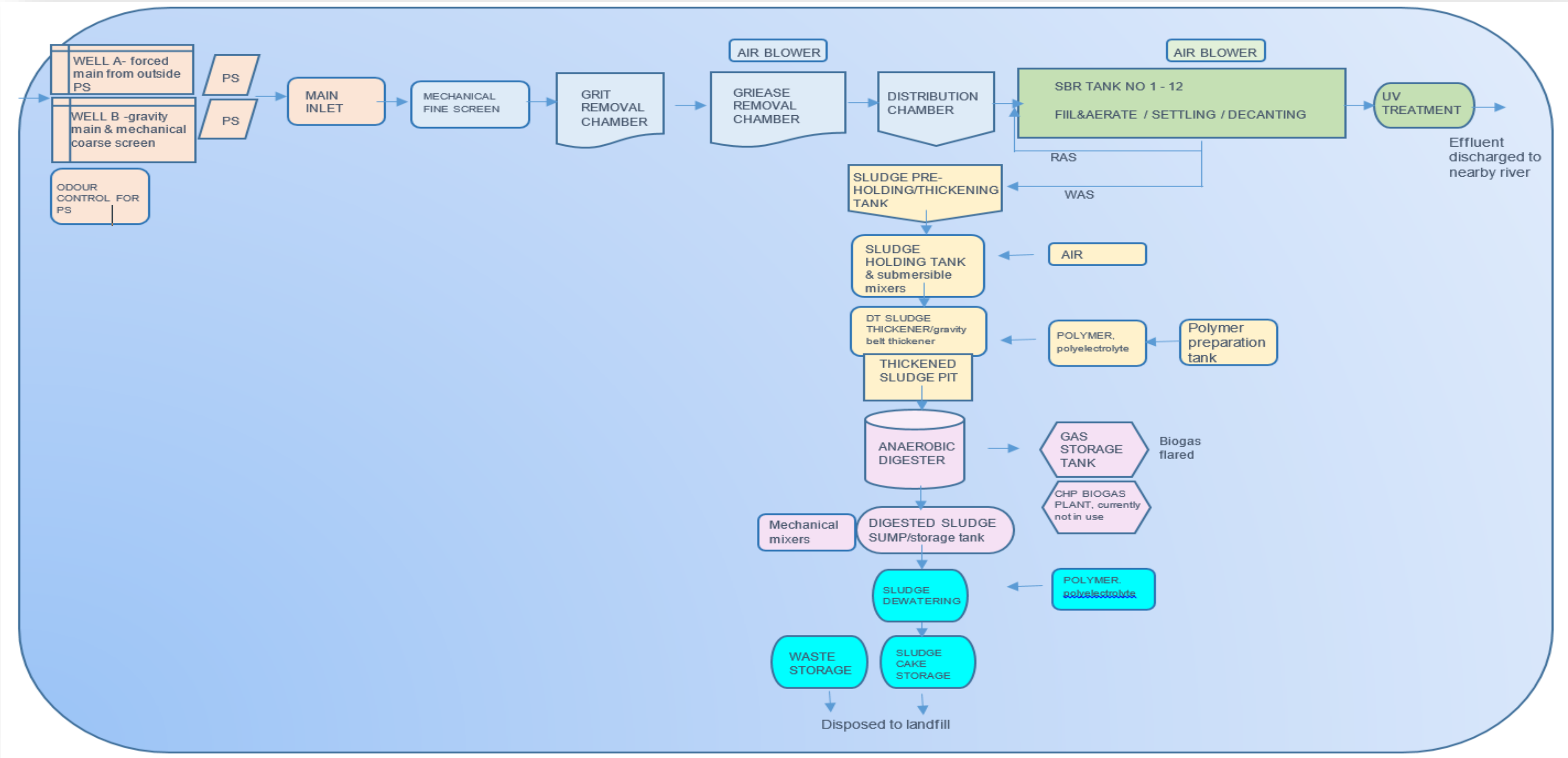
Overall, this thesis specifically captured comprehensive local conditions and databases for the modelling of the environmental and economic impacts of WWTP. This work also identifies critical factors related to wastewater operation that affects results in various scenarios, while proposing improvement options towards lower impact and sustainable operation. The result in this research will benefit the wastewater management practitioner and guide LCA users to abridge the gaps in the methodology limitation for future application of LCA-WWTP.

6.3 Recommendations for future research

The outcomes of this thesis provide the basis for further investigations into a variety of research topics, which are discussed below.

- In the seasonal effect of WWTP, it is suggested for future research to assess two different WWTPs in the same region but with different strength of wastewater characteristic (e.g. municipal WWTP with low strength wastewater and industrial WWTP with high strength wastewater) to obtain more comparable result based on regional impact especially to eutrophication.
- In the toxicity study, besides metals and PPCPs, it is suggested to include other priority pollutants such as pathogens from both in effluent and sludge, with more number of local pollutants considered. It is to further compare the contribution/composition of each category of pollutants in toxicity impacts for guidance to future regulations. In addition, LCA assessment from the industrial or hospitals WWTPs with high strength wastewater could be conducted and compared to municipal WWTPs to identify the difference in terms of the occurrence of toxic pollutants and their contribution to the environment.
- For the upgrading of WWTP, further research could investigate the integration of more advanced technologies such as Anammox or A-B process (which is more suitable with higher strength wastewater) to further compare if advanced treatment could achieve better impact in terms of environmental and economic. Furthermore, besides the electricity and P recovery potential, another recovery potential such as thermal energy from the incineration process of sludge is also interesting for the technology integration especially in a developed country situation in order to achieve net-zero impact from WWTP.

Appendix A Flow Diagram of Malaysian Sewage Treatment Plant



Appendix B Aerial photo of Malaysian Sewage Treatment Plant in Penang, Malaysia

(Source: www.sepakatsetiaperunding.com)

Appendix C The calculation of number of reactors required in 3 upgraded processes which is processes A, B and C

The number of reactors required is dependent on treatment capacity of each reactor. The treatment capacity of each reactor is determined by operation cycle. The treatment capacity of each reactor after upgrade was calculated using Eq.(5.2.3.1).

$C_1 = (C_0 \times N_1) / N_0$ $Eq.$(5.2.3.1)

Where C_0 is treatment capacity of each reactor before upgrade, N_0 is number of cycles in each reactor each day before upgrade, and N_1 is number of cycles in each reactor after upgrade.

Number of cycles each day was reduced with longer operation cycle. Thus, treatment capacity of each reactor was reduced, and number of reactors required in upgraded processes was calculated using Eq.(5-2).

$n_1 = (n_0 \times C_0) / C_1$ $Eq.$(5.2.3.2)

Where n_0 is number of reactors required before upgrade.

<u>Number of reactors required in process A is calculated using following steps:</u>

Step 1: calculate treatment capacity of 1 reactor in process A/B/C

For example : $C_A = (C_0 \times N_A) / N_0$

Step 2: calculate number of reactors required based on treatment capacity

$n_A = (n_0 \times C_0) / C_A$

Number of reactors in existing and three upgraded processes:

Process	Treatment capacity of 1 reactor (m^3/day)	Number of cycles each day	Number of reactors
Existing	37,237.62	6	4
Process A	24,825.08	4	6
Process B	24,825.08	4	6
Process C	29,790.10	4.8	5

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