SCHOOL OF COMPUTING, ENGINEERING AND BUILT ENVIRONMENT

DISSERTATION

ASSESSMENT GREENHOUSE GAS EMISSIONS COMING FROM WASTEWATER TREATMENT PLANTS

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ABSTRACT

Wastewater treatment plants play a vital role in protecting the water environment. WWTPs help remove pollutants in wastewater before being discharged into the environment, ensuring that the quality of wastewater meets certain wastewater quality standards and regulations. However, the wastewater treatment process of WWTPs generates greenhouse gases (GHGs) both, directly and indirectly, affect the air environment profoundly.

This report is made for the main purpose of "*Assessing the greenhouse gas emissions coming from wastewater treatment plants*" in the range of Scope 1 emissions: direct GHG emissions from WWTPs themselves. Through the study of documents for data collection and analysis, the report has basically drawn out a schematic diagram of direct GHG emissions from WWTPs. The report found that the GHG emissions are released in all steps of the wastewater treatment process. The number of GHG emissions generated in each treatment stage of WWTPs has been synthesized. In which, the pre-treatment phase generated 78.3 kg h^{-1} of CO_2, 1.8 kg h^{-1} of CH_4 and 1.025 kg h^{-1} of N_2O. These numbers for the primary treatment stage were 5.29 kg h^{-1}, 0.8 kg h^{-1}, and 0.1 kg h^{-1}, respectively. The secondary treatment phase released a huge amount of GHGs. For the AAO technology, the quantity of CO_2 emissions was about 3,046.92 kg h^{-1}, the number of CH_4 emissions was 8,812 kg h^{-1}, and the figure for N_2O emissions was 407.45 kg h^{-1}. For SBR technology, the emissions of CO_2, CH_4 and N_2O were 2,044.21 kg h^{-1}, 5,306 kg h^{-1} and 14,135 kg h^{-1}, respectively.

It can be seen that the amount of three main GHGs, namely CO_2, CH_4, and N_2O arising from WWTPs themselves are significant. However, the estimation of GHG emissions in previous studies often ignores CO_2 emissions as it is considered to be carbon neutral, which leads to the total amount of GHG emissions generated from WWTPs may be underestimated. The assessment of direct GHG generated directly from wastewater treatment plants is a huge challenge due to many influent factors, the accuracy and lack of data. In the UK, the water sector has not updated the GHG Conversion Factors since 2012, which may cause uncertainty of the GHGs estimation from WWTPs.

The water industry should consider and apply mitigation measures such as source controls, technological measures, GHG collection and treatment, energy recovery, and develop legal GHG-related plans and strategies for the water sector's activities.

The UK water industry has developed "*The UK net-zero 2030 routemap*" to help support drinking water treatment plants and WWTPs transition to a lower emissions future and enabling an acceleration and step change in decarbonisation. This has crucial implications for the responsibility of the water sector to the global problem as well as the direction of development activities of the water sector in the UK.

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LIST OF ABBREVIATIONS

AAO/A^2O	:	Anaerobic/Anoxic/Oxic
AD	:	anaerobic digestion
ADM1	:	Anaerobic Digestion Model No.1
AOB	:	Ammonia-oxidizing bacteria
ASM1	:	Activated Sludge Models No.1
BNR	:	Biological Nitrogen Removal
BOD	:	Biological oxygen demand
BSM1	:	Benchmark Simulation Model No. 1
BSM2	:	Benchmark Simulation Model No. 2
CDM	:	Clean Development Mechanism
CH_4	:	Methane
COD	:	Chemical oxygen demand
CO_2	:	Carbon dioxide
DO	:	Dissolved oxygen
DWTP	:	Drinking water treatment plant
EFs	:	Emission factors
EPA	:	United Stated Environmental Protection Agency
GHG	:	Greenhouse gas
GWP	:	Global Warming Potential
IDMM	:	Inverse Dispersion Modelling Method
IPCC	:	Intergovernmental Panel on Climate Change
MBBR	:	Moving Bed Biofilm Reactor
MBR	:	Membrane Bio Reactor
MLE	:	Modified Ludzack and Ettinger
MTDM	:	Mobile Tracer gas Dispersion Method
NASA	:	The National Aeronautics and Space Administration
NOAA	:	The National Oceanic and Atmospheric Administration
N_2O	:	Nitrous dioxide
OSPM	:	On-site point measurements
PAC	:	Poly-Aluminium Chloride
PE	:	population equivalent

ppm	:	parts per million
ppb	:	part per billion
SBR	:	Sequencing Batch Reactor
STDM	:	Static Tracer Gas Dispersion Method
TKN	:	Total Kjeldahl Nitrogen
TN	:	Total Nitrogen
TSS	:	Total Suspended Solids
UK	:	United Kingdom
USA	:	United State of America
USEPA	:	United States Environmental Protection Agency
VOCs	:	Volatile Organic Compounds
VSS	:	Volatile Suspended Solids
WEF	:	Water Environment Federation
WWTP	:	Wastewater treatment plant

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CHAPTER 1: INTRODUCTION

1.1. Background

In recent years, climate change has become a worldwide concern and discussion topic. Due to the increased concerns in global warming, human awareness of the greenhouse gas (GHG) emissions effects has expanded globally. Climate change is a natural phenomenon, however, because of human economic development activities, which are releasing GHGs into the atmosphere, it makes the Earth's environmental components constantly change. Global atmospheric carbon dioxide (CO_2) was 409.8 ± 0.1 ppm in 2019, a new record high. That is an increase of 2.5 ± 0.1 ppm from 2018 (Lindsey, 2020). According to the recorded figures by the National Oceanic and Atmospheric Administration (NOAA), the global methane (CH_4) concentrations rose from 1875.4 ppb in October 2019 to 1890.9 ppb by October 2020. Nitrous dioxide (N_2O) also witnessed an average uptrend of about 1.3 ppb.

All the sectors including the water sector are somehow contributing to an increase in the number of GHGs releasing into the air. The water industry has several activities including the extraction, transportation, and treatment of raw water for a variety of purposes, wastewater collection, treatment, and discharge. These activities throughout the whole life cycle of water and wastewater treatment plants are directly and indirectly emitting GHGs such as CO_2, CH_4, and N_2O. In the UK, the water industry contributes 0.8 per cent of annual UK greenhouse gas emissions (Reffold et al, 2008).

Numerous researches have been carried out in the water industry and those achieved data are adequate to prove that wastewater treatment plants (WWTPs) are a potential source of anthropogenic GHG emissions. The operation of WWTPs directly releases GHGs through biological processes as well as indirectly from the production and use of energy and chemicals. Each WWTP does not generate the same amount of GHGs because it depends on many different factors such as capacity, treatment technology, the type of wastewater treated (domestic or industry), energy, and chemical uses.

In 2006, the Intergovernmental Panel on Climate Change (IPCC) developed guidelines for the National Greenhouse Gas Inventory (IPCC, 2006). There were five volumes, in which Chapter 6 of volume 5 guides how to calculate the GHG emissions from

Wastewater Treatment and Discharge. To restrain GHG emissions into the atmosphere, the United Nations encourages countries to annually report on the amount of anthropogenic GHG emissions. Although CO_2 is also one of the gases released from WWTPs, it is not considered a GHG due to their biogenic origin (IPCC, 2006). Only CH_4 and N_2O emitted from WWTPs are guided. Besides that, some models have also been set up to calculate greenhouse gases coming from WWTPs such as Anaerobic Digestion Model No.1 (ADM1); Benchmark Simulation Model No. 1 (BSM1); Activated Sludge Models No.1 (ASM1); and Bridle model.

The water sector, the fourth most energy-intensive industry in the UK, is a major national contributor to emissions responsible for around five million tonnes each year (WWTonline, 2020). Obviously, it is imperative to take action to minimize the amount of GHG emissions from WWTPs. In the UK, the water sector has launched the world's first sector-wide plan to deliver net zero carbon emissions by 2030. In order to achieve that plan, the water sector should understand key sources of emissions, how much GHG they emit, and then plan details what they should do to reduce GHG emissions.

This report will define the scope of the study based on three emission scopes defined by the United Nations through the GHG Protocol. The report will then critically review previous studies to assess the GHG emissions generated by WWTPs, and present popular quantitative GHG emission methods from WWTPs in the methodology chapter. At the same time, approaches to reduce GHG emissions from WWTPs will also be recommended.

1.2. Research boundaries

To assess the amount of GHG emissions generated by industry, the United Nations has defined 3 Scopes through GHG Protocol in order that the government and business leaders would understand, quantify and measure the GHGs. According to the Greenhouse Gas Protocol Initiative, three scopes are as follows:

Scope 1 emissions: Direct emissions from owned or controlled sources.

Scope 2 emissions: Indirect GHG emissions from consumption of purchased electricity, heat or steam.

Scope 3 emissions: Other indirect emissions (not included in Scope 2), such as the extraction and production of purchased materials and fuels, transport-related activities in vehicles not owned or controlled by the reporting entity, electricity-related activities not covered in Scope 2, outsourced activities, waste disposal.

The boundaries in this research are determined from the scope 1. The research will evaluate the emissions generated from the operation of the plant itself through the wastewater treatment processes.

1.3. Research Aim and Specific Objectives

The overall aim of this study is to assess the direct GHG emissions releasing from WWTPs in order to contribute to develop and implement the net-zero carbon emissions of the water sector in the UK as well as proposing some mitigation measures.

Research objectives:

1. To critically review previous studies on GHG emissions of the WWTPs;
2. To assess the direct GHG emissions generated by the WWTPs;
3. To analyse the environmental impacts of the WWTPs;
4. To propose solutions to reduce the direct GHG emissions coming from the WWTPs.

Research questions: The questions that relate to the research objectives are identified as follows:

1. *Where do greenhouse gas emissions come from in WWTPs?*

   The answer to this question will support the achievement of Objective 2. The report will select common wastewater treatment processes to assess the amount of GHG emissions.

2. *What are the possible consequences and impact of these emissions?*

   Answering this question will help to gain Objective 3. The report will be based on the results obtained from question 1 to analyse the environmental impacts of WWTPs.

3. *What must we do to reduce greenhouse gas emissions from the water industry sector in order to reach net-zero carbon emissions?*

The answer to this question will assist in achieving Objective 4 by proposing some viable solutions to reduce GHG emissions from WWTPs.

1.4. The importance of research

Like any industry, the water industry sector needs to take actions to reduce its GHG emissions into the atmosphere. In order to take appropriate measures, they must understand their contributions to the release of GHGs.

Although WWTPs treat wastewater aiming at environmental protection, they also generate directly and indirectly a relatively large amount of GHGs during the treatment process and consuming energy and chemicals.

Understanding responsibilities in preventing climate change and cutting GHG emissions, the water industry in the UK is developing a plan towards net-zero carbon emissions. By implementing this report, it will make a certain contribution to identifying sources of GHG generation from the operation of WWTPs and clarifying its impacts on the environment, and at the same time, gradually prepare for a road map and implementation of the net-zero carbon emissions within the water industry sectors.

1.5. Dissertation structure

Chapter 1: Introduction

Chapter 1 of the study demonstrates the motivation to conduct research, defines the scope of the research, the objectives, and the importance of the study. Besides, it also provides an overview of the whole report, implementation plan, and presentation structure.

Chapter 2: A literature review

This chapter critically reviews the relevant studies that have been performed, the findings of previous studies, and the contributions of those studies. The similarly previous studies and findings that have been published are important as they help define and confirm the results of this study.

Chapter 3: Research Methodology

This chapter will illustrate how the study is to be conducted and common methods for estimating GHG emissions arising from the operation of WWTPs themselves. It also includes methods' limitations and how to mitigate them.

Chapter 4: Critical analysis and discussion

This chapter analyses critically the review study and also discusses similarities and differences from previous studies, the validity of the findings, and the uncertainties. Additionally, it assesses the potential impacts of GHG emissions arising from WWTPs as well as recommending some mitigation measures to cut down GHG emissions.

Chapter 5. Conclusion and Future Work Recommendations

Chapter 5 concludes what is found and what it contributes to further research and on the road to net zero carbon emissions, recommendation on future work direction.

CHAPTER 2: LITERATURE REVIEW

2.1. Overview

Both domestic or industrial wastewater must be treated up to certain standards before being discharged into the environment. Wastewater is treated through many various stages in WWTPs, which require different operating conditions and generate different amounts of GHG emissions. GHG emissions from these WWTPs are fugitive, as they are unintended discharges escaping from process units, and they are diffusive because they occur from sources scattered throughout the facility (Delre, 2018).

Calculating these types of emissions from wastewater treatment plants is a huge challenge since facilities with different plant layouts, and using different process units and treatment technologies. Therefore, it could require alternative ways of applying the method. Operations at a WWTP involve direct and indirect GHG emissions. Direct GHG emissions are fugitives discharged directly into the atmosphere, whereas indirect GHG emissions are caused by the consumption of chemicals and energy (Yoshida et al., 2014a cited in Delre, 2018). During the operation, these plants directly release GHGs such as CO_2 from the bio-treatment process, N_2O from nitrification and denitrification processes, and CH_4 from sludge treatment. In addition, WWTPs indirectly emit GHGs, mainly CO_2, due to the consumption of chemicals and energy (Delre, 2018). *Presura and Robescu, 2017* calculated the number of CO_2 emitted from DWTP and WWTP based on energy consumption. They concluded that energy used was the main source of CO_2 generation in the water industry. Previous studies, by many different methods, have quantified the number of GHGs generated both directly and indirectly from WWTPs.

In Chapter 2, the report will critically review the published studies that have been done related to the research. Reviewing similar studies will help formulate and validate the study's findings.

2.2. Wastewater treatment processes

Wastewater is generated from anthropogenic domestic and industrial activities. Bacteria, chemicals, and toxic substances enter the wastewater stream and escape into the environment. Therefore, domestic and industrial wastewater must be collected and

treated to reduce contaminants to acceptable levels according to wastewater quality standards and regulations to ensure the safe before discharging into the environment.

The choice of wastewater treatment technology processes is mainly driven by the quality standards and regulations for each type of wastewater (domestic or industrial). There are two main sorts of WWTPs: biological wastewater treatment plants and chemical or physical wastewater treatment plants, which correspond to two different types of domestic and industrial wastewater. Biological WWTPs use biological matter and bacteria to break down waste matter. Alternatively, physical WWTPs use chemical reactions as well as physical processes to treat wastewater. While biological treatment systems are ideal for treating wastewater from households and business premises, physical wastewater treatment plants are mostly used to treat wastewater from industries, factories, and manufacturing firms (Rinkesh, 2020). This study will only focus on domestic wastewater treatment processes.

Nowadays, wastewater treatment technologies are constantly being improved. Depending on many different factors such as construction area, cost, and characteristics of wastewater, each WWTP chooses suitable technology for its wastewater treatment. The technologies that are commonly applied include Moving Bed Biofilm Reactor (MBBR), Anaerobic/Anoxi/Oxic (A2O or AAO), Membrane Bio-Reactor (MBR), and Sequencing batch reactor (SBR). Each type of treatment technology generates different amounts of GHG emissions.

Normally, a domestic wastewater treatment process could be broken down into five basic stages: preliminary treatment, primary treatment (sedimentation), secondary treatment (biological), tertiary treatment (final discharge quality), and sludge treatment. For sludge treatment, it could break into a separate treatment line with water lines. The general layout of a wastewater treatment plant is shown in Figure 1.

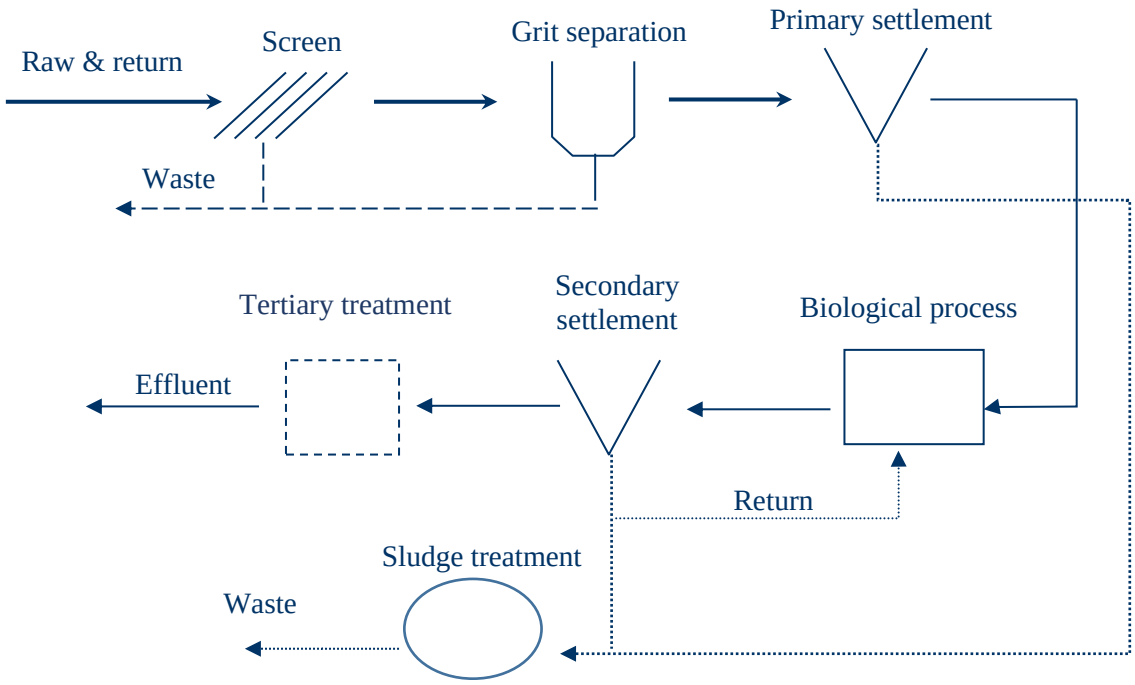

Figure 1: General layout of a WWTP

(George et al., 2003)

❖ **Preliminary treatment**

Regardless of the choice of any treatment technology, the first step of a wastewater treatment technology chain is a pre-treatment step to remove the gross solids, grit, stormwater, and oil by using a screen and grit chamber. The raw waste will be removed from the water stream and treated.

❖ **Primary treatment**

After preliminary treatment, wastewater is moved to the following step of primary treatment (sedimentation). This step refers to the sedimentation tank or conditioning tank or balancing tank. The large suspended solids in sewage are removed by sedimentation. The main goal of this step is to separate the organic matter and sludge from the rest of the water.

❖ **Secondary treatment**

Treatment processes can be different in this steps. This study will not cover all treatment technologies but select AAO and SBR technologies to focus on. These are two popular technologies that are widely applied in numerous domestic WWTPs.

The secondary treatment (biological treatment) step is often applied to remove dissolved and colloidal organics by biological methods. In this step, the technology selected for

wastewater treatment can be either A2O or SBR technology. The A2O technology is the combination of Anaerobic/Anoxic/Oxic processes, by using microorganisms to decompose pollutants in wastewater, while SBR is a technology that uses a separate treatment section with five phases including: fill, react, settle, decant, and idle.

- ***A2O (or AAO) technology***

+ *Anaerobic process:*

In the anaerobic tank, anaerobic microorganisms do not require oxygen to break down organic matter forming CH_4, CO_2, and excess biomass. The anaerobic processes can be divided into three main stages:

• *Hydrolysis:* enzyme-mediated transformation of complex organic compounds (polysaccharides, protein, lipid) into simple compounds (sugars, amino acids, fatty acids).

• *Acidogenesis:* anaerobic bacteria convert simple compounds into substrates for methanogenesis (acetate, formate, hydrogen, carbon dioxide).

• *Methanogenesis:* methanogenic substrates continually are converted into methane and carbon dioxide.

The anaerobic digestion process can be simplified with the following biochemical reactions:

i. Organic matter + anaerobic bacteria → $CO_2 + H_2S + CH_4$ + other substances + energy

ii. Organic matter + anaerobic bacteria + energy → $C_5H_7O_2N$ (new cells)

The resulting gas mixture is often referred to as biogas, can be captured for the recovery of energy.

+ *Anoxic process:*

Anoxic process is typically used to remove nitrogen from wastewater, which is commonly known as denitrification. Denitrification can occur in biological systems that are:

• *Anabolism:* The process of conversion nitrate into ammonia (NH_4^+) used for cell synthesis. It occurs when ammonia is not available and independent of the inhibition of oxygen.

• *Catabolism (or denitrification):* this involves transforming nitrate into nitrite, nitrous oxide, and nitrogen gas.

$$NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$$

+ Oxic process:

The oxic process is based on the living activities of aerobic microorganisms which use dissolved oxygen to decompose organic matter (pollutants need to be treated). Microorganisms convert NO_3^- to N_2 and release into the air.

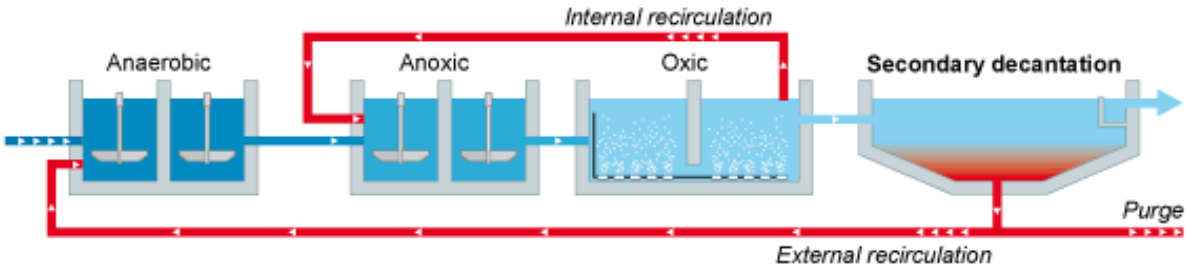

Figure 2: AAO process

(Source: Membrane Solutions LLC, 2017)

- ***SBR technology***

SBR technology is a technology to treat domestic wastewater by the sequencing batch reactor. This can be considered as an aerotank. The SBR process includes 5 different steps: Fill, React, Settle, Decant and Idle, as shown in Figure 3.

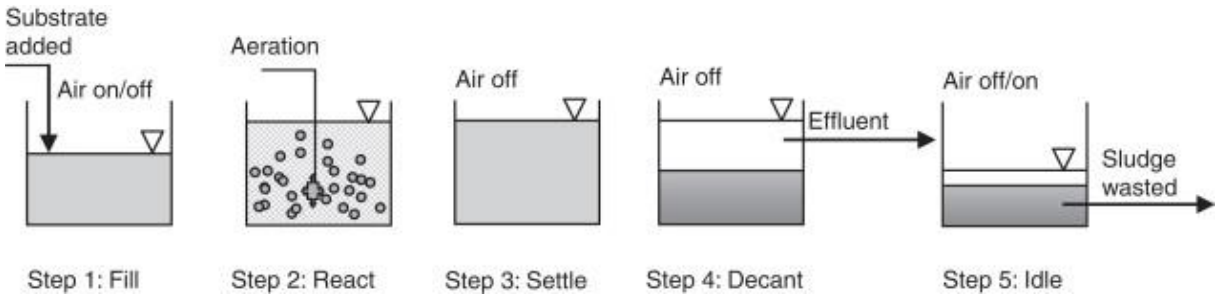

Figure 3: SBR process

(Source: Ghangrekar and Behera, 2014)

The wastewater is filled in the “Fill” step. Depending on operation strategy, there are three different variations that can be used one or all of them in this step, namely static fill, mixed fill, and aerated fill.

"Static fill" is characterized by no mixing or aeration (EPA, 1999), and biomass is added in the influent wastewater, whereas, "mixed fill" means that organic matters are mixed with added biomass so that denitrification reaction occurs under anoxic conditions. Anaerobic conditions can also be achieved during the mixed fill phase (EPA, 1999).

The "aerated fill" occurs by aerating the contents of the reactor to create the environment for aerobic reactions to take place in the react step.

In the "react" step, biological reactions are performed and completed in this step. Ammonia-nitrogen is converted to nitrite-nitrogen and ultimately to nitrate-nitrogen. Anaerobic conditions can also be achieved during this phase. Depending on what types of wastewater, reaction under controlled conditions: anaerobic, anoxic or aerobic could happen.

"Settle" is usually conducted under quiescent conditions, meaning no inflow/outflow conditions, in order to settle the active sludge. "Draw" or "Decant" step uses a decanter to remove the clear supernatant effluent (EPA, 1999). "Idle" step is classified as sludge wasting (Dohare and Bochare,2014).

- **Tertiary treatment**

Tertiary treatment is the final treatment step to remove pathogenic microorganisms and improve the quality of wastewater before it is reused or discharged into the environment. In this step, chemicals such as Chlorine, sodium hypochlorite (NaOCl), sodium hydroxide (NaOH), ozone, Poly-Aluminium Chloride (PAC) can be used to remove the inorganic compounds, and substances.

- **Sludge treatment**

Sludge can be considered as a by-product of the wastewater treatment process. Sludge is collected to the sludge storage tank and then can be treated by different methods to remove water. Popular sludge treatment methods include a combination of thickening, digestion and dewatering; thermal hydrolysis with biological treatment; sludge reuse and disposal, and incineration, to name but a few.

2.3. Overview of direct GHG emissions from wastewater treatment plants

2.3.1. Carbon dioxide emissions

Carbon dioxide (CO_2) is an important heat-trapping (greenhouse) gas, which is released through human activities as well as natural processes (NASA, 2020). During the wastewater treatment process, CO_2 is one of the three main GHG emissions released into the atmosphere. Research by *Campos et al, 2016*, has shown that the WWTPs produce carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) during the biological wastewater treatment process and CO_2 is also emitted during the production of the energy required for the plant operation. In the USA, the United States

Environmental Protection Agency (USEPA) and Water Environment Federation (WEF) database suggested that methanol CO_2 was likely to represent 1/4 of all Scope-1 emissions from wastewater treatment (Koh and Shaw, 2018). *Willis, Al-Omari et al. 2017* as cited in *Koh and Shaw, 2018,* presented a case study of the Blue Plains AWTP in DC, USA where methanol CO_2 amounted to 60 to 85% of the Scope-1 emissions.

Regarding the direct CO_2 emission from WWTPs, during anaerobic treatment, the BOD_5 in the wastewater is either converted to CO_2 and CH_4 by endogenous respiration. Other sources of CO_2 emissions directly coming from WWTPs are caused by sludge digesters and digestion gas combustion. In the aerobic process, CO_2 is produced by the decomposition of organic substances (Taseli, 2019).

There are only a few studies that research on direct CO_2 emission from WWTPs including research of *Yan et al., 2014, Bao et al., 2015 and 2016, and Kyung et al., 2015*. Research by *Yan et al., 2014* and *Bao et al., 2015* both quantified CO_2 emissions from each unit in the AAO treatment process. The results of the two studies are summarized by *Nguyen et al, 2019* as shown in Table 1.

Table 1: CO_2 emissions from each unit in the AAO treatment process

Treatment unit	Yan et al., 2014		Bao et al., 2015	
	Area (m^2)	***CO_2 emissions (kg/day)***	***Area (m^2)***	***CO_2 emissions (kg/day)***
Aerated grit tank	346	78	504	1,879.13
Anoxic tank	-	172	3564	215.66
Anaerobic tank	-	70	3564	242.00
Oxic tank	15,051	24,637	25,011	72,651.20

(Source: Nguyen et al., 2019)

In both studies, the number of CO_2 in the oxic tank is the highest which corresponds to the largest area. When considering the rate between the number of emissions and the areas of tanks, the results of *Bao et al., 2015* showed that the aerated grit tank had the highest emission rate, while the highest rate of emission flux was from the oxic tank according to the results of *Yan et al., 2014*. However, the previous studies all indicated that the aerobic area emitted significant volumes of CO_2 (Nguyen et al., 2019).

A similar trend was also found in the SBR treatment process. The amount of carbon dioxide released from feeding and aeration period was an average of 334.6 g CO_2eq m^{-3} wastewater (Bao et al., 2015 cited in Nguyen et al, 2019) and 343.86 g CO_2eq m^{-3} wastewater (Bao et al., 2016 cited in Nguyen et al, 2019), which amounted to 99% of the total emissions. At the beginning of the feeding and aeration stages of the SBR treatment process, the CO_2 is mostly released in these two phases.

Although WWTPs directly emit CO_2, the direct CO_2 emissions originate from the microbial respiration of organic matter in the aeration tanks which is short-cycle carbon and therefore it does not contribute to the increased carbon dioxide concentration in the atmosphere (Daelman, 2014). The IPCC guidelines also say explicitly that carbon dioxide emissions are not included in the panel's tabulations for WWTP emissions because they are thought to be derived from natural biological sources and are carbon neutral (Magill, 2016). As a result, there was not much research on the direct CO_2 emissions from WWTPs.

2.3.2. Methane emissions

Methane (CH_4) is the second most important greenhouse gas in terms of concentration and impact on the climate (Marmier and Schosger, 2020). Methane, having a global warming potential of 25 CO_2-equivalents over a 100-year time horizon, is expected to be formed in the sewer system (Guisasola et al., 2008) and in those parts of the WWTP where anaerobic conditions prevail (Daelman et al, 2012). In 2013, IPCC had announced for CH_4 a new GWP factor of 34 CO_2-equivalents over a 100-year time horizon. It can be seen that CH_4 is a major contributor to global warming.

CH_4 is usually generated in anaerobic conditions such as in sewer systems. Also, CH_4 is dissolved in water and enters into WWTPs. Sources of CH_4 emissions can be found in both water and sludge process lines in WWTP (Noyola et al, 2018). In the water line, CH_4 can be formed in the primary sedimentation and then stripped out with the aeration air in activated sludge tanks (Parravicini et al., 2016). The sludge anaerobic digestion in the wastewater treatment process generates a large amount of CH_4 (around 72% according to *Campos et al, 2016*; and 81% according to Samuelsson et al, 2017), which could be released into the air if it is not trapped to produce biogas. Therefore, in the guidelines of IPCC, 2006, for National GHG Inventories from Wastewater Treatment and Discharge, CH_4 and N_2O are the two GHGs mentioned.

Direct CH_4 emissions is higher than N_2O emissions in most wastewater treatment facilities, in terms of CO_2 equivalent (Zhan et al, 2018). *El-Fadel and Massoud, 2001 and Conrad, 2009*, (as cited in Daelman, 2014), found that wastewater treatment can attribute about 4-5% of the global anthropogenic CH_4 emissions. However, CH_4 has received very little research interest compared to N_2O.

There are a few prominent case studies on CH_4 emissions from municipal WWTPs, including the study of *Czepiel et al* (1993) in Durham, NH, USA; *Wang et al.* (2011) in Jinan, China; *Daelman et al.* (2012) in Netherlands, *Rena et al* (2013) in China; *Liu et al.* (2014) in Beijing, China; *Oshita et al*, (2014) in Japan; and *Kwok et al*, (2015) in France, to name but a few.

Czepiel et al. 1993, researched on a small WWTP with 4 x 10^3 m^3/day (12,500 PE). They found that the CH_4 emissions were 39 g CH_4/(capita.year), 0.140 m^3 CH_4/m^3 influent, or 0.16% of the incoming COD (Xinmin Zhan et al, 2018). For two research in WWTPs in China, *Wang et al, 2011*, studied a WWTP with the capacity of 3 x 10^5 m^3/day (1,500,000 PE, A^2O process), while *Liu et al, 2014*, conducted research at two different WWTPs with the capacity of 5 x 10^5 m^3/day (1,200,000 PE) with A^2O process, and 8 x 10^4 m^3/day (231,000 PE) with sequencing batch reactor (SBR) process, respectively. These studies applied grab sampling techniques to monitor direct off-gas emissions in the WWTPs. Compared with the findings by *Czepiel et al*, *1993*, the amounts of CH_4 gas generated from WWTPs in China were comparatively lower despite the larger treatment capacity. CH_4 emission factors found by *Wang et al, 2011*, were 11 g CH_4/(capita.year), 0.16 m^3 CH_4/m^3 influent, or 0.16% of the incoming COD, respectively. In the investigation of *Liu et al., 2014*, the emission factors in the A^2O process were 0.182 g CH_4/m^3 influent and 24.8 g CH_4/(capita.year), whereas the amount of CH_4 emissions for SBR process were 0.457 g CH_4/m^3 influent and 36.5 g CH_4/(capita.year) (Zhan et al, 2018).

Daelman et al., 2012, studied the WWTP with a 360,000 PE in the Netherlands which included an anaerobic digestion facility for surplus sludge, and the CH_4 emissions from this step were also considered. This had a profound influence on the research results, specifically, the amount of CH_4 emissions generated were 306 g CH_4/(person.year), or 3.44 g CH_4/m^3 influent. *Oshita et al*, 2014, had monitored two full-scale WWTPs with anaerobic digestion of sludge in Japan for over 1-year period and found that average

emissions of CH_4 was 509 ± 72 mg/m^3-influent (wastewater). Studies with WWTPs that apply anaerobic digestion of sludge process suggested that CH_4 continues to be emitted from digested sludge after leaving the anaerobic digester (Oshita et al, 2014).

In 2015, *Kwok et al*, conducted an CH_4 emission estimation using chamber and tracer release experiments for a municipal WWTP in Valence, France. They confirmed that the open basins were not a major source of CH_4 on the WWTP (about 10% of the total emissions), but the pretreatment and sludge treatment plant were the main emitters.

Delre, 2018, has synthesized previous studies on CH_4 emission factors (EFs) measured at WWTPs, expressed in percentages of kg CH_4 (per kg COD in the influent) and (per kg CH_4 in the production) in Table 2 and Table 3. In the previous studies, the research of *Yoshida et al, 2014* in the whole plant by Mobile Tracer Gas Dispersion Method (MTDM) showed the highest value of CH_4 emissions, 9.08 kg CH_4 (per kg COD in the influent) and 32.7 kg CH_4 (per kg CH_4 in the production), respectively.

Table 2: CH_4 emission factors (EFs) measured at WWTPs, expressed in percentage as kg CH_4 (per kg COD in the influent)

Value (min-max)	Source	Method	References
0.29	Wastewater Reactors	OSPM	Czepiel et al. (1993)
0.53 - 1.2	Wastewater Reactors	OSPM	STOWA (2010)
0.034	Wastewater Reactors	OSPM	Toyoda et al. (2010)
0.06 - 0.1	Wastewater Reactors	OSPM	Wang et al. (2011)
1.13	Whole plant	OSPM	Daelman et al (2012)
0.04 – 0.1	Wastewater Reactors	OSPM	Aboobakar et al. (2013b)
0.25	Wastewater Reactors	OSPM	Gustavsson, Tumlin (2013)
0.046 – 1.33	Wastewater Reactors	OSPM	Ren et al. (2013)
0.016	Wastewater Reactors	OSPM	Rodriguez-Caballero et al. (2014)
0.16 – 0.6	Sludge treatment units	OSPM	Oshita et al. (2014)
0.61 – 4.35	Whole plant	MTDM	Yoshida et al. (2014b)
9.08[a]	Whole plant	MTDM	Yoshida et al. (2014b)
0.15 – 0.69	Wastewater Reactors	OSPM	Masuda et al. (2015)

Note: [a] Value measured during digester malfunctioning. OSPM: on-site point measurements. MTDM: Mobile tracer gas dispersion method.

Table 3: CH_4 emission factors (EFs) expressed in percentage as kg CH_4 (per kg CH_4 in the production)

Value (min-max)	Facility	Source	Method	References
0.8 – 3.2	Biogas plant	Whole plant	IDMM	Flesch et al. (2011)
26.6 [a]	Biogas plant	Whole plant	IDMM	Flesch et al. (2011)
2.8	Biogas plant	Digester	NA	CDM (2012)
5	Biogas plant	Digester	NA	CDM (2012)
10	Biogas plant	Digester	NA	CDM (2012)
0.15 -2.6	WWTP	Sludge treatment units	OSPM	Petersson (2012)
0.3 – 5.25	Biogas plant	Whole plant	OSPM	Petersson (2012)
1.1 – 13.7	Biogas plant	Whole plant	OSPM	Liebetrau et al. (2013)
2.1 – 4.4	WWTP	Whole plant	MTDM	Yoshida et al. (2014b)
32.7 [a]	WWTP	Whole plant	MTDM	Yoshida et al. (2014b)
4	Biogas plant	Whole plant	IDMM	Groth et al. (2015)
1.3	Biogas plant	Digester	OSPM	Thomsen (2016)

Note: [a] Value measured during digester malfunctioning. OSPM: on-site point measurements. MTDM: Mobile tracer gas dispersion method. IDMM: inverse dispersion modelling method. NA: Not available. CDM: Clean development mechanism EFs according to the type of digester.

From the above studies, it can be seen that the different treatment technologies emit differing amounts of GHG emissions. *Rena et al, 2013*, had demonstrated this by conducting research on N_2O and CH_4 emissions from different treatment processes in full-scale municipal WWTPs. Three typical biological wastewater treatment processes were studied in WWTP of Northern China: pre-anaerobic carrousel oxidation ditch (A+OD) process, pre-anoxic anaerobic-anoxic-oxic (A-A/A/O) process and reverse anaerobic-anoxic-oxic (r-A/A/O) process. The authors observed that the CH_4 conversion ratio of r-A/A/O process was the lowest among the three WWTP, which were 89.1% and 80.8% lower than that of A-A/A/O process and A+OD process, respectively.

In addition to the application of monitoring and sampling methods, plant-wide models can also apply to quantify the number of GHG emissions resulting from WWTPs. Although CH_4 and CO_2 are the 2 gases considered in the first models. However, later models mainly focus on N_2O quantification. For CH_4 gas, the Anaerobic Digestion Model 1 (ADM1) model proposed by *Batstone et al. (2002)* described CH_4 and CO_2 emissions under

anaerobic conditions (Corominas et al, 2012). *Laura Snip, 2009,* had applied the Bridle model and Benchmark Simulation Model 2 (BSM2) model to quantify GHG emissions from WWTPs. The author found that the mean values of the Bridle model and the BSM2 model were 1085.8 kg CH_4/day and 1059.5 kg CH_4/day, respectively.

2.3.3. Nitrous dioxide emissions

N_2O is a greenhouse gas which remains in the atmosphere for more than 100 years, on average. According to IPCC, 2013, N_2O has a Global Warming Potential (GWP) 265–298 times that of CO_2 for a 100-year timescale. The N_2O emissions from wastewater management accounts for about 26% of the total GHG emitted from water sector (Frison et al, 2015 cited in Nguyen et al, 2019). In a WWTP, N_2O is emitted during nitrification and denitrification processes used to remove nitrogenous compounds from wastewater. Nitrification is an aerobic bacterial respiratory process that releases N_2O as by-product (Inamori et al., 2008 2008 cited in Delre, 2018), while denitrification is a microbial-mediated process carried out mainly in anoxic conditions, where N_2O is an intermediate step (Tallec et al., 2008 cited in Delre, 2018). The quantity of N_2O emissions accounts for over 88% of the total GHGs released from the WWTPs (Daelman et al 2012). There is a significant increasing trend for N_2O from domestic wastewater and global N_2O volume between 1970 and 2012, from about 150 and 200 ktons of N_2O to roughly 350 and 365 ktons of N_2O, respectively as shown in Figure 4.

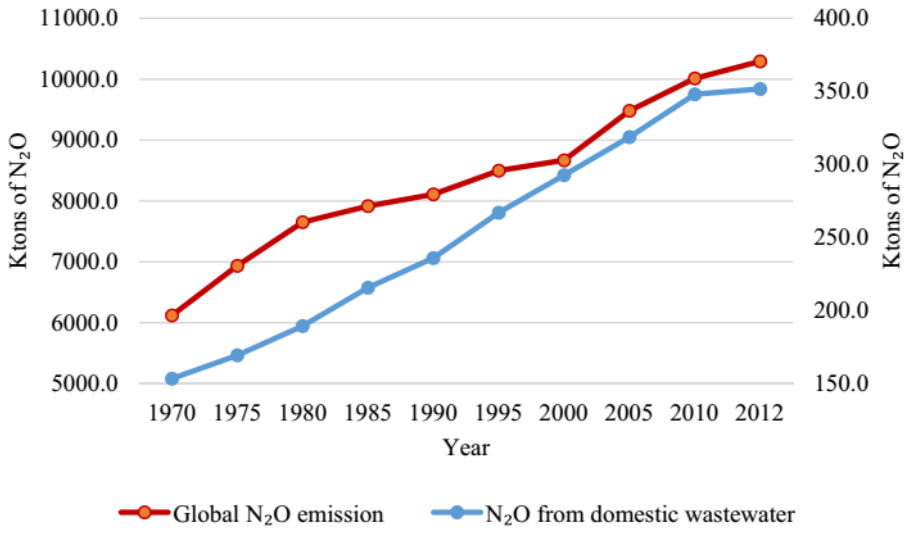

Figure 4: Total global N_2O emissions and N_2O emissions from domestic wastewater *(Janssens-Maenhout et al., 2017b cited in Nguyen et al., 2019)*

Kampschreur et al, 2009 cited in *Hass, 2018* noted that the global emissions from human sewage treatment was estimated to be 3.2% of total anthropogenic N_2O emissions. They also discussed that there were uncertainties around the N_2O emission factors and historical ambiguities over how the factors were derived or have been applied in GHG emission calculation protocols. Moreover, research has shown that measured N_2O emission rates vary widely according to type of treatment systems or unit processes, and can also vary widely both spatially and temporally within a given process (Kampschreur et al., 2009; Law et al., 2012; Ye et al., 2014; Ni et al., 2015; Pan et al., 2016 cited in Hass 2018).

The N_2O emission factor is typically represented as the ratio between the mass of emitted N_2O-N (kg-N d^{-1}) and the amount of influent Total Kjeldahl Nitrogen (TKN) load (kg-N d^{-1}). In some cases, the emission factors are represented as the ratio between the mass of N_2O-N emitted and the amount of N removed through nitrification and denitrification in the treatment plant (Law et al., 2012).

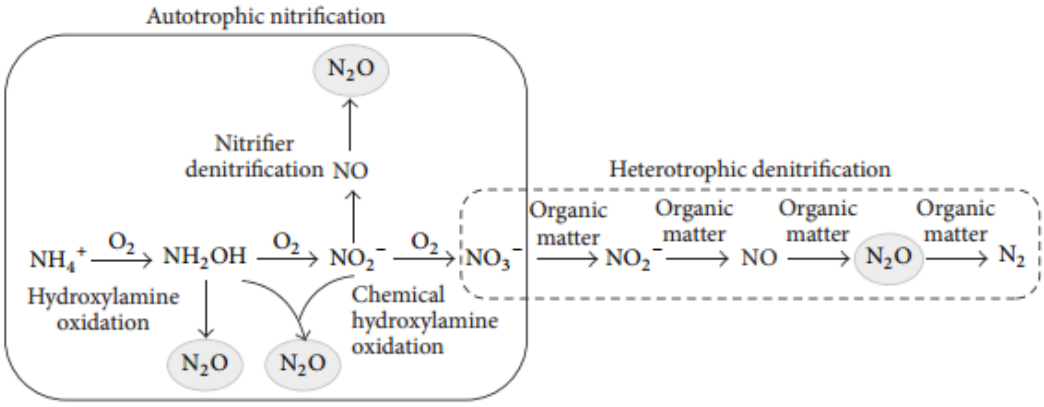

Figure 5: Biological and chemical pathways of N_2O production in the nitrification and denitrification processes

(Campos et al, 2016)

In 1995, *Czepiel et al*. had first published data on N_2O emissions from municipal wastewater treatment. They measured the N_2O emissions from primary and secondary wastewater treatment processes during spring and summer 1993 in Durham, New Hampshire, USA. The results showed that emission factors derived from field measurements included per capita emissions of 3.2 g of N_2O $person^{-1}$ yr^{-1} and flow based emissions of 1.6 x 10^{-6} of N_2O (L of wastewater)$^{-1}$ (Czepiel et al, 1995). Their measurements indicated that 90% of the N_2O emissions were from the activated sludge compartments, while grit tanks and sludge storage tanks released only 5% each.

Wicht and Beier, 1995 conducted single grab samples at 25 activated sludge plants in full-scale to estimate N_2O emissions. The author found that the N_2O emissions (per cent of N-load) was ranging from 0 to 14.6% (0.6% average), and N_2O emissions increased with increasing nitrogen load. *Sumer et al., 1995* and *Sommer et al., 1998* carried out fortnightly grab samples for one year, and 1 or 2 weeks sampling during 1.5 years, respectively at activated sludge plants (60,000 PE). While the result of *Sumer et al., 1995* indicated N_2O emissions was 0.001% of N-load and increased with nitrite and nitrate concentrations, *Sommer et al., 1998* found that N_2O emissions (0.02% N-Load) was higher 20 times than the results of *Sumer et al*., 1995.

By online measurement during 4 aeration cycles in 2 hours at an activated sludge plant (1,000 PE), *Kimochi et al, 1998*, had found the N_2O emissions were from 0.01 to 0.08 % of N-load and decreased with proportionally shorter aeration periods. *Kampschreur et al., 2008* had conducted two different research on nitritation - anammox sludge water treatment by online measurement during 4 days; and taking 3 grab samples during one day at the nitrification stage (nitrogen removal stage) of activated sludge plant (620,000 PE). The results demonstrated that N_2O emissions of nitritation reactor and anammox reactor were 1.7% and 0.6% of N-load respectively. The authors remarked that decrease in oxygen concentration (aerated stage) and increase of nitrite concentration (anoxic stage) made N_2O emissions increase. For the activated sludge plant, the nitrification stage emitted 4% N_2O emissions of the nitrogen load.

Besides research conducted in full-scale of WWTPs, there were several lab-scale WWTPs studies. The N_2O emissions data in the lab-scale ranged from 0 to 95% of nitrogen load. The results by *Benthum et al, 1998* showed the highest value 5 - 95% N_2O emissions of nitrogen load was from continuous oxic-anoxic SBR activated sludge treatment process in artificial wastewater (380 days). *Chung and Chung, 2000* had conducted studies on Batch tests denitrifying activated sludge for artificial wastewater by grab samples while *Itokawa et al., 2001* used online measurement to carry out research on continuous oxic-anoxic SBR activated sludge for artificial wastewater. They both asserted that N_2O emissions increased with decrease of COD/N. The findings by *Park et al., 2000* also proved that N_2O emissions decreased upon methanol addition for higher COD/N ratio. They grabbed samples in continuous nitrifying and denitrifying

active sludge processes for real wastewater and measured that 0.2 to 4.5% N_2O emissions of N-load was released in these processes.

Tsuneda et al., 2005 had daily grab samples in three different processes for artificial wastewater (30-300 days), including continuous nitrifying, denitrifying, and oxic-anoxic activated sludge processes. The N_2O emissions in the oxic-anoxic activated sludge treatment process was the highest, ranging from 0.7 to 13% of N-load while the lowest percentages of N_2O emission was for the continuous denitrifying activated sludge treatment process with 0.005 to 0.02% of N-load. The figure for the continuous nitrifying sludge activated treatment process was 0.2-0.5% of nitrogen load. The N_2O emissions increased with an increasing salt concentration in the continuous nitrifying and oxic-anoxic activated sludge treatment processes whilst it was independent of salt concentration in the continuous denitrifying sludge activated treatment process.

Research by *Foley et al, 2009* (as cited in Hass, 2018) measured N_2O emissions in seven WWTPs with the main process systems included an oxidation ditch, SBR, and several compartmentalized continuous-flow configurations (Johannesburg, A2/O or 3-stage Phoredox, and three different Modified Ludzack and Ettinger (MLE) types). They expressed results in kg N_2O-N/kg N denitrified (% TN removed). *Hass, 2018* had converted Foley et al, 2009 results to % influent TN as follows: average N_2O emissions factors were in the range 0.6-1.3% for the oxidation ditch; 0.7-5.0% for the SBR; 1.0-2.0% for the Johannesburg process; 1.0-1.7% for the A2/O process; and 0.5-8.5% for the MLE types.

Ahn et al., 2010b applied a 24-hour online to measure a wide range of N_2O fluxes from biological nitrogen removal (BNR) process in 12 WWTPs operated at different temperatures, configurations and influent TKN loads. The average emissions in each sampling period was calculated. On average, N_2O emission fractions varied from 0.01 to 1.8 or 0.01 to 3.3% when normalized to influent TKN load or influent TKN load processed, respectively (Ahn et al, 2010b).

Foley et al, 2011, measured online the N_2O emissions in four treatment plants (completely mixed, plug-flow, and membrane bioreactor). They found that the N_2O emissions coming from four WWTPs were from 0 to 0.3% of N-influent and the NH_4-N and DO concentration also affected N_2O emissions. The identification of factors that influence the amount of GHG emissions is of importance as it helps to calibrate models for calculating GHG emissions from WWTPs.

A research in a step-feed 'plug-flow' activated sludge plant in Australia by *Pan et al., 2015* achieved that the N_2O emissions was 75% TN removal on average with the emission factor for the plant overall was 1.9% of influent TN.

Table 4 below summarizes a few studies related to the direct emissions of N_2O from wastewater treatment plants based on summary tables of studies compiled by *Kampschreur, 2009* and *Delre, 2018* in their research.

Table 4: N_2O emissions (% of N load) from WWTPs

N_2O emissions (% of N-load)	**Sources**	**Method**	**References**
0.035	Activated sludge plant (11,000 PE)	Grab samples	Czepiel et al., 1995
0 – 14.6	25 activated sludge plants	Grab samples	Wicht and Beier, 1995
0.001	Activated sludge plant (60,000 PE)	Grab samples	Sümer et al., 1995
0.001 – 0.04	Activated sludge	Grab samples	Benckiser et al., 1996
0.02	Activated sludge plant (60,000 PE)	Grab samples	Sommer et al., 1998
0.01 – 0.08	Activated sludge plant (1,000 PE)	Online measurements	Kimochi et al., 1998
2.3	Nitritaion-anammox sludge water treatment	Online measurements	Kampschreur et al., 2008b
4	Nitrification stage (Nitrogen removal stage) of active sludge plant (620,000 PE)	Grab samples	Kampschreur et al., 2008b
0.02 – 0.08	Partial Nitritation and Anammox in One SBR	Grab samples	Joss et al., 2009
0.01 – 1.8	12 WWTPs	Online measurements	Ahn et al., 2010b
0 – 0.3	4 WWTPs	Online measurement	Foley et al, 2011
0.013 – 0.07	Sludge treatment	OSPM	Oshita et al, 2014
0.1 – 2.72	whole WWTP	MTDM	Yoshida et al., 2014
0.1 – 5.2	7 WWTPs	MTDM	Delre, 2018

There are several model that have been built and applied to quantify emissions of N_2O such as ASM1, BSM2, Bridle, ASM_2N4DN. *Snip, 2009* had applied models in quantifying GHG emissions from WWTPs. She found that by extending the ASM1 into the ASM_2N4DN using the equations proposed by Hiatt & Grady (2008), the N_2O emissions can be modelled dynamically and in more detail because it was taken into account the effects of different conditions on the production of GHGs including DO concentration in the aerobic tanks, ammonia and nitrate concentration in the last aerobic tank. *Ni et al., 2015* applied a mathematical N_2O model incorporating two N_2O production pathways by ammonia-oxidizing bacteria (AOB) namely the AOB denitrification and the hydroxylamine pathways) and the N_2O production pathway by heterotrophic denitrifiers to describe and provide insights into the large spatial variations of N_2O fluxes in a step-feed full-scale activated sludge plant. The authors found that the AOB denitrification pathway decreased and the NH_2OH oxidation pathway increased along the path of both steps due to the increasing dissolved oxygen concentration. The overall N_2O emissions from this step-feed WWTP would be largely mitigated if 30% of the returned sludge were returned to the Second Step to reduce its biomass nitrogen loading rate.

In general, N_2O emissions from wastewater treatment plants are relatively large. In 2007, IPCC reported that N_2O emissions from wastewater account for approximately 2.8 per cent of total anthropogenic sources. Between 2005 and 2020, global N_2O emissions from wastewater treatment were expected to increase by approximately 13 per cent (Law et al, 2012).

2.4. Summary of the literature review

This review of previously published research has shown that the study of direct GHG emissions generated from WWTPs has received the attention of many researchers. However, a lack in several studies is ignoring CO_2 emissions from WWTPs. Compared with CH_4 and N_2O emissions, CO_2 emissions have received very little attention although CO_2 emissions generated directly from WWTP are also significant and responsible for global warming. Besides, the overall diagram of GHG emissions from WWTPs has not completely been formed yet. Previous studies mainly focused on GHG emissions in the

secondary treatment and sludge treatment. This will be further clarified in chapter 4 of the report.

The studies assessing GHG emissions generated directly from WWTPs have been conducted by different measures and each measure has certain uncertainties. This will be discussed in more detail in the next chapter of the report.

CHAPTER 3: RESEARCH METHODOLOGY

3.1. Introduction

The main purpose of Chapter 3 is to present the methods and tools that can be applied to achieve the research objectives. The main method applied is the literature study for data collection, evaluation, and analysis of collected data. Moreover, it is necessary to understand the quantitative methods of GHG emissions generated directly from WWTPs. Therefore, in this chapter, methods and guidelines for the estimation of GHG emissions are also illustrated.

3.2. Data collection and analysis method

This study used the documentary study method for data collection and analysis. The method is conducted by studying previous topic-related documents and analysing the collected data, finding similarities, and limitations of the review studies. The reviewed documents are related to the direct GHG emissions from WWTPs. Through the study of published documents, the data, that are suitable with the research objectives and subjects, are gathered and synthesized. From the collected data, the report analyses and draws correct conclusions.

The structure of the report is as follows: the next section (Chapter 4) presents the evaluation of the gathered data in the reviewed papers, discusses the similarities, drawbacks, and challenges in assessing the direct GHG emissions from WWTPs, how the water industry could mitigate GHG emissions, and the UK net-zero 2030 route-map. In the last section (Chapter 5), the conclusion and suggestions for future research will be shown.

3.3. Methods for emission quantifications

Previous studies have used a variety of methods such as on-site measurements; MTDM; ASM1, Bridle, BSM2, and ADM1 models, IPCC guidelines for quantitative GHG emissions generated from WWTPs. Each method has advantages and different measurement ways of GHG emissions from WWTPs. This study will not present all the methods, only typical and popular methods for quantifying direct CH_4 and N_2O emissions from WWTPs are selected to present in this report.

3.3.1. CH_4 emission measurements

Methane (CH_4) is one of the three main GHG generated from wastewater treatment plants and the dominant sources are from anaerobic digesters and anaerobic sludge storage tanks as well as the sewer system. Previous research applied several methods to measure CH_4 such as grab samples or online measurements, and mobile tracer gas dispersion method (MTDM). Because CH_4 belongs to the chemical group of Volatile Organic Compounds (VOCs) indicating that it can easily vaporize (Kosse et al, 2018), the methods for quantifying and qualifying CH_4 released from WWTPs have taken advantage of this property to develop suitable methods. Chamber measurements and tracer release methods are two popular methods for CH_4 quantification that are based on this property.

3.3.1.1. Chamber methods

Chamber techniques are a widely popular method of studying GHG emissions for aquatic studies. This method can be applied to measure both CH_4 and N_2O generated from the WWTPs. For CH_4 emission measurements, the chamber techniques can be divided into 2 different measurements including accumulation (closed-chamber) measurements (Frakignoulle, 1988 cited in Kwok et al, 2015), and flow-through (open-chamber) measurements (Kwok et al., 2015).

- **The accumulation (closed-chamber) measurements**

The closed chamber had a small vent hole (ca. 10 mm in diameter) vented to the atmosphere to allow the chamber pressure to equalize to atmospheric pressure when the chamber is first placed on the water (Kwok et al, 2015). The vent after a certain of time is closed against ambient air. According to *Kwok et al, 2015*, the methane flux from the chamber is calculated from linear increase of the measured gas mole fraction in the chamber with time as Equation 1 below.

$$\mathbf{F_{CH4}} = \frac{\Delta C}{\Delta t}\ \frac{pVMA_{basin}}{RTA} \qquad \text{(Eq.1)}$$

Where: F_{CH4}*: the methane flux from the chamber (g/s);*

$\frac{\Delta C}{\Delta t}$*: the fitted linear increase of the measured gas mole fraction in the chamber with time (mol mol^{-1} s^{-1});*

p: the pressure in the floating chamber (Pa);

V: the volume of the chamber (m^3);

A_{basin}: the area of the basin (m^2);

M: the molar mass of methane (g mol^{-1});

T: the temperature (K);

R: the universal gas constant, R=8.314 m^3Pa $K^{-1}mol^{-1}$;

A: the water surface area enclosed by chamber (m^2).

Note: For the temperature and ambient pressure parameters, it is possible to use measuring equipment or collect data at the nearest weather station.

- **Flow-through (open-chamber) measurements**

Oppositely, the open chamber allows methane-free air to be blown into one end of the chamber and excess injected air to escape from the other end. The air in the chamber was replaced by the injected air. Therefore, the methane flux from the chamber is simply the measured concentration in the exit-air multiplied by the flow-rate of air used (Hobson, 2000) (Eq.2).

$$\mathbf{F_{CH4}} = \sum_t (\boldsymbol{C_{chamber}} - \boldsymbol{C_{baseline}}) \frac{\boldsymbol{M}}{\boldsymbol{V_m}} \frac{\boldsymbol{dV_{aeration}}}{\boldsymbol{dt}} \mathbf{A_{aeration}} \qquad \text{(Eq.2)}$$

Where: *F_{CH4}: the methane flux from the chamber (g/s);*

$C_{chamber}$: the gas measured concentration in the chamber (mol mol^{-1});

$C_{baseline}$: the injected gas concentration (mol mol^{-1});

M: the molar mass of methane (g mol^{-1});

V_m: the molar volume of ideal gases (m^3 mol^{-1});

$\frac{dV_{aeration}}{dt}$: the flow-rate of injected air (m^3 h^{-1});

$A_{aeration}$: the surface area of the aeration area in the aeration basin (m^2).

Each method has different outstanding features, however, they also have limitations and errors. While the errors of the closed-chamber measurement are related to the volume of the chamber, the water level in the chamber, the sensors for pressure and temperature, for open-chamber measurement, the uncertainties include the injected air volume, the background CH_4 concentration, and the error of the CH_4 measurement (Kowk et al, 2015).

In general, the above limitations are unavoidable. Nevertheless, in order to minimize errors in the measurements, it is necessary to take into account the possible errors and determine the ranges of errors by studying previous research. The differences between two chamber measurement methods is shown in Figure 6.

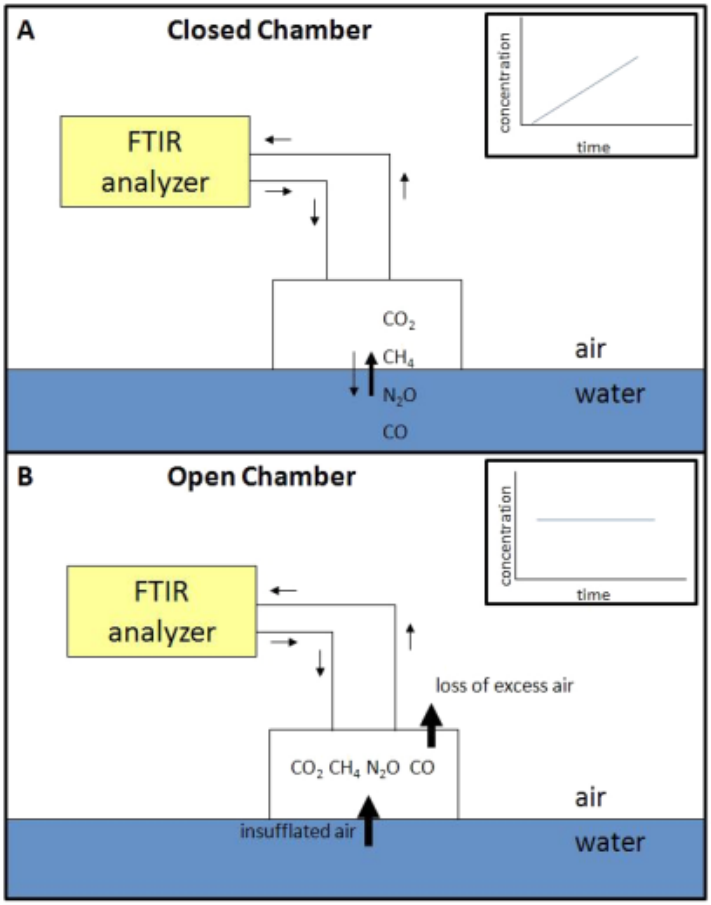

Figure 6: Schematic showing different modes of two chambers deployment.

(a) Conventional floating chamber used on a calm surface (accumulation closed-chamber measurements). The schematic concentration vs. time points out how the gas accumulates in the chamber over time (in case of a positive net flux from water to air). This increase is linearly approximated and from the slope, the flux is calculated.

(b) Flow-through open chamber: the excess air escapes and the concentration measured in the chamber relates directly to the concentration in the emitted air.

(Kowk et al, 2015)

3.3.1.2. Tracer release method

The tracer release method can be conducted in two ways including mobile tracer gas dispersion method (MTDM) and static tracer gas dispersion method (STDM). The

MTDM combines the known release of tracer gas with measurements of downwind atmospheric gas concentrations carried out with a vehicle (Delre, 2018), while the STMD uses a static instrument. The releasing tracer gas is C_2H_2 at a known rate and the unknown emission of a trace gas to be determined is CH_4.

For the MTDM, concentrations of the tracer as well as the gas of interest are measured using a mobile instrument downwind in the co-propagating plumes (Kwok et al, 2015). The vehicle with GPS device carried gas analysers which are used to analyse samples taken from the roof of the vehicle. The C_2H_2 released gas is controlled by gas cylinders with calibrated flow meters (Delre, 2018).

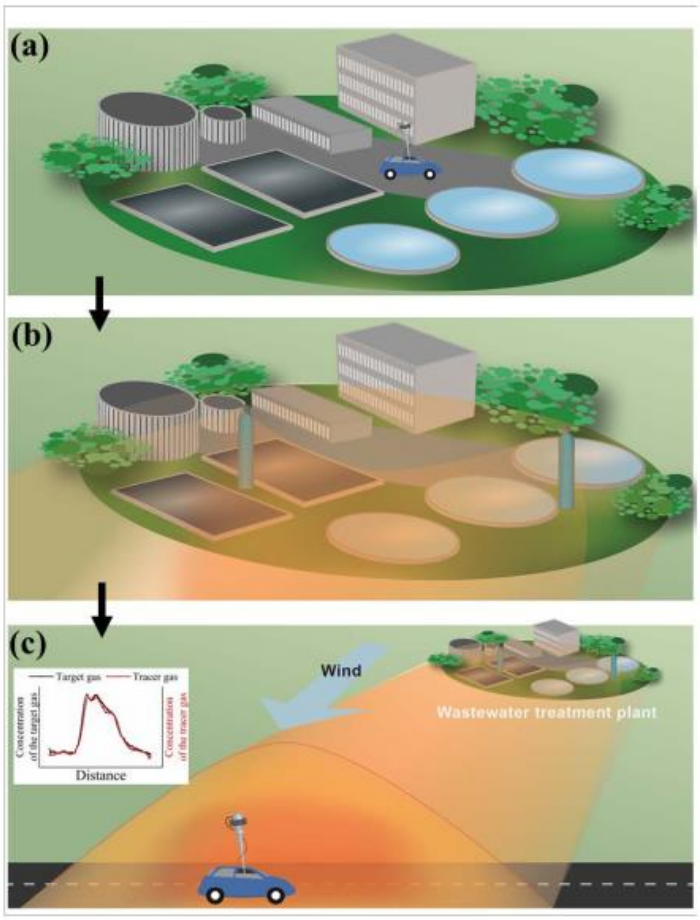

Figure 7: Application of the mobile tracer gas dispersion method at a wastewater treatment plant

(a) The on-site screening phase; (b) Tracer gas placement and controlled release; (c) The quantification phase, when the plume is traversed for measurement of target and tracer gas concentrations.

(Delre, 2018)

Regarding the STDM, C_2H_2 was released in the enclosed ventilated duct upstream of a fan in the WWTP building, which allowed for the proper mixing of tracer and target gases where gas sampling occurred. The gas samples were taken at the end of the duct, where the gas analyser was placed at a fixed position (Delre, 2018).

According to Kwok et al, 2015, the emission rate of the determined gases can be calculated as follows.

$$F_{CH_4} = F_{C2H2} \cdot \frac{A_{CH4}}{A_{C2H2}} \cdot \frac{M_{CH4}}{M_{C2H2}} \qquad \text{(Eq.3)}$$

Where:

F_{CH_4} *: The emissions of* CH_4 *(kg/h);*

F_{C2H2}*: The known emission of* C_2H_2 *(kg/h);*

$\frac{M_{CH4}}{M_{C2H2}}$ *: The ratio of the molar masses of* CH_4 *and* C_2H_2

$\frac{A_{CH4}}{A_{C2H2}}$ *: The ratio of* CH_4 *concentration and* C_2H_2 *concentration*

+ For STMD, the ratio is just between the concentrations of two gases ($C_{CH_4\ detected}$, and $C_{C2H2\ detected}$).

+ For MDTM, the ratio needs to subtract the background (A_{CH_4} = $C_{CH_4\ detected}$ - $C_{CH_4\ baseline}$ and A_{C2H2} = $C_{C2H2\ detected}$ - $C_{C2H2\ baseline}$).

The units of CH_4 and C_2H_2 concentrations are parts per billion (ppb).

The main drawbacks of the tracer release methods are that they can be affected by weather conditions, surrounding activities of the background environment (noise, ambient electronic equipment), different background CH_4 concentrations in space and time, and gas dispersion. To address this issue, the background for each CH_4 plume is calculated using a linear regression between the first and last point of the peak instead of removing an average background value for the whole event (Kowk et al, 2015). In addition, the analyser's resolution and accuracy also should be taken into account during performing tracer release measurements.

3.3.2. N_2O emission measurements

Domestic wastewater usually contains relatively high concentrations of nitrogen, around 20-70 mg l^{-1} total nitrogen as N (Law et al, 2012). Nitrogen removal in the secondary

treatment stage (biological process) in the WWTPs releases a significant amount of N_2O. This has caught the attention of several researchers.

There are many different methods that can be applied to quantify NO_2 in the wastewater treatment process, in which, gas-phase (chamber) and liquid-phase nitrous oxide measurements are two methods that have been used in several previous studies.

3.3.2.1. Chamber measurement

As mentioned above, chamber methods can be used to measure both CH_4 and N_2O emissions from WWTPs. Similar for measuring CH_4 emissions, an open-chamber is used to blow nitrogen-free air into the headspace of the chamber during non-aerated phases, and strip dissolved N_2O from the liquid phase into the gas during aeration. The gas flow through the chamber in non-aerated zones can be recorded with a rotameter. For aerated zones, the gas flow out of the chamber is equal to the air flow for aeration (Law et al, 2012). To calculate nitrogen flux, Equation 2 can be applied for N_2O.

3.3.2.2. Liquid-phase nitrous oxide measurement

This method used off-line grab samples and then using a gas chromatograph to analyse the samples that can be conducted in both laboratory-scale reactors and full-scale plants. A liquid sample containing N_2O is injected into a vacuum vial and allowed to reach liquid-gas equilibrium (Law et al, 2012), and then measure and note the gas-phase N_2O concentration ($C_{N2O\ gas}$). Henry's law then is applied to calculated liquid-phase N_2O ($C_{N2O\ liquid}$). The total N_2O concentration in the sample is obtained by dividing the total amount of N_2O in both the gas and liquid phases by the total liquid volume (Law et al, 2012) (Eq.4).

$$\Sigma\ C_{N2O\ \text{in the sample}} = \frac{C_{N2O\ gas} + C_{N2O\ liquid}}{\Sigma V_{liquid}} \qquad \text{(Eq.4)}$$

To measure dissolved N_2O concentrations, N_2O microsensors, which are connected to a highly sensitive pico-ammeter, can be used. N_2O penetrates through the sensor tip membrane and is reduced at the metal cathode surface (Law et al, 2012). When the current running in the sensor is reduced, a signal can be captured and recorded on a computer. The response of electrochemical microsensors is known to be linear in the range 0-1,2 mM (Andersen et al, 2001 cited in Law et al, 2012).

Because the N_2O concentration is varied at different sites, it is necessary to determine the spatial variation of N_2O concentration. Parameters including pH, DO, temperature TSS, VSS are usually measured at the sampling points and at the influent for mass balance and correlation analysis of N_2O emission fluxes.

While N_2O microsensors have a low detection limit, the high sensitivity can render it susceptible to interference especially in full-scale measurements (Law et al, 2012). Moreover, it is difficult to estimate the mass transfer coefficient between the liquid and gas phases, especially in full-scale plants.

3.4. Guidelines for GHG emission estimations

❖ IPCC Guidelines

Since the amount of GHG emissions from the water industry is significantly contributing to accelerating global warming and climate change, in 2006, the IPCC developed a guide for estimating GHG emissions for many industries which includes GHG emissions from WWTPs.

For annual CH_4 emissions, the IPCC guideline indicates three tiers based on the data available. Each country can base on that to develop their national guideline.

- Tier 1 is applied when country-specific emission factors (EFs) are not available. The EFs and activity parameters are defaulted in this tier. Tier 1 is the simplest method for any country but it has the lowest accuracy.
- Tier 2 estimation emissions is the same as Tier 1 but used country-specific EFs. This tier method is more complicated but it may not improve the accuracy.
- Tier 3 estimate emissions is applied when a country-specific method is available.

For annual N_2O emissions, direct emissions from nitrification and denitrification at WWTPs may be considered as a minor source (IPCC, 2006) and the guidance offers a specific formula to estimate these emission as follows.

$$\mathbf{N_2O_{PLANTS} = P \times T_{PLANTS} \times F_{IND\text{-}COM} \times EF_{PLANT}} \quad \text{(Eq. 5)}$$

Where:

N_2O_{PLANTS} : *total N_2O emissions from plant in inventory year, kg N_2O/yr;*

P : *human population;*

T_{PLANTS} : *degree of utilization of modern, centralized WWT plants, %;*

$F_{IND-COM}$ *: fraction of industrial and commercial co-discharged protein (default = 1.25, based on data in Metcalf & Eddy (2003) and expert judgment)*

EF_{PLANT} *: emission factor, 3,2 g N_2O/person/year*

❖ **The UK Environmental Reporting Guidelines**

In the UK, the UK Government had built Environmental Reporting Guidelines in 2019 including streamlined energy and carbon reporting guidance helping businesses and organisations manage their GHG emissions from their activities. The guidance provides that GHG emissions are equal to the activity data, that is recorded or estimated, multiplied by an emission (conversion) factor.

Activity data x Emission factor = GHG emissions (Eq.6)

This equation is applied to estimate annual GHG emissions from any activity in the UK. The UK Government GHG Conversion Factors in 2019 only described the Scope 3 emissions for water EFs. Therefore, this report cannot apply the Eq.5 to estimate the direct GHG emissions from WWTPs because the boundary of the research is only the Scope 1 emission (the direct GHG emissions from WWTPs).

The limitations of IPCC and UK Government guidelines are the lack of information, data accuracy, and uncertainty in EFs. Because the formulas in IPCC guidelines are only for the direct and indirect amount of CH_4 and N_2O emissions from WWTPs, it does not fully take into account all the GHG emissions from WWTPs. CO_2, for instance, is also a direct and indirect GHG emissions from WWTPs due to microbial activities and energy consumption, but it is not considered in the IPCC Guidelines. Therefore, the GHG emissions generated from WWTPs may be underestimated. Besides, some data in guidelines formulations default and it only can apply under certain conditions. In cases, WWTPs apply different treatment technologies under different conditions, applying the guidelines calculated formula can affect the accuracy of the data. Another important factor is the EFs, according to the IPCC guidelines the default EFs can be applied to any country. Nevertheless, it is suggested to use the country-specific factors when available (Zhan et al, 2017 cited in Nguyen et al, 2019). The EFs in the formula of UK guidelines are the determinant of total GHG emissions, however, since 2012, the water UK has discontinued its “Sustainability Indicators” report and so no longer produces further

updates to these emission factors (Department for Business, Energy & Industrial Strategy, 2019).

To mitigate the above limitations on the calculation of GHG emissions generated by WWTPs in the UK, it is suggested that Water UK continues to have the proper synthesis and calculation to provide suitable EFs with the development of wastewater treatment technologies being applied in the UK. In addition, in order to verify the results from the application of the calculated formula according to the UK and IPCC guidelines, it is recommended to measure direct GHG emissions generated from WWTPs by applying described methods above.

3.5. Summary

The goal of this chapter was to present the methods used to answer the research questions. The main method to conduct this research was the documentary study for the data collection and analysis findings. The report also presented a summary of applicable methods for estimating CH_4 and N_2O emissions generated directly from WWTPs, noting the uncertainty and limitations of each measure.

CHAPTER 4: CRITICAL ANALYSIS AND DISCUSSION

This chapter contains the evaluation of the data collection and discussion of the similarities and difficulties in estimating the direct GHGs from WWTPs. Moreover, GHG emission mitigation methods and plans to reduce GHG emissions from the water sector in the UK will be introduced in this chapter. Chapter 4 aims to gain the goals of assessing the direct GHG emissions generated by the WWTPs; analysing the environmental impacts, and proposing solutions to reduce the direct GHG emissions coming from the WWTPs.

4.1. Direct GHG emissions from WWTPs

The direct GHG emissions from WWTPs occur at different treatment steps in varying conditions and are mainly from the biological treatment process. CO_2 is mainly released from microbial respiration activities while N_2O is fluxed from denitrification, nitrification stages, and CH_4 mainly comes from anaerobic digestion (Zhang et al, 2017 cited in Nguyen et al, 2019). Based on the layout of a WWTPs shown in Chapter 2 and the literature review, a direct GHG emissions map can be indicated in Figure 8.

- **Preliminary treatment**

At the pre-treatment step, there have yet not any studies related to direct GHG emissions at this step. However, CH_4 is supposed to emit from a WWTP after it enters the plant via stripping from the incoming wastewater, or after it is formed at the plant itself (Parravicini et al., 2016) at primary treatment and sludge line. Moreover, the influent of a WWTP contains dissolved methane that is formed in the sewer system. Recent studies indicate that methane formation in sewer systems can be substantial (Foley et al., 2009; Guisasola et al., 2008), but actual quantities of methane entering a WWTP have as yet not been reported (Daelman et al., 2012).

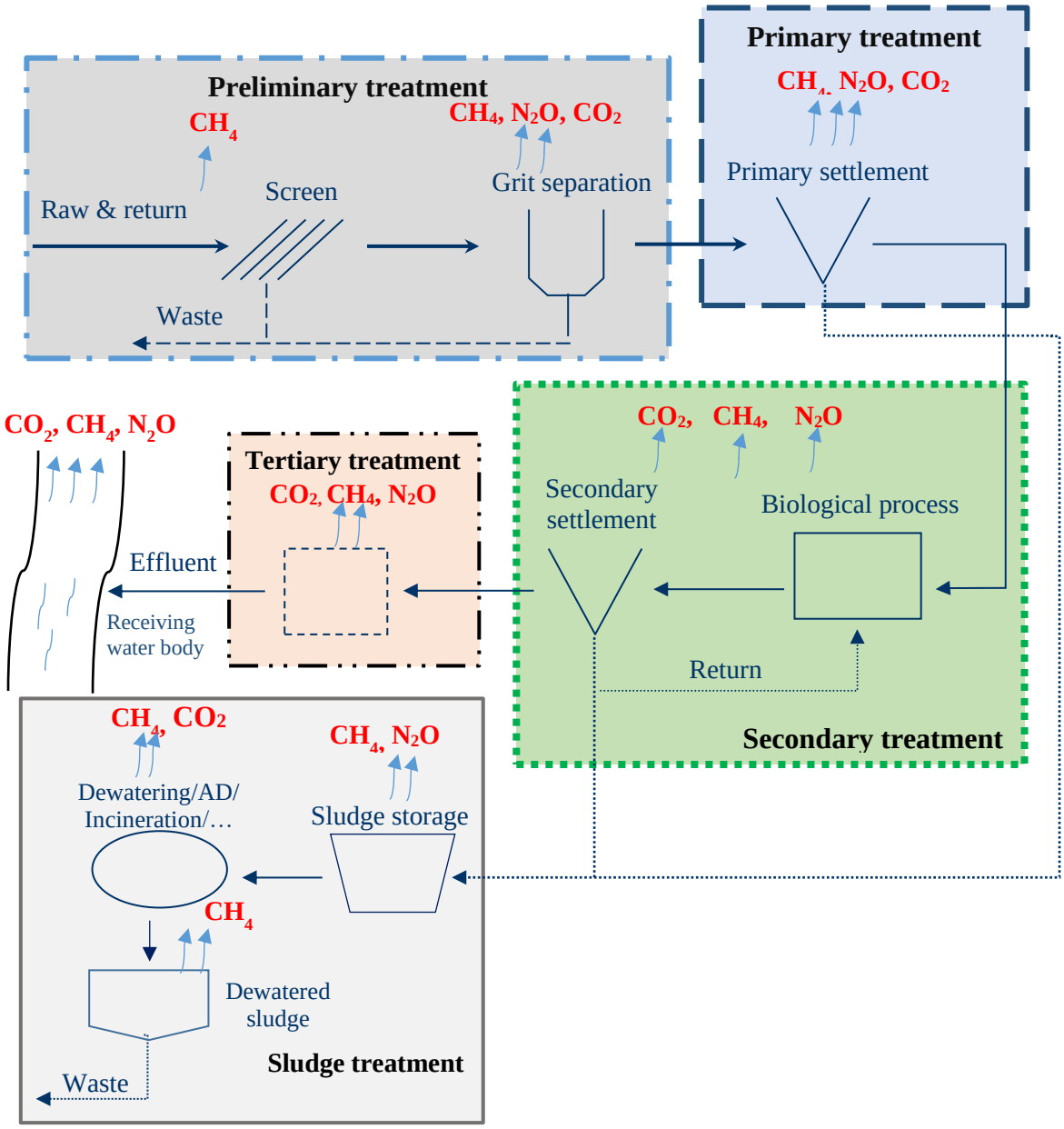

Figure 8: General layout of a WWTP with direct GHG emissions

A few studies on each type of GHG emissions have proved that the pre-treatment generates all three main GHG emissions, in which the amount of CO_2 emissions in this stage is considered significant.

In the results published by *Samuelsson et al, 2017*, sand traps in the pre-treatment stage contributed to the total of 9% of CH_4 emissions and <1% of N_2O emissions, with emission rates of 1.8 kg h^{-1} and 0.01 kg h^{-1}, respectively. In the research by *Liu et al.,2014* and *Wang et al., 2011*, the results indicated that the aerated grit tank emitted 0.026 g/m^3 (equivalent to 0.542 kg h^{-1}) and 0.022 g/m^3 of CH_4, respectively. The N_2O

emissions from the aerated grit tank was different in research by *Sun et al., 2013; Ren et al., 2013;* and *Yan et al., 2014* (5.51 kg d^{-1}, 6.00 kg d^{-1} and, 24.60 kg d^{-1}, respectively). Research on characteristics of direct CO_2 emissions in four full-scale wastewater treatment plants by *Bao et al., 2015* showed that 34.32 g CO_2/m^2 d (or 960.96 g d^{-1}) of dissolved CO_2 was emitted in the swirl grit tank in the plant using the SBR process to treat wastewater, while in the aerated grit tank in the WWTP using the AAO process, the average CO_2 flux was 3,728.44 g CO_2/m^2 d (1,879.13 kg d^{-1}), ranging from a minimum of 969.16 g CO_2/m^2 d (488.46 kg d^{-1}) to a maximum of 7,650.7 g CO_2/m^2d (3,855.95 kg d^{-1}).

- **Primary treatment**

The primary treatment generates CH_4 in the primary sedimentation/settlement tank but it then is stripped out with the aerobic air in the activated sludge tank in the next treatment process (Parravicini et al., 2016). This stage contributed about 4% of CH_4 emissions to the total (Samuelsson et al, 2017). Research by Bao et al., 2015 also proved that the primary sedimentation tank emitted 5.04 g CO_2/m^2d (or 127.01 kg d^{-1}), and with a range of 3.35 g CO_2/m^2d – 9.65 g CO_2/m^2d (equivalent to 84.42– 242.18 kg d^{-1}), which was quite small due to the limited biological activities and the stable water surface.

Although there are not many studies showing that N_2O is emitted in the primary treatment, Samuelsson et al, 2017 indicated that the N_2O emissions in the primary settlers accounted for 3% of the total N_2O emissions from WWTPs.

The emissions from primary treatment could be negligible compared to the secondary treatment and sludge treatment stages, however, it is unavoidable some leakage and escape from this treatment stage, and also necessary to be evaluated.

- **Secondary treatment**

In the secondary treatment, nitrogen is removed and it mainly releases N_2O due to biological breakdown. For AAO technology, the largest amount of N_2O was emitted in the oxic zone due to nitrifying activities of the ammonia-oxidising bacteria (AOB) (Massara et al, 2017, cited in Nguyen et al, 2019). This is also demonstrated in previous studies by *Ren et al., 2013; Sun et al, 2013a; and Yan et al., 2014.* The amount of N_2O

emissions generated at each AAO treatment unit of the three studies above was gathered by *Nguyen et al, 2019* as shown in Table 5.

Table 5: N_2O emissions in AAO process

Treating unit	*Sun et al, 2013a, 2013b*		*Ren et al, 2013*		*Yan et al, 2014*	
	kg/day [(a)]	**kg/h [(b)]**	**kg/day [(a)]**	**kg/h [(b)]**	**kg/day [(a)]**	**kg/h [(b)]**
Anoxic zone	1.32	0.055	14.05	0.59	22.10	0.92
Anaerobic zone	7.59	0.32	7.66	0.32	10.80	0.45
Oxic zone	471.70	19.65	6,030.00	215.25	9,745.90	406.08
Total	**480.61**	**20.025**	**6,051.71**	**216.16**	**9,778.80**	**407.45**

Note: "a": Nguyen et al, 2019; "b": this study

For the SBR process, more than 90% of nitrous oxide emissions occurred during the aeration phase due to air-stripping of dissolved nitrous oxide (Kampschreur et al, 2009). *Sun et al, 2013a* indicated that in the SBR WWTP, the emissions of N_2O was 339.24 kg d^{-1} with 99.9% of the total emissions coming from the periods of feeding and aeration. During aeration phase, dissolved N_2O was stripped, and in the non-aerated zone, N_2O emissions contributed 94 ± 4% to the total N_2O emissions in the first 15 min of the aeration phases (Law et al, 2011). *Bao et al, 2016*, also found that the nitrous oxide emitted dominantly in the feeding and aeration periods with 1.13 and 0.75 $g.m^{-3}$, respectively.

Several studies have mainly indicated that N_2O is emitted during nitrifying and denitrifying. However, CO_2 and CH_4 are also released in this step as both technologies have biochemical reactions under anaerobic and anoxic conditions converting organic matters into CH_4 and CO_2, and ammonia-nitrogen into nitrite-nitrogen and then nitrate-nitrogen. Previous research has proved that large amounts of CO_2 and CH_4 were stripped in the secondary treatment. Most CO_2 and CH_4 emissions were found in the oxic tank in the AAO process whilst, these two GHGs were mainly detected in the feeding and

aeration phases in the SBR process due to the aeration respiration and aeration stripping process (Bao et al, 2015). The results of direct CO_2 emissions in the AAO and SBR processes in the study by *Bao et al., 2015* are shown in Table 6.

Table 6: Direct CO_2 emission from the AAO and SBR processes

AAO process	**CO_2 emissions**		**SBR process**	**CO_2 emissions**	
	kg/day	**kg/h**		**kg/day**	**kg/h**
Anoxic tank	215.66	8.99	Feeding and aeration period	48,877.02	2,036.54
Anaerobic tank	242.00	10.08	Settling period	94.38	3.93
Oxic tank	72,651.20	3,027.13	Decanting period	89.75	3.74
Total	***73,108.86***	***3,046.92***	**Total**	***49,061.15***	***2,044.21***

Source: Bao et al., 2015

Liu et al., 2014 discovered the CH_4 emissions from each treatment unit under different conditions from the AAO and SBR processes in two municipal WWTP with capacity of 5 x 10^5 m^3 day^{-1} and 8 x 10^4 m^3 day^{-1}, respectively. The results are shown in Table 7.

Table 7: Methane emitted from each treatment unit from AAO process

AAO process unit	**Emissions (g CH_4/m^3)**	**Emission rate (kg/h)**	**SBR process unit**	**Emissions (g CH_4/m^3)**	**Emission rate (kg/h)**
Anoxic tank	0.007	0.145	Feeding and aeration phase	1.59	5.3
Anaerobic tank	0.019	0.396	Settling phase	0.001	0.003
Oxic tank	0.371	7.729	Decanting phase	0.001	0.003
Total		**8.812**	**Total**		**5.306**

Source: Liu et al.,2014

Figure 9 shows the CO_2, CH_4 and N_2O emissions in both two different treatment technologies, namely SBR and A2O processes.

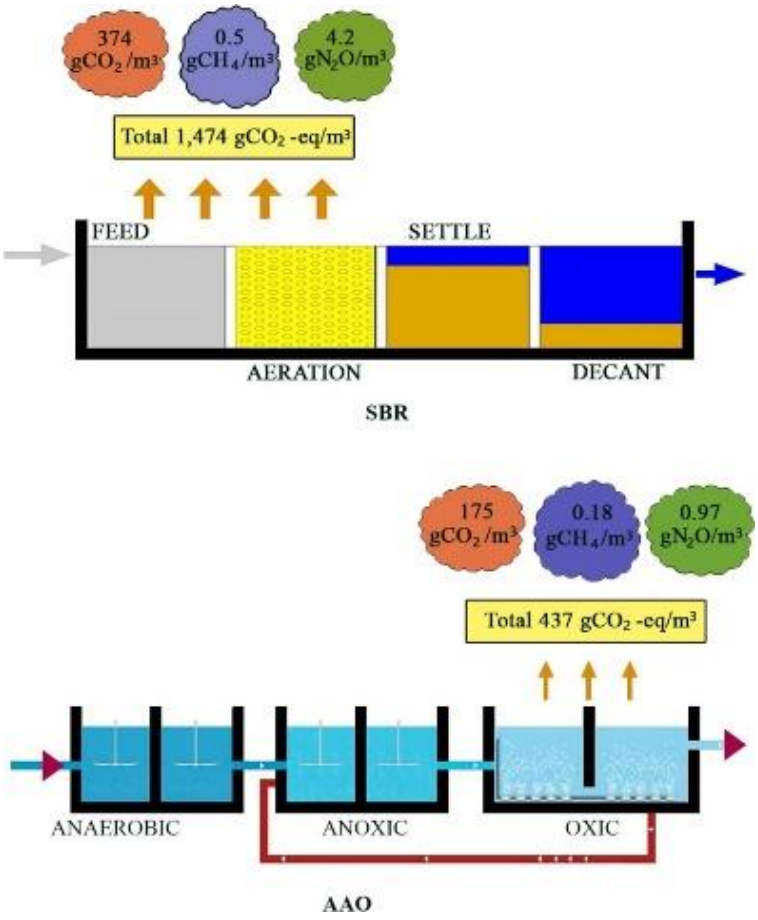

Figure 9: GHG emissions from SBR and AAO processes in WWTPs

(Nguyen et al, 2019)

Nguyen et al, 2019, had carried out research on insight into GHG emissions from the two popular treatment technologies (SBR and AAO) in municipal wastewater treatment processes. The authors found that a quantity of total CO_2 emissions from SBR (374g/m^3 of wastewater) was double that of AAO whilst 10% of these were direct CO_2. CH_4 emitted from an SBR was 0.50g/m^3 wastewater while 0.18g CH_4/m^3 wastewater was released from an AAO. The level of N_2O from AAO and SBR accounts for 0.97g/m^3 and 4.20g/m^3 wastewater, respectively.

- **Tertiary treatment**

In this stage, wastewater is continuously treated by some different methods such as chemical dosing, advanced oxidation systems, and sand filters before discharging into the environment. Depending on the technique and chemicals used to disinfect, there may be some biological breakdown that could emit CO_2, CH_4, and N_2O, however, there is not any specific research on the amount of GHG emissions generated from the tertiary

treatment stage. Since the majority of the BOD, COD, phosphorus and nitrogen are removed at previous stages, the emissions in this final stage could be considered negligible.

Although wastewater is treated, it still can generate GHGs. The COD and TN leaving the WWTP with the treated effluent can promote further CH_4 and N_2O production and emissions in the receiving water bodies, depending on existing milieu conditions (Parravicini et al, 2016).

- **Sludge treatment**

For the treatment of sludge from WWTPs, various methods can be applied to reduce water in the sludge. Different sludge treatment methods will generate different quantities of GHG emissions. *Piippo et al, 2018*, conducted research on GHG emissions from different sludge treatment methods in Northern Finland. The results showed that the anaerobic digestion (AD) method generated the least CO_2eq emissions of all treatment methods studied. The second best option was the incineration of sludge without thermal drying, while the third-best was composting or incineration of sludge after thermal drying with fossil or other fuels.

The sludge treatment process from a WWTP is the largest methane sink with 81% of methane emissions of the total methane emissions from the plant itself according to *Samuelsson et al, 2017*. All tanks in the sludge treatment line generate CH_4 emissions. Numerous studies have proved that sludge storage contributes significantly to the CH_4 emissions. CH_4 is produced both in the digested sludge buffer tank, as well as in the dewatered sludge storage tank (Daelman, 2014).

Since the influent sludge can be converted to biogas, and this process occurs continuously for a certain period of time, the sludge digestion and sludge storage produce a significant quantity of CH_4 emissions. *Daelman, 2012* concluded that the methane emissions related to the anaerobic digestion of primary and secondary sludge counts for about three quarters with respect to the WWTPs overall methane emissions. *Czepiel et al, 1993* (cited in Hobson, 2000), made measurements of CH_4 emissions from sludge treatment and found that the primary treatment (including preliminary treatment) and secondary treatment emitted 0.0056 and 0.0079 Tg of US annual emissions of CH_4,

respectively. Sludge digestion was the process that produced the highest methane emissions with 0.84 Tg of US annual CH_4 emissions.

In the UK, *Watt, 1994* made an estimate of methane emissions from the treatment of sewage sludge (Hobson, 2000). The author calculated the total annual methane emissions from the sludge treatment was 69 kt (0.069 Tg), of which 0.049 Tg of CH_4 emissions was generated from the treatment process itself, and 0.02 Tg was from the disposal of sewage sludge to landfill.

Besides CH_4 emissions, sludge line treatment also generates a small amount of CO_2 and N_2O emissions. While the decomposition of organic matter in the sludge from WWTPs emits CO_2 emissions, the sludge storage tanks release N_2O emissions. Although the amount of CO_2 and N_2O emissions may be insignificant compared to the quantity of CH_4 emissions in this stage, it still should be considered when estimating CO_2 and N_2O emissions generated directly from WWTPs.

4.2. Environmental Impacts of GHG emissions from WWTPs

The wastewater treatment process itself generates GHGs into the environment. During the biological treatment process of WWTPs, a portion of dissolved organic matter is converted into the components of the microorganisms through anabolism and then be oxidized to CO_2 and CH_4 and emitted into the air; the residual material will participate during catabolism and eventually also be converted into gasses and emitted into the air (Wanqiu et al, 2019). Moreover, the nitrogen removal in wastewater treatment also releases a large amount of N_2O into the air.

Besides the GHG emissions from the waterline, the sludge line from WWTPs is a large methane sink. Long-term storage of sludge in the lagoons is no longer common in the UK, instead sludge is treated by modern treatment technologies, which rapidly dewater in sludge, before it is transported to landfill. Regardless of the technology, sludge treatment contributes a large amount of methane to the air.

Moreover, the operation of WWTPs requires large amounts of energy and chemicals, which emit even higher quantities of GHGs than the plant itself.

Both direct and indirect GHG emissions arising from WWTPs can affect three basic environmental components: soil, water, and air. CO_2, CH_4 and N_2O are the three main

GHGs responsible for global warming and climate change. While the global warming potential (GWP) of CO_2 is 1 over a 100-year time horizon, the figure for CH_4 is 34 CO_2 – equivalents, and N_2O is even more powerful with a GWP 265-298 times that of CO_2 for a 100-year timescale according to IPCC, 2013. Once the GHGs emitting into the air, they will trap the heat in the atmosphere and make the planet warmer. Consequently, they alter environmental components.

Table 8 below summaries the GHG emissions in each stage from WWTP based on the data in section 4.1.

Table 8: Summary GHG emissions from in WWTP

Treatment stage		**CO_2 emissions (kg/h)**	**CH_4 emissions (kg/h)**	**N_2O emissions (kg/h)**
Preliminary treatment (the aerated grit tank)		**78.30** (Bao et al., 2015)	1.8 (Liu et al.,2014)	1.025 (Yan et al., 2014)
Primary treatment		**5.29** (Bao et al., 2015)	0.8 (Samuelsson et al, 2017)	0.1 (Samuelsson et al, 2017)
Secondary treatment	AAO process	**3,046.92** (Bao et al., 2015)	**8.812** (Liu et al.,2014)	**407.45** (Yan et al, 2014)
	SBR process	**2,044.21** (Bao et al., 2015)	**5.306** (Liu et al.,2014)	**14.135** (Sun et al, 2013a)
Tertiary treatment		-	-	-
Sludge treatment		-	-	-

Note: "-": missing data

The amounts of GHG emissions from WWTPs with distinct operating conditions and treatment technologies are different. However, based on the aggregated data in Table 8, it can be seen that the quantities of GHG emissions generated from WWTPs are significant.

In the UK, the contribution to GHG emissions from water industry operations to the annual UK GHG emissions are relatively high, nearly 1% in 2008 (Reffold et al, 2008).

Among water sector activities, wastewater treatment contributes the highest amount of emissions (including direct and indirect GHG emissions) with 56%, followed by clean water with 39%, and administration and transport with 5% (Charles et al, 2009). Although the amount of GHG generated by WWTPs is mainly from energy consumption, it is undeniable that the direct GHG emissions also have certain effects on the air environment.

4.3. Discussion of the similarities and difficulties in assessing the direct GHGs from WWTPs

Research on the GHG emissions from WWTPs is not a new topic and has received the attention of a certain number of researchers in many countries. By critically reviewing previous studies, this research has recognized that there are some similarities between studies as well as the challenges faced by previous studies in assessing the GHG emissions coming directly from WWTPs.

❖ **Similarities**

There are three main similarities found during the critical review of previous studies on the GHG emissions from WWTPs, namely neglect of CO_2 emissions, the method of implementation; and GHG emissions trends.

- *The neglect of CO_2 emissions from WWTPs*

The most obvious similarity in previous studies is that it is often overlooked direct CO_2 emissions from WWTPs. According to IPCC Guidelines, 2006, only CH_4 and N_2O emissions from WWTPs are considered. Carbon dioxide (CO_2) emissions from wastewater are not considered in the IPCC Guidelines because these are of biogenic origin and should not be included in national total emissions (IPCC, 2006). Besides, CH_4 and N_2O exist and have a longer-lasting impact than CO_2 in the atmosphere. Therefore, previous studies have often focused on CH_4 and N_2O emissions from WWTPs.

Only a few studies have considered CO_2 generated from WWTPs such as the studies of *Bao et al, 2015; Yan et al, 2014; Schneider et al, 2015; Kyung et al, 2015;* and *Ren et al, 2015*. These studies have substantiated that the amount of CO_2 generated from the aerobic zone during secondary treatment of the WWTPs is relatively significant.

Since CO_2 emissions from WWTPs are not considered, the actual total amount of GHG emissions generated from WWTPs could be underestimated.

- *Methods of implementation*

The methods for estimating the direct GHG emissions from WWTPs are described in Chapter 3 of the report. Among the above methods, the chamber-based methods are the most popular methods chosen by the majority of researchers because the methods are able to apply to quantify CO_2, CH_4, and N_2O emissions from WWTPs. Xiao et al., 2016 cited in Nguyen et al, 2019 mentioned that the static floating chamber method was widely used to evaluate the direct GHG emissions because of its convenience and low cost. The static chamber with gas chromatography is a simple and economic method used to analyses for the GHG concentrations in the lab.

- *GHG emissions trends*

Although previous studies were conducted at various WWTPs, under different types of technologies and operating conditions, and results showing different amounts of GHG emissions, the tendency in GHG emissions at WWTPs are quite similar. For instance, most research results show that the direct GHG emissions are generated mainly from the biological treatment process. In the secondary stage, the biological reactions occur breaking down pollutants in the wastewater and removing them from the wastewater. The nitrification and denitrification processes in this step generate all three main GHGs, namely CO_2, CH_4, and N_2O, of which N_2O is emitting the most in this stage. This is also mentioned in the IPCC guidance, 2006.

For the direct CH_4 emissions, the sludge treatment process in the WWTPs generates the largest amount of CH_4 emissions, accounting for the majority of the total emissions generated from the WWTP itself.

❖ **Difficulties**

Assessment of the Scope 1 GHG emissions from WWTPs is a huge challenge for researchers. During conducting research, difficulties that researchers face include the uncertainties of the methods, the influential factors in wastewater; and accuracy and lack of information.

- *The uncertainties*

As discussed in Chapter 3 of the report, all the methods that could be applied to the quantification of GHG emissions have uncertainties. For estimating CH_4 emissions, for example, the largest uncertainties of the tracer release method come from the collocation assumption of the signals and the baseline estimates, while the injected airflow measurement and water level in the chamber are two main uncertainties of open- and close-chamber methods, respectively (Kwok et al, 2015).

The IPCC guidance, 2006 provides the emission factors (EFs) that are applied by default to most countries when country-specific EFs are unavailable. The EFs are important factors affecting the total GHG emissions from WWTPs. Estimation based on emission factors can be high uncertainty due to the lack of reliable information on the operation of the treatment process and the local environmental situation (Noyola et al., 2018 cited in Nguyen et al, 2019).

- *Influential factors*

Numerous previous research on direct GHG emissions from WWTPs have proved that the concentration of DO, COD/N ratio, pH, aeration ratio, dissolved CO_2, CH_4, and N_2O in wastewater are the influent factors that have profound impacts on the amount of GHG emissions from the WWTPs themselves. Of which, the concentration of DO is one of the most important parameters when controlling GHG emissions released from WWTPs. A low concentration of DO limits the growth of microorganism while high concentration could influence the denitrification process (Nguyen et al, 2019). If the concentration of DO and aeration rate decrease, the quantities of N_2O and CH_4 emissions increase, while the figure for CO_2 emissions decreases.

In addition to the factors that come from the treatment itself, indirect emission factors can also affect the calculation of GHG emitted directly from WWTPs. The biological process in WWTPs requires very high energy consumption which is linked to the amount of CO_2 emitted when generating electricity at a power plant. The biological wastewater treatment accounts for about 73.9% from total power consumption for wastewater treatment (Presura and Robescu, 2017). Besides, the use of chemicals for phosphate precipitation and chemical production also produces GHGs. The indirect

GHG emissions from WWTPs may make researchers misunderstand and miscalculate the amounts of direct GHG emissions.

- *The accuracy and lack of information*

Data accuracy is of paramount importance in any calculation. Applying default data in any country following the IPCC guidelines can lead to large errors as WWTPs often have different capacities, technologies, and operations conditions depending on the quality standards of effluent wastewater in each country. In the UK, although country-specific guidelines have been developed, the EFs for the water sector has not been updated since 2012.

The lack of data greatly affects the ability to estimate the total quantity of the direct GHG emissions from WWTPs. Due to the developments in wastewater treatment technologies and new policies related to wastewater quality, the reduction of GHG emissions schemes, the amount of GHG emission from WWTPs could increase or decrease. Therefore, it is essential to regularly update data and carry out more studies related to direct GHG emissions from WWTPs in the UK particularly and in the world generally.

4.4. GHG emission mitigation methods

In the context of increasing climate change forcing all sectors to have plans to reduce GHG emissions, the water sector is no exception.

To reduce the number of GHGs generated directly from WWTPs, several measures have been proposed to minimize GHG emissions generated from the treatment process itself. These include source control, technological solutions, GHGs collection and treatment, and energy recovery. Also, the water management agencies could adopt new policies, plans, and strategies reducing GHG emissions, for example, the UK net zero-2030 routemap.

- **Source control**

Source control solution is an indirect approach to help reduce the amount of GHG emissions coming from WWTPs. Controlling wastewater at the source can bring down the amount of wastewater generated as well as reduce the concentration of pollutants in the wastewater. As a result, the number of GHG emissions generated both directly and

indirectly by WWTPs can be cut down because they do not have to treat large amounts of wastewater and reduce the number of chemicals and energy consumption.

Although this solution is difficult to control and achieve, it should also be considered by lawmakers and educational institutions to encourage people to use water economically.

- **Technological solutions**

Technology solutions are considered feasible because of their versatility. For new treatment plants, it is important to select wastewater treatment technologies which meet emissions standards, have low GHG emissions, and appropriate to site-specific and economic conditions. Although several advanced treatment technologies can treat wastewater well, they might still be high GHG emissions. Current understanding of end-of-pipe treatment options shows significant variability in the difference in embodied and operational carbon across treatment techniques (Georges et al, 2009).

- *New wastewater treatment plants*

A viable technology proposition for new domestic wastewater treatment plants is trickling filters (see Figure 10). The trickling filter process is a biological treatment method based on biological breakdown for the removal of pollutants in wastewater. Wastewater is trickled over a filter bed to which the biomass is attached with the effluent leaving the bed via an underdrain before passing into a sedimentation tank (Georges et al, 2009). The bacteria and microorganisms attach to the media surface of the trickling filter and feed on contaminants in the wastewater. Taking advantage of the biological oxidation of pollutants, the bacteria and microorganisms then use oxygen in the air to convert the pollutants into harmless by-products. The media can use rock, stone, coke or plastic, of which plastic media is an advanced design with high performance treating wastewater meeting the stringent standards.

This technology has noticeable advantages of being able to limit both direct and indirect GHG emissions from WWTPs. For direct GHGs, during the treatment process itself, it limits denitrification on the filters which highly links to N_2O emissions. Also, CH_4 tends not to be formed in the aerobic environment (Georges et al, 2009). As a result, it could reduce the direct N_2O and CH_4 emissions. For indirect GHGs, the energy demand of the filters is relatively low leading to fewer CO_2 emissions in energy generation.

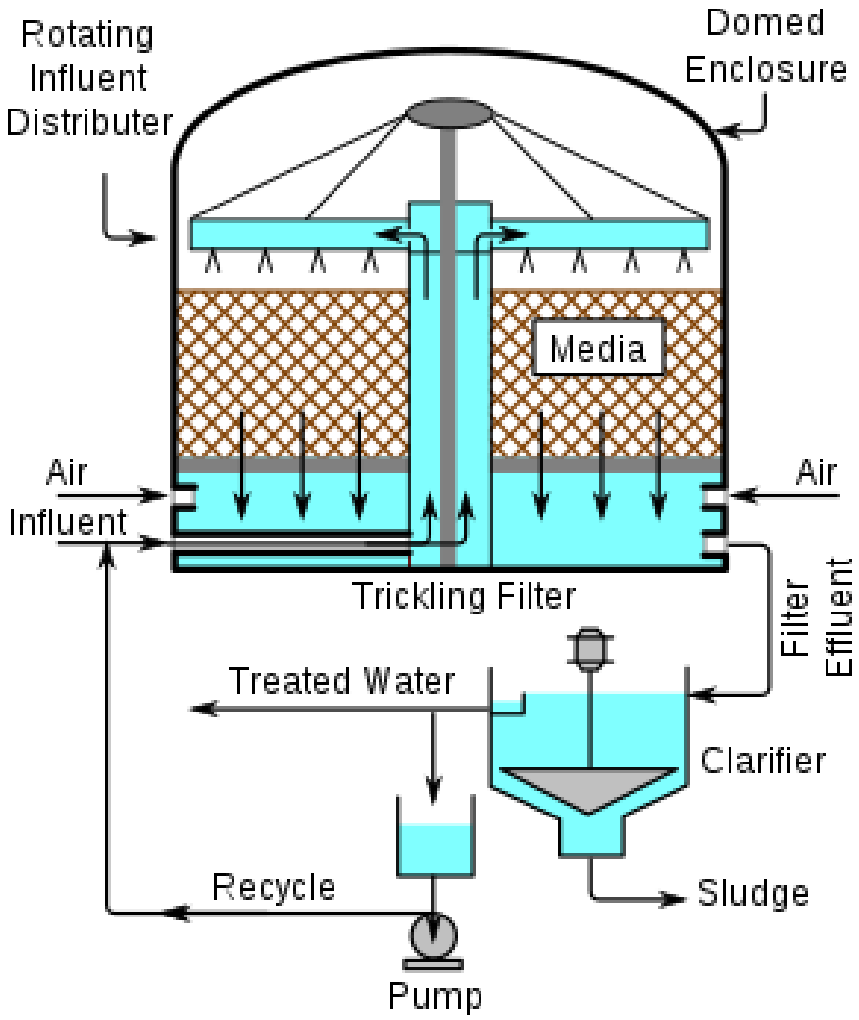

Figure 10: Trickling filter system

(Mbeychok, 2007)

- *Operated wastewater treatment plants*

For wastewater treatment plants already in operation, changing to new technologies may not be an economic option. To limit direct GHG generation from the treatment process, changing operating conditions may be a viable option.

In order to determine the operating conditions that help minimize direct GHG emissions, it is necessary to understand the influent factors. For example, the low concentration of DO, low COD/N ratio, pH, high dissolved GHGs are factors that have been proved to affect direct CO_2, N_2O, and CH_4 emissions by several previous research. Therefore, during operation, each WWTP needs to calculate and adjust operating parameters such as solid retention time in the bioreactor to match the capacity and operating conditions.

- **GHGs collection and treatment**

Collecting the GHGs generated by WWTPs and treating them could also be a possible solution. GHGs arising directly from the wastewater treatment process can be absorbed by absorbent materials or membranes, or bio-filters. Depending on each type of GHGs, it could choose different methods to remove them. This solution, while helping to partially reduce direct GHG emissions from WWTPs, may increases construction, operation, and maintenance costs.

- **Energy recovery**

Methane is a powerful greenhouse gas and a major component in natural gas, which can be captured and converted to heat or electricity. The biological treatment process and sludge treatment of wastewater treatment plants generate a large amount of CH_4. If it is collected and converted into energy, it can help plants reduce the overall total amount of GHGs generated, saving energy as well as operating costs.

- **The UK net zero 2030 routemap**

In the UK, each tonne of GHG emissions as carbon dioxide equivalent (CO_2eq) costs £26 following the Defra Shadow Price of Carbon guidance in 2008 (Reffold et al, 2008), which meant each year, it would cost the UK water industry billions of pounds for the GHG emissions. With the importance of climate change in mind, the UK water sector has made the commitment to achieve net-zero carbon emissions by 2030 and developed the net-zero 2030 routemap to help support its transition to a lower emissions future, building on the work that it is already doing, and enabling an acceleration and step change in decarbonisation (Ricardo PLC and Mott MacDonald Group, 2019).

Through this routemap, the water industry in the UK gradually develops plans to minimize emissions by combining technological, technical, and management solutions. Although the net-zero route does not mean there will be no GHG emissions, it will help minimize the number of GHGs emitted from the water industry, and implement measures to return the amount of GHGs generated by the UK water industry. This also shows the sense of responsibility of the water sector in the UK for the global problem and helps to direct water-related companies and businesses in the UK to apply measures to mitigate the GHG emissions from their activities.

4.5. Summary of findings

Through the critical review, the report discussed the GHG emissions in each stage of the wastewater treatment process and developed a diagram of the GHG emissions directly from the WWTPs. The report analysed the impact of GHG emissions on the environment, assessed the similarities as well as the challenges that studies are facing during implementation. The report also proposed a few measures that can be applied to reduce GHG emissions generated directly from WWTPs, the development direction of the water sector with a commitment to reducing greenhouse gas in the future.

CHAPTER 5: CONCLUSION AND FUTURE WORK RECOMMENDATIONS

5.1. Conclusion

The water industry plays an important role in life as its activities help ensure the quality of life and also protect the environment. The WWTPs are a part of the water industry that help to treat pollutants in wastewater, ensuring the quality of wastewater before being discharged into the water environment. However, the wastewater treatment process generates both direct and indirect GHGs that profoundly affect the air quality.

This report has made "*Assessment of GHG emissions coming from WWTPs*" in the boundary of the Scope 1 emissions, direct GHG emissions from the plant itself. The report has the literature review of the research-related documents in chapter 2 of the report as a basis for confirming the findings in the result and discussion chapter. By critical review for data collection and analysis, the report has generally answered research questions.

Question 1: *Where do greenhouse gas emissions come from in WWTPs?*

The report has basically built up a general layout of a WWTP with direct GHG emissions in Figure 8. Also, the report analysed the direct GHG emissions in each step of the treatment process. The data related to direct GHG emissions in each stage from WWTPs are summarized in Table 8. Although some emission data for the tertiary treatment and sludge treatment were missing, it still can be seen that the direct GHG emissions generated from all stages of wastewater treatment, from pre-treatment until tertiary treatment and sludge treatment. Particularly, the secondary treatment in both the AAO and SBR technologies considered in the study emits a very significant amount of GHGs. The AAO process has total emission rates of CO_2 with 3,046.92 kg h^{-1}, CH_4 with 8,812 kg h^{-1}, and N_2O with 407.45 kg h^{-1}, while the figures for the SBR process are 2,044.21 kg h^{-1} of CO_2; 5,306 kg h^{-1} of CH_4 and 14,135 kg h^{-1} of N_2O.

Through the critical review, the report found that the biological treatment process in the secondary treatment step and the sludge treatment process are the two biggest GHGs emission sources. While the AAO technology emits the GHGs mainly in the oxic zone, the SBR technology generates the largest emissions in the feeding and aeration phases.

The sludge treatment process is the largest methane sink in the WWTPs up to 81% of the total methane emissions from the plant itself.

Question 2: *What are the possible consequences and impact of these emissions?*

The GHG emissions from WWTPs negatively affect the air environment and are responsible for global warming and climate change.

The amount of CO_2 emissions generated in the preliminary, primary and secondary treatment phases are very large. Although the GWP of CO_2 is just 1, with a large amount of direct CO_2 emissions from WWTPs, it can still have significant impacts on the air environment. The CH_4 is generated in all treatment processing stages of both the water line and sludge line. In which, the CH_4 emissions in the sludge line account for up to 81% of the total CH_4 generated from WWTPs. With a GWP of 34 CO_2eq, the amount of CH_4 emissions will absorb more heat many times than that of CO_2. The N_2O is a powerful gas with a GWP of 268-298 CO_2eq and emitted mainly in the secondary treatment stage with 407 kg h^{-1} in the AAO process and 14,135 kg h^{-1} in the SBR process. Once the N_2O is released into the atmosphere, it will last a long time and trap a large amount of heat in the atmosphere.

Previous studies are sufficient to prove that the direct GHG emissions from WWTPs are significant, and have a similar view on the trend of direct GHG emissions from WWTPs. Additionally, those research have similar implementation methods, and most of them ignore direct CO_2 emissions. Moreover, the assessment of GHG emissions from WWTPs encounters difficulties such as uncertainties, influent factors, and the accuracy and lack of data and information, which can cause the estimation of direct GHG emissions from WWTPs to be underestimated.

Question 3: *What must we do to reduce greenhouse gas emissions from the water industry sector in order to reach net-zero carbon emissions?*

The report has presented and discussed some possible solutions to reduce the direct GHG emissions from WWTPs including source control, technological measures, GHG collection and treatment, energy recovery, and the UK net zero 2030 routemap. Of which, the technological measures are highly feasible, significantly reducing the number of GHG emissions directly from the wastewater treatment process. Source control is the

indirect solution that is difficult to control and achieve; however, it should also be considered for inclusion in educational programs. The UK net zero 2030 routemap of the water sector is an integrated mitigation measure that helps guide companies and businesses to adopt greenhouse gas mitigation measures appropriate to their operating and business conditions.

The operation of wastewater treatment plants not only needs to treat pollutants in wastewater but also must ensure that the impacts on the air environment are limited. Therefore, the application of GHGs mitigation measures is essential.

5.2. Future work recommendations

Although the report partly assessed the GHG emissions directly from WWTPs, it still has certain limitations and needs to conduct more research in the future. Here are some suggestions for future studies to help improve the assessment of GHGs from WWTPs as well as the direction to develop better solutions to mitigate the GHG emissions of the water sector.

1. Research on evaluation of the amount of greenhouse gas generated at the influent wastewater of WWTPs.
2. Assessment of CO_2 and some other GHGs emissions from WWTPs.
3. Research on technological measures to reduce GHG emissions directly from WWTPs.
4. Assessment or comparison of the number of GHG emissions for different types of wastewater treatment technologies such as MBBR and MBR technologies.
5. Research on indirect solutions such as policies, plans, and actions to help reduce GHGs from WWTPs.
6. Applying models to forecast the number of GHG emissions coming from WWTPs.

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