NITROGEN MANAGEMENT IN WASTEWATER INDUSTRY FROM CIRCULAR ECONOMY PERSPECTIVE

MATTHIAS CHAI ZHENG FENG

A project report submitted in partial fulfilment of the requirements for the award of Bachelor of Engineering (Honours) Chemical Engineering

Lee Kong Chian Faculty of Engineering and Science
Universiti Tunku Abdul Rahman

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DECLARATION

I hereby declare that this project report is based on my original work except for citations and quotations which have been duly acknowledged. I also declare that it has not been previously and concurrently submitted for any other degree or award at UTAR or other institutions.

Signature : 

Name : Matthias Chai Zheng Feng

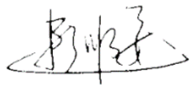
ID No. : 1703190

Date : 16th May 2022

APPROVAL FOR SUBMISSION

I certify that this project report entitled **"NITROGEN MANAGEMENT IN WASTEWATER INDUSTRY FROM CIRCULAR ECONOMY PERSPECTIVE"** was prepared by **MATTHIAS CHAI ZHENG FENG** has met the required standard for submission in partial fulfilment of the requirements for the award of Bachelor of Engineering (Honours) Chemical Engineering at Universiti Tunku Abdul Rahman.

Approved by,

Signature : 

Supervisor : Dr. Lai Soon Onn

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ABSTRACT

The municipal wastewater industry is known as a resourceful hub as it contains large amount of valuable nitrogen that can be recovered. However, the operation of conventional nitrogen management in the municipal wastewater industry is by nitrogen removal, mainly through biological nitrogen removal (BNR) processes. The downside of BNR processes includes high energy and cost requirement, and the emission of greenhouse gases (GHGs) only served the purpose of meeting the standard discharge limit of nitrogen. Besides, the sole reliance on energy-intensive Haber-Bosch process to produce synthetic fertilizers triggered the eminent nitrogen recovery process. Therefore, existing nitrogen recovery pathways were explored in the municipal wastewater industry for the production of useful bioproducts. The pathways reviewed in this project included bio-membrane integrated systems (BMISs), capacitive deionization (CDI), coupled aerobic-anoxic decomposition operation (CANDO) process, ammonium recovery through adsorbents, stripping and struvite precipitation, and through the usage of microalgae and cyanobacteria. The feasibility and challenges of its implementation were explored, in which low-strength of municipal wastewater being the major challenge causing economical and technical infeasibility in large-scale execution. Amongst the pathways reviewed, the BMISs, FCDI and stripping and struvite precipitation were discovered to be the potential pathways for large-scale application. Therefore, more intensive research and development should be conducted for these recovery technologies for the techno-economic viability, coupled with other initiatives such as governmental incentives and transparent communication between involved parties that includes local farmers, research scientists, stakeholders and consumer for the successful implementation of nitrogen recovery pathways.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF SYMBOLS / ABBREVIATIONS

AEM	Anion Exchange Membrane
AnMBR	Anaerobic Membrane Bioreactor
AnNFMBR	Anaerobic Nanofiltration Membrane Bioreactor
AOB	Aerobic Oxidising Bacetria
BES	Bioelectrochemical system
BMIS	Bio-membrane Integrated System
BNR	Biological Nitrogen Removal
CANDO	Coupled Aerobic-Anoxic Decomposition Operation
CDI	Capacitive Deionization
CEM	Cation Exchange Membrane
COD	Chemical Oxygen Demand
DON	Dissolve Organic Nitrogen
ECOA	Electrochemical Oxidation
ED	Electrodialysis
EDR	Electrodialysis Reversal
FCDI	Flow Capacitive Deionization
FO	Forward Osmosis
GHG	Greenhouse Gas
IEM	Ion Exchange Membrane
MEC	Microbial Electrolytic Cell
MFC	Microbial Fuel Cell
MD	Membrane Distillation
MPBR	Membrane Photobioreactor
N_2O	Nitrous Oxide
NF	Nanofiltration
NH_3	Ammonia
NH^{4+}	Ammonium Ion
NO_2^-	Nitrite Ion
NO_3^{2-}	Nitrate Ion
NOAA	National Oceanica and Atmospheric Administration
NOB	Nitrifying Oxidising Bacteria
OMBR	Osmotic Membrane Bioreactor

RO	Reverse Osmosis
TKN	Total Kjeldahl Nitrogen
TN	Total Nitrogen
US EPA	United State of Environmental Protection Agency
WWTP	Wastewater Treatment Plant

CHAPTER 1

INTRODUCTION

1.1 General Introduction

The increasing demand for nitrogen sources over the years had exerted pressure on the food supply as a result of estimated continual increase of world population from eight to ten billion by 2050 (van der Hoek, Duijff and Reinstra, 2018). Besides, other factors such as climate change, the process of urbanization and the substantial depletion of natural resources have forced the society to prepare for uncertain availability of nitrogen sources around the world (Shaddel et al., 2019).

The main usage of nitrogen sources is to manufacture nitrogen-based fertilisers from the Haber-Bosch process through the nitrogen fixation process. Besides, other application of nitrogen sources includes the growth of algae and bacteria in the biogas industry to become food source for humans and animals (Beckinghausen, et al., 2020). Thus, the wastewater industry plays a crucial role for the recovery and transformation of nitrogen sources into valuables products. According to Shaddel et al. (2019), there were key drivers in place from the legal, environmental and economic sectors for the furtherance of the recovery of nitrogen sources from the wastewater sources.

Although nitrogen is widely available in the atmospheric air, it exists in its non-reactive and stable form of nitrogen (N_2) gas. Nitrogen in its reactive forms can only be uptake by plants for growth and development which can be derived from lighting (2 %) and biological nitrogen-fixation (98 %) (van der Hoek, Duijff and Reinstra, 2018). The introduction of Haber-Bosch process in the 1909 have revolutionised the agriculture industry as nitrogen can be industrially fixed to form fertilisers that enabled the substantial increase on the productivity of crops by four times. The increased food production by nitrogen-based fertilisers were converted into urea and ammonium ions, NH_4^+ through human metabolic activity and discharged into the wastewater industry.

The common practice of the wastewater industry is through nitrogenous compounds removal by converting into nitrogen gas and releasing it into the atmosphere that brings a closure to the nitrogen cycle. However, it posed significant drawbacks towards the environment. Firstly, converted atmospheric nitrogen is lost into the atmosphere rather than being reused. Secondly, the process of converting into nitrogen gas is energy intensive. Thirdly, potent GHG of nitrous oxide are emitted in the removal process (van der Hoek, Duijff and Reinstra, 2018). Therefore, the consideration of nitrogen recovery in the wastewater industry should be highlighted as it can potentially be transformed from the current linear economy to the circular economy concept. Through the new approach of circular economy, the wastewater industry seeks a more sustainable treatment process to transform the conventional wastewater industry into a resourceful hub of nutrients (Noriega-hevia et al., 2021).

1.2 Importance of the Study

The wastewater industry acts as a resourceful hub for nutrients such as nitrogen which is in increasing demand with the growing world population that exerted its pressure on the food production. The opportunities presented in the wastewater industry for its nitrogen management through a circular economy help fill the gap of the current practice of nitrogen removal. Besides, the current practice of nitrogen removal does not only pose environmental drawbacks, but only serves the purpose of meeting the discharge standards. In addition, the current Haber-Bosch process for the manufacture of nitrogen-based fertilisers is not cost effective, and is unable to keep up with the growing needs of food production. Therefore, the nitrogen management in the wastewater industry from a circular economy perspective is provided in this study to fulfil the growing needs on the food supply and thus revolutionizing the wastewater industry as a resourceful centre for nitrogen.

1.3 Problem Statement

The growing world population in need of food supply, environmental drawbacks of the wastewater industry and its potential to act as a resourceful source for nitrogen have led to the contribution of this study. The current Haber-Bosch industry that supplies the nitrogen-based fertilisers for the growing of crops requires large amounts of energy. An estimated yearly energy consumption of 6.4×10^{12} MJ is required, which is approximated to the usage of energy by 80,000,000 people from a global warming perspective (Noriega-hevia et al., 2021). In addition, an estimated 949 m^3 of natural gas is required coupled with the emission of 1,600 kg of carbon dioxide to produce 1,000 kg of anhydrous ammonia fertilisers have led to idea of nitrogen recovery from the wastewater industry due to its environmental cost and increasing cost of fertilisers around the world (Beckinghausen et al., 2020). Thus, this acts as a catalyst for the implementation of nitrogen recovery from the nutrient-enriched wastewater from the industrial or municipal wastewater so as to prevent valuables from entering into the water bodies in a linear economy pathway.

1.4 Aim and Objectives

This project aimed to investigate the nitrogen management in the wastewater industry from the circular economy perspective. The objectives of the project were listed such as follows:

1. To investigate the current nitrogen management in the municipal wastewater industry
2. To identify the potential nitrogen recovery technologies/pathways in the municipal wastewater industry
3. To analyse the feasibility of the nitrogen recovering technologies in the municipal wastewater industry
4. To identify the challenges for the implementation of circular economy on nitrogen management in the municipal wastewater industry

1.5 Scope and Limitation of the Study

The scope of this study encapsulated the current nitrogen management in the wastewater industry and the different routes for the nitrogen recovery pathways through the circular economy approach. The scope extended to the study of the opportunities or sustainability gaps in the conventional practice of nitrogen management in the wastewater industry. In order to explore the different technologies applied in the wastewater industry for the recovery of nitrogen in a realistic approach, the present and future challenges in its implementation were also reviewed as part of the scope. The limitation of the study includes the actual feasibility of the proposed technologies for the nitrogen recovery on a real production plant due to several factors, such as the advancement of technologies available in the region and the different types of wastewaters that entered into the wastewater industry. Besides, the study only incorporated the nitrogen recovery technologies that were feasible in the municipal wastewater industry and not into any specific industrial wastewater industry. Therefore, the focus of this study presented the nitrogen recovery pathway in a conventional municipal wastewater treatment plant.

1.6 Contribution of this Study

This study offered potential feasible technologies in the municipal wastewater industry for the nitrogen recovery from a circular economy approach which is in rising demand. Therefore, the current problems that the wastewater industry brings were tackled and insights into the refurbishment of facilities for the nitrogen recovery pathway were provided. The continuous growing demand of food supply in the agricultural industry that relied on nitrogen-based fertiliser have caused the wastewater industry to take initiatives to recover valuable resources from the enriched-wastewater. This contributed by and large to all the municipal wastewater industry on the potential development of technologies to achieve the circular economy. Lastly, this study also highlighted the present and future prospect of the maturing technologies.

1.7 Outline of the Report

The overall outline of the report were divided into six sections as follows:

Chapter 1: Introduction

Overview on the purpose of the report was expounded, which included: (1) brief introduction of the topic, (2) importance of the study, (3) problem statements, (4) aim and objectives, (5) scope and limitation, and (6) its contribution.

Chapter 2: Literature Review

Overview of nitrogen, overview of the wastewater industry, nitrogen management in wastewater industry, conventional and current nitrogen removal in wastewater industry and the overview of circular economy were focused.

Chapter 3: Methodology and Workplan

The methodology for conducting the research in order to achieve its aim and objectives were highlighted.

Chapter 4: Discussion/Results

The research outcomes and discussion by analysing the different types of technologies available for nitrogen recovery in the municipal wastewater industry were being expounded.

Chapter 5: Conclusion and Recommendations

The conclusion of the research that navigate the reader on the potentiality of the technologies discussed earlier and recommendations to undertake for its application into the municipal wastewater industry were looked into.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

In order to clearly establish the nitrogen management in wastewater industry from the concept of circular economy, a preliminary background research was conducted to understand the current management of nitrogen. While there had been much research on the removal of nitrogen from the wastewater industry, few researchers had undertaken the research process on the recovery of nitrogen. The nitrogen recovery pathway that complimented the concept of circular economy was expected to cause an overturn of the conventional linear economy of "take-make-dispose". Thus, the focus of this literature review was to grant a holistic view on the current approaches of nitrogen management in the wastewater industry by exploring the overview of nitrogen and wastewater industry, nitrogen management in wastewater industry, conventional and current nitrogen removal methods in wastewater industry as well as the overview of circular economy. The main purpose was to shed light on the importance of introducing the concept of biocircular economy in managing nitrogen resource in the wastewater industry.

2.2 Overview of Nitrogen

It was of utmost important to lay down the fundamentals in regard to the topic, such as the properties of nitrogen and its various forms, the nitrogen cycle, the application of nitrogen in various industries. By exploring the different aspects on the overview of nitrogen, the emphasis on the importance of nitrogen management in the wastewater industry was highlighted.

2.2.1 Properties of Nitrogen and its Various Forms

According to Connor (2020), nitrogen is an element that can be found in the periodic table as a gas whose atom consists of 7 electrons, 7 protons and 7 neutrons and commonly exists as an inert diatomic gas at standard temperature and pressure. Nitrogen gas is also widely available in the atmosphere as it consists of 78 % in the air (Connor, 2020). Several physico-chemical properties of nitrogen gas were summarised and tabulated in Table 2.1 as follows:

Table 2.1: Physico-Chemical Properties of Nitrogen Gas (Connor, 2020)

Properties	**Description**
Element Category	Non-Metal
Phase (25 ℃, 1 atm)	Gas
Odour	Odourless
Colour	Colourless
Atomic Mass [amu]	14.0067
Density (25 ℃,1 atm) [g/cm^3]	1.251
Melting Point (℃)	-209.9
Boiling Point (℃)	-195.8

According to Killpack and Buccholz (2021), nitrogen acts as the source of life for all plants and it comes with various forms for its specific usage in soils and plants. Although nitrogen is abundantly available in the atmosphere, it needs to be converted into usable forms from its original state which is nutritionally unavailable. This resulted in the inability of plants to assimilate the existing inorganic form of nitrogen which needs to be converted into organic forms such as ammonia, nitrates and nitrites to be nutritionally available (Science Struck, 2021). The different forms of nitrogen as well as its specific uses for soils and plants were being highlighted in Table 2.2.

Table 2.2: Various Forms of Nitrogen and its Uses in Soil and Plants (Killpack and Buccholz, 2021)

Nitrogen Forms	Symbol	Uses in Soils and Plants
Atmospheric Nitrogen	N_2	It can be taken up by minority of legume plants through nitrogen fixation process by certain bacteria, algae, lightning etc.
Nitrate	NO_3^{2-}	It is the most used forms of nitrogen by plants for growth and development. It can easily be lost from soils into the groundwater.
Ammonium Nitrogen	NH_4^+	Taken directly into plants from soil for protein building. It is not easily lost from the soil.
Organic Nitrogen	C-NH_2 (C represents a complex organic group)	It exists in various forms. Able to convert into nitrate and ammonium nitrogen which can be used in plants.

2.2.2 Nitrogen Cycle

While there are only limited number of pathways nitrogen can be converted into its reactive forms, the majority of it is fixed biologically by a small group of microbes by carrying out enzymatic activities to convert the dinitrogen gas into ammonia (Holmes, Dang and Smith, 2019). The remaining non-biological fixing of nitrogen pathway consists of lightning, combustion, industrial fixation as well as the burning of biomass. Through the formation of lightning, it breaks the triple bonds of the dinitrogen gas and combines with the oxygen present in the atmosphere to produce nitrogen dioxide, NO_2 which in turns dissolves in rainwater to form nitric acid, HNO_3. The acidified rainwater then falls onto the earth through rain, snow, or the formation of fail which then can be utilised by living organisms as it is now converted into reactive inorganic nitrogen sources.

Besides relying on natural phenomenon for the production of reactive nitrogen forms, human activities also contributed to the nitrogen fixation process according to Galloway et al. (2013). In the manufacturing of plants fertilisers, extreme operating condition of elevated temperature and pressure through an iron-based catalyst is required to be undergone by raw materials of nitrogen and hydrogen gases for ammonia production. Other human activities such as the combustion of biomass and the clearance of forests and grasslands in the agriculture industry have also contributed to the generation of reactive nitrogen (Holmes, Dang and Smith, 2019).

After the conversion of inert dinitrogen gas, plants will uptake the reactive forms of nitrogen to produce proteins and nucleic acids. Ammonia cannot be utilised directly by plants and thus needed to be converted into nitrates by nitrifying bacterias. The nitrification pathway eventually processes the ammonia or ammonium ions into nitrate ions by a two-step process which can be utilised by plants for growth and development. Lastly, the decomposition of living organisms by certain group of microbes aids the conversion of the available organic nitrogen sources into ammonium through mineralization. This process acts to transform the inorganic into organic nitrogen forms for the reintroduction of nitrogen in the nitrogen cycle. Another way of undergoing denitrification or anaerobic ammonium oxidation (anammox) pathways also is to convert the reactive nitrogen into dinitrogen gas and released into the atmosphere (Holmes, Dang and Smith, 2019). The illustration of the entire process of nitrogen cycle in a holistic view was shown in Figure 2.1.

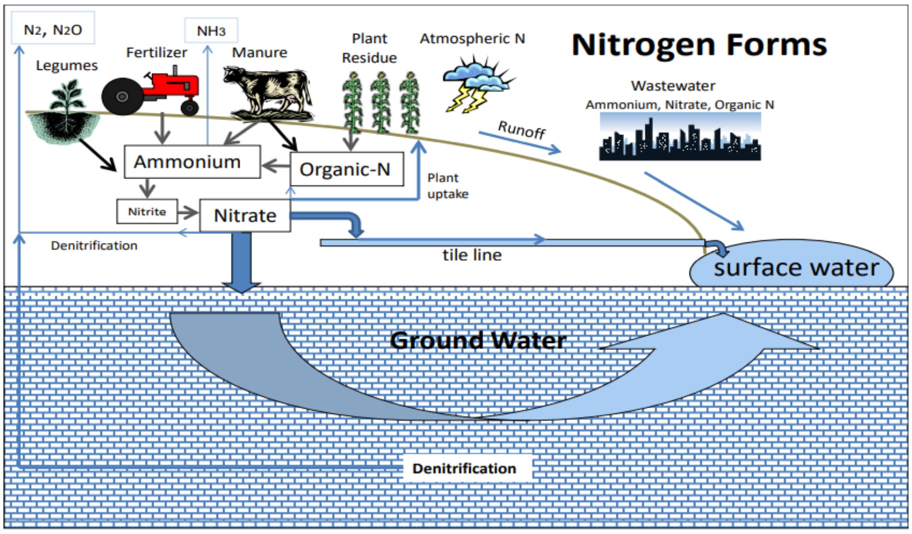

Figure 2.1: Nitrogen Cycle (Wall, 2013)

2.2.3 Application of Nitrogen in Various Inudstries

Nitrogen is important for the survival of humanity and all living organisms on earth. Such as an example was that nitrogen is used as an active ingredient in the manufacturing of fertilisers for the growth of plants and its supply (Hoek, Duijff and Reinstra, 2018). They further claimed that the nitrogen-based fertilisers can be manufactured from the Haber-Bosch process where the process of nitrogen-fixation was being carried out to be converted into ammonia. According to Science Struck (2021), the main two raw materials needed for the Haber-Bosch process were hydrogen (H_2) and nitrogen (N_2) gases respectively, in which approximately 454 million tons of nitrogen fertilisers were being produced to be used extensively in the agricultural field for increasing crop yields presently. The visual illustration of the conventional Haber-Bosch was shown in Figure 2.2.

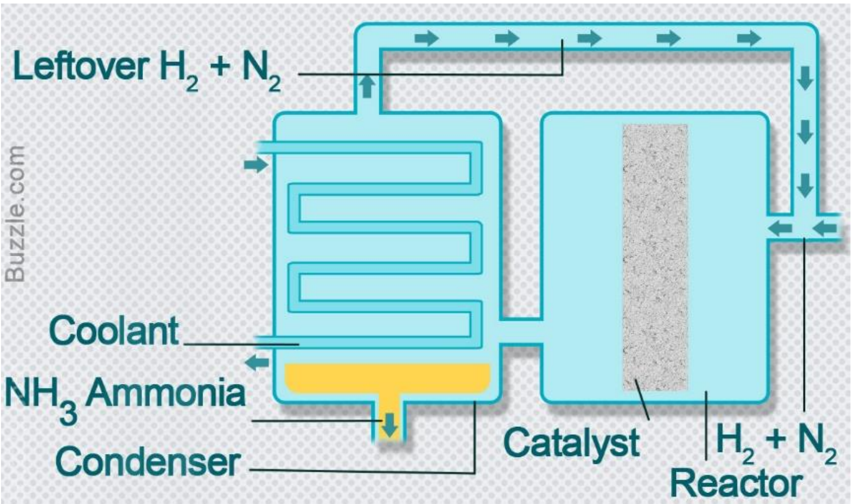

Figure 2.2: Haber-Bosch Process (Science Struck, 2021)

The use of nitrogen was not limited in the agricultural field but across other industrial fields. Nitrogen in the form of nitrogen gas can be used in the food packaging industries where it is used to preserve packaged foods by preventing the occurrence of oxidation in food by displacing oxygen present in the products. In the lighting and blub industry, it is used to replace the more expensive argon gas in the light bulb as a cheaper alternative. In the automobile industry, nitrogen gas is used extensively to fill tires by offering a longer lifespan by reducing the oxidation process. By filling car tyres with nitrogen gas, it is known to further improve the retention of tires pressure which provides better gas mileage to drivers (World of Chemicals, 2021)

In addition, the use of nitrogen gas has also exerted its influence in the stainless-steel manufacturing industries in the process of electroplating. Through the process of electroplating, the stainless-steel product is fortified and more resistant to corrosion. Other than that, nitrogen gas is widely used in the pharmaceutical industry to produce antibiotics. One of the forms of nitrogen, nitrous oxide, is commonly used in anesthetic as well. Lastly, nitrogen gas is used in the effort of pollution control. One of the concerns in the pollution control industries are the presence of volatile organic compounds (VOC) in liquid that acts as GHGs in the atmosphere. Thus, the application of nitrogen gas in removing the VOC present in the liquids acts as one of the remediation methods for pollution control before it is discarded into the landfills (World of Chemicals, 2021).

2.3 Overview of Wastewater Industry

According to US Geological Survey (2021), the term wastewater denotes used water with the inclusion of foreign substances such as human wastes, food scraps, oil, soaps and various chemicals. The wastewater was originated from various places such as homes, businesses, industrial areas and surface runoffs as a result of rain. The wastewater industry plays a very important role as wastewater were collected from various sources to be treated and released back into the environment according to the standard discharge effluent regulations set by local authorities. The presence of wastewater industry does not guarantee a complete prevention on the release of waste into the surroundings, but it reduced the pollutants released into the water bodies to an acceptable level.

According to Riffat (2013), the wastewater engineering has progressed significantly over the recent years. It progresses from the only the collection and open dumping of waste, which proceed to collection and disposal without proper treatment procedure, to collection and treatment before disposal and finally to the revolutionised collection and treatment before reusing it back into usable forms. This progression has definitely led to a greener engineering and chemistry in the wastewater industry.

2.3.1 Sources of Wastewater

There are four common sources of wastewater, which are the municipal wastewater, industrial wastewater, infiltration and inflow, as well as stormwater (Rifat, 2013). Municipal wastewater, also known as domestic wastewater, incorporates discharges from residential areas, institutions, and commercial activities while wastewater generated from the industrial activities originates from industrial processes. The source of infiltration and inflow denotes water that comes from leaking pipes, drains, submerged manholes and groundwater infiltration that eventually enters into the sewage system. Lastly, the stormwater source here refers to water run-offs that comes from rainfall and the melting of snow.

2.3.2 Constituent of Wastewater

In the municipal wastewater, the major components that can be found are suspended solids, organic matters and pathogens (Riffat, 2013). High concentration of elements such as nitrogen are commonly present in the residential wastewater can cause numerous problems relating to the environment and human health. In the industrial wastewater bodies, it contains contaminants such as heavy metals, toxic compounds and refractory organics substances as well as those found in the municipal wastewater. One of the sources of wastewater which is stormwater contains petroleum compounds, silt, and pesticides due to surface runoff from the urban and agricultural regions. The presence of these constituents have led to the possible negative environmental impacts. The major constituents of wastewater and its environmental impacts on the water bodies were highlighted in Table 2.3.

Table 2.3: Pollutants of Wastewater and its Environmental Impact (Riffat, 2013)

Pollutants	**Source**	**Environmental Impact on Water Bodies**
Suspended solids	Municipal wastewater and stormwater	Water surface filled with scum layers; sludge deposits can be found
Organic matter	Municipal and industrial wastewater	Depletion of dissolved oxygen, lead to anaerobic conditions, killing aquatic plants and animals
Nutrients	Municipal and industrial wastewater	Eutrophication and degradation of water quality
Pathogens	Municipal wastewater	Ease of transmission of disease
Heavy metals	Industrial wastewater	Highly toxic to aquatic life
Refractory organics	Industrial wastewater	Toxic or carcinogenic to living orgaisms
Endocrine disrupting compounds	Municipal wastewater	Fish feminization and possible broader scope of impacts

2.3.3 Wastewater Treatment Process

The general wastewater treatment process consists of five successive steps, consisting of the preliminary, primary, secondary, tertiary treatment processes and the final treatment of sludge as shown in Figure 2.3.

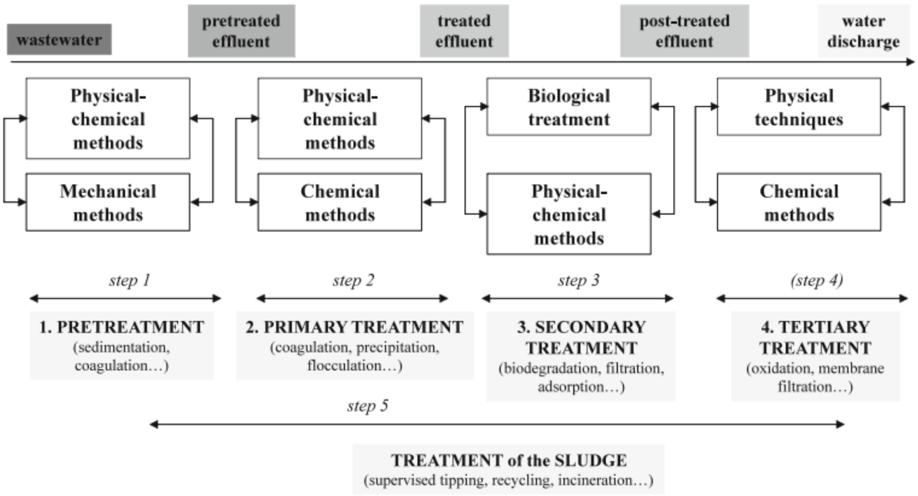

Figure 2.3: Overview of Processes in Wastewater Treatment (Gregorio and Lichtfouse, 2018)

In the pre-treatment process, also known as the preliminary treatment stage, physical and mechanical means are applied to separate the solid particles and suspended substance from the waste effluent. It is carried out by grit removal and screening processes (Skibinski et al., 2015). This stage is important for subsequent steps of treatment as large objects present are able to hinder the process by causing damage of the unit equipment thus rendering it less efficient (Gregorio and Lichtfouse, 2018). After the removal of solid waste in the pre-treatment stage, the primary treatment process commences where several processes such as oxidation for cyanide destruction and chromium reduction, pH adjustment as well as reduction of organic load are conducted. Other processes are included such as coagulation, precipitation and flocculation of wastes (Gregorio and Lichtfouse, 2018). This stage aids the removal of most of the suspended solids by adding coagulants which eventually results in the sludge formation at the bottom of the tank and subsequently pumped to the solids treatment process for complete removal (Skibinski et al., 2015).

In the secondary treatment process, the main functionality is to remove dissolved and suspended organic materials from the effluent by means of a biological processes with the inclusion of microorganisms (Wisconsin Department of Natural Resources, 2011). In this stage, the organic material present in the wastewater will be broken down and consumed by the microbes in an aerated tank for the supply of oxygen requirement. A tertiary treatment is followed by where it functions to remove the remaining pollutants produced from the secondary purification process. However, the inclusion of tertiary treatment is limited in Europe (Gregorio and Lichtfouse, 2018). Some of the technologies involved in this stage uses the activated carbon, ion-exchange, membrane filtration, advanced processes of oxidation as well as constructed wetlands. The last process includes the treatment of the sludge produced from previous stages by means of incineration and recycling such as for the use of fertilisers (Gregorio and Lichtfouse, 2018).

2.3.4 Challenges Faced in Wastewater Industry

There are many challenges encountered in the wastewater industry as the load and composition of wastewater generated from various sources poses a threat to the human and the environment. Hence, it is important to ensure the effective management of wastewater which comprises of the process of collection, treatment, and discharge with the smallest possible hazards (Villarín and Merel, 2020). Due to the vastness of contaminants present in the wastewater, this further increases the challenges for the present-day wastewater industries in order to remove the contaminants before discharging to the environment.

One of the challenges faced in the wastewater industry is the types of chemical contaminants to be removed. According to Villarín and Merel (2020), there are several types of chemical contaminants that poses significant challenges in this field, these include nutrients, pharmaceuticals and care products, biocides, heavy metals, dyes, radionucleocides, as well as transformation products. Besides, they further added that there are also several types of non-chemical contaminants such as microplastics, nanoparticles, and pathogens which are challenging to be treated as well.

With the increasing population in the world, the problem of increasing exploitation of resources by mankind becomes more prevalent. This further challenges the wastewater industry to deal with various environmental pollution issues due to the increasing wastewater generated from diverse sources (Guerra-Rodriguez et al., 2020). A paradigm shift is essential in the wastewater treatment sector in order to resolve this issue which can be achieved through circular economy. Through circular economy, a significant waste reduction can be predicted as the raw materials are recovered and reuse. However, many of the efforts of achieving circular economy in the wastewater industry are far from becoming a reality (Guerra-Rodriguez et al., 2020).

2.4 Nitrogen Management in Wastewater Industry

According to Beckinghausen et al. (2020), the management of nitrogen which practices the recovery of nitrogen should be the improvement made in the current wastewater industry. Nitrogen carries intrinsic value and is considered an important nutrient for the manufacturing of fertilisers in order to decrease the use of energy, petrochemicals and its impacts towards the environment (Beckinghausen et al., 2020). The wastewater industry, by and large, have utilised the customary treatment process which is energy-demanding to remove nitrogen from the wastewaster effluent. Such process gives no additional benefits but simply complying with the effluent concentration limits from the standard discahrge set by local authorities. Thus, the process of recovering nitrogen as a result of nitrogen management in the wastewater industry allows the simultaneous treatment of wastewater while giving rise to benefits such as the collection of concentrated ammonia products in order to create a circular economy solution (Beckinghausen et al., 2020).

According to Sánchez and Martins (2021), nitrogen and phosphorus can be managed and recovered from the wastewater and be used as fertilisers. This is because urban wastewater contains rich amount of these nutrients which is suitable to be used for plant growth. For example, a total load of 80 % of nitrogen and 45 % of the total load of phosphorus can be found in urine in the wastewater. Thus, the effort of recovering such nutrients as a form of management in the wastewater industries is able to reduce the need for the manufuring of industrial fertilisers as the production process is energy-intensive and depending on non-renewable sources of minerals.

2.4.1 Forms of Nitrogen in Wastewater Industry

According to Wall (2013), there are various forms of nitrogen entering into water bodies which consists of inorganic and organic nitrogen. Inorganic forms of nitrogen in water bodies consists of ammonia, ammonium, nitrate and nitrite while organic nitrogen derived from proteins, amino acids, urea, living or dead organisms as well as plant material as a result of decay. There are two categories of nitrogen which can be known in the wastewater industry, known as the Total Kjeldahl Nitrogen (TKN) that measures the combination of organic nitrogen with the addition of ammonia and ammonium and Total Nitrogen (TN) which measures the totality of nitrogen which includes every form of nitrogen present in the water. It is known that the amount of various forms of nitrogen are dependent on a number of factors, which includes the distance from the point and non-point pollution sources, influence of discharged groundwater into water bodies, amount and types of wetlands, presence of intersecting reservoirs and lakes to the stream and other natural and human-causing factors. The various forms of nitrogen present in the wastewater industry were shown in Figure 2.4.

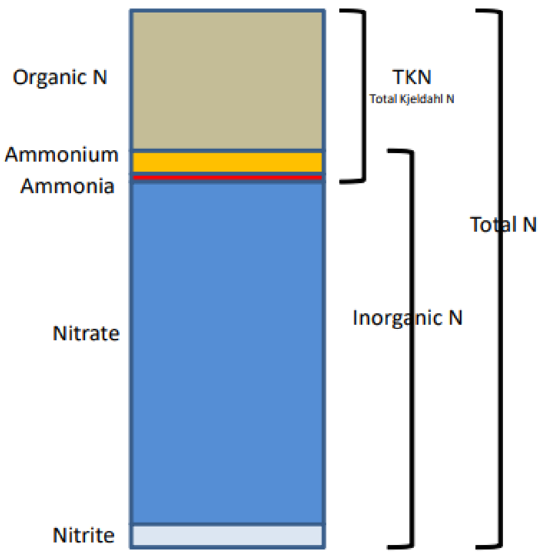

Figure 2.4: Various Forms of Nitrogen in Wastewater Industry (Wall, 2013)

According to Kumar et al. (2020), a point source is referred to any single identifiable source of nitrogen pollution into water bodies which includes industries and municipal sewage treatment plants. It is also known that the point source from urban areas contributed to the majority of the nitrogen pollution in water bodies as it made up of 50 % of the total nitrogen. On the other hand, a non-point source refers to nitrogen pollution source that is not well-defined. The contribution of the non-point source of nitrogen pollution are more evident in the rural areas. For example, significant nitrogen run-offs from the agricultural fields can be found in the stream in rural areas.

According to Zhang et al. (2021), one of the forms of nitrogen that can be found in the wastewater industry is in the form of dissolved organic nitrogen

(DON). DON compounds is known as the faction of organic nitrogen that is capable of passing through a 0.1 μm pore size filters and can be widely found in the environment in the form of nucleic acids, urea, amino acids, humic and fulvic substances, . Zheng et al. (2021) further explains that DON plays a very crucial role in the global nitrogen cycle as it exists majorily as fixed nitrogen in the surface water. One of the dangers posed by DON is that its presence in the surface water bodies will cause the generation of toxic nitrogenous disinfection by-products and growth of algae being stimulated. Due to its hazards posed towards the environment as well as to the human health, scientists and researchers have been focusing on its sources, composition as well as its bioavailability of DON in freshwater systems for the past decades (Zheng et al., 2021). According to Pehlivanoglu-Mantas and Sedlak (2008), the primary form of nitrogen in the effluent discharge is DON due to the increasing use of nitrification and denitrification processes in the municipal industry. Their findings resonate with that from Zheng et al. (2021) which states that wastewater-derived DON will lead to the stimulation of algal growth and acts as a precursor for the formation of carcinogenic products.

2.4.2 Nitrogen Level in Different Types of Wastewater

The municipal wastewater treatment plants receive a variety of wastewater ranging from household, industrial, commercial building, surface run-offs and more. Thus, there are many factors that contributed to the quality of the wastewater entering into the municipal wastewater treatment plants which includes the number of inhabitants, types of industrial discharge and environmental factors such as rainfall patterns (Guida et al., 2021). It is also known that the nitrogen discharge for each source varies according to the type of wastewater source which includes different industrial areas. The wastewater influent nitrogen levels from different sources into the municipal wastewater industry were tabulated in Table 2.4.

Table 2.4: Nitrogen Levels from Different Sources of Wastewater Influent (Al-Dhabi et al., 2021; Sonkar, Kumar and Dutt, 2020; Xu et al., 2020; Tejido-Nuñez et al., 2019; Said et al., 2020; Wang et al., 2020; Shin et al., 2022; Kong et al., 2021; Guida et al., 2021; Roslan, Debbra and Tan, 2019)

Nitrogen Sources	Location	Nitrogen Level (mg/L)	Sources
Date Processing	Riyadh, Saudi Arabia	NO_3^{2-}: 4.6 ± 0.52	Al-Dhabi et al., 2021
Paper Mills	Saharanpur, India	TKN: 14.8 ± 4.7	Sonkar, Kumar and Dutt, 2020
Agriculture – Swine manure	-	TN: 21.60 NH_4^+: 7.84 NO_3^{2-}: 0.06 NO_2^-:0.03	Xu et al., 2020
Aquaculture (Nile tilapia)	Wädenswil, Switzerland	NO_3^{2-}: 152.8	Tejido-Nuñez et al., 2019
Coffee Processing	Pulau Pinang, Malaysia	NH_3-N: 137	Said et al., 2020
Tofu Processing	-	TN: 1273.5	Wang et al., 2020
Municipal Sewage and Poultry Farms	Gwanju, Korea	TN: 58.4 ± 0.3 NH_4^+: 34.9 ± 0.3 NO_3^{2-}: 0.5 ± 0.1	Shin et al., 2022

Municipal	-	TN: 44.1 ± 3.0	Kong et al., 2021
Municipal	-	NH_4^+: 37.7	Guida et al., 2021
Ruminant Abattoir	Seremban, Malaysia	NH_4^+/NH_3: 29 – 70	Roslan, Debbra and Tan, 2019
Ruminant Abattoir	Banting, Malaysia	NH_4^+/NH_3: 55 - 144	Roslan, Debbra and Tan, 2019

2.4.3 Nitrogen Effluent Dicharge Regulation

It is important that the wastewater produced from all wastewater industries to comply with the regional wastewater discharge standard. This is because excessive input of pollutant into the water bodies will cause severe negative consequences to the environment, human health, and ecosystem as a whole. According to the Environmental Quality Industrial Effluent Regulations 2009 found in the Environmental Quality Act 1974 (Department of Environment, 2009), the conditions to be accepted for the nitrogen discharge from of industrial effluent Malaysia was tabulated in Table 2.5. There are two standards in place for the discharge of nitrogen in Malaysia. Standard A is applicable for discharge of water supply at upstream intake points and Standard B for water intake points at downstream discharge and other areas that do not fall under Standard B (Roslan, Debbra and Tan, 2019).

Table 2.5: Maximum Permitted Values of Ammoniacal Nitrogen of Industrial Effluent in Malaysia (Department of Environment, 2009)

Parameter	**Unit**	**Maximum Permitted Values**	
		Standard A	**Standard B**
Ammonia/Ammonium	mg/L	10.0	20.0

Besides, there is also the acceptable conditions for the discharge of sewage systems regulated by the Environmental Quality Sewage Regulations 2009 in the Environmental Quality Act 1974 in Malaysia (Department of Environment, 2009) which was shown in Table 2.6. It is known that more stringent discharge limits are imposed in Malaysia as the previously existing discharge limit of ammonia-nitrogen has been reduced from 50 mg/L to 5 mg/L (Kutty, Isa and Leong, 2011). Thus, these stricter regulations are needed to be complied to ensure the reduced negative impacts in Malaysia.

Table 2.6: Maximum Permitted Values of Nitrogen of Sewage Discharge in Malaysia (Department of Environment, 2009)

Parameter	**Standard (unit in mg/L)**	
	A	**B**
Ammoniacal Nitrogen (enclosed water body)	5	5
Ammoniacal Nitrogen (river)	10	20
Nitrate – Nitrogen (river)	20	50
Nitrate – Nitrogen (river)	10	10
Nitrate – Nitrogen (enclosed water body)	5	10

Besides Malaysia, the European Union (EU) had also set the best available technique of associated emission of nitrogen level into the water bodies as according to Table 2.7. The best available technologies (BAT) are used as the sole reference for the permitted emission levels for various types of pollutants, such as nitrogen, under normal operating condition.

Table 2.7: Allowed Nitrogen Emission Level into Receiving Water Bodies in Countries of European Union (Official Journal of the European Union, 2016)

Parameters	**Nitrogen Emission Level (mg/L)**	**Conditions**
Total Nitrogen (TN)	5.0 – 25.0	Nitrogen emission level exceeds 2.5 tons/year.
Total Inorganic Nitrogen	5.0 – 25.0	Nitrogen emission level exceeds 2.0 tons/year.

In Japan, the national effluent standards for nitrogen emission levels can be divided into 2 categories, which are standards that protect human health and living environment respectively. The discharge limit of nitrogen into the water bodies in Japan was summarised in Table 2.8.

Table 2.8: Permissible Limit of Nitrogen Discharge Level to Water Bodies in Japan (Ministry of the Environment, 2015)

Category	**Parameters**	**Permissble Limit of Nitrogen Discharge (mg/L)**
Protection of Human Health	Ammonia, Ammonium compounds, Nitrate and Nitrite compounds	100.0
Protection of Living Environment	Total Nitrogen	120.0 (Daily average 60.0)

In China, it is known that a maximum permissible limit of nitrogen discharge to surface water of only 1.0 mg/L is allowed according to the Ministry of Ecology and Environment by The People's Republic of China (2002). Such stringent limit as compared to other countries is imposed due to the environmental impact it brought forth due to increased population in the country.

2.4.4 Conseqeunces of Improper Nitrogen Management

Although the wastewater industry aims to minimise the discharge of pollutants such as nitrogen to be released into the water bodies, potential mismanagement of nitrogen can occur in the industry if equipment are not well-maintained, or process conditions are not monitored closely. With the improper management of nitrogen in the wastewater industry, the pollution issues that are derived from the existence of nitrogenous compound will surface and cause severe negative impact to the surroundings. It is known that nitrogen is regarded as one of the major contaminants that results in deterioration of water and depletion of oxygen level in the water bodies due to the absence of effective treatment process prior to discharge (Li et al., 2021).

According to Rahimi, Modin and Mijakovic (2020), the problem of nitrogen contamination of water sources is a growing environmental issue and it brings severe consequences to the human health and the environment. Some of the examples of sources of contamination due to nitrogen can be found in ground and surface waters. The increasing use of nitrogen-based fertilisers in the agricultural field has significantly polluted the ground and surface water sources, which amount to 293 000 tonnes usage of these fertilisers annually alone in the region of Canada. Besides, untreated waste dumped into the water sources from various places such as from the industries and residences have caused the contamination of water with nitrogen. Rahimi, Modin and Mijakovic (2020) also added that many of the wastewater industry have contributed to the nitrogen loading issue into the surface and groundwater which amounts to 80 000 tonnes yearly.

According to Ye et al. (2021), the majority of the nitrogen used in the agricultural field is lost to the aquatic environment as well as to the atmosphere, and only approximately 17 % of the nitrogen is uptake by crops and livestock and eventually consumed by humans. Thus, it is evident that there will be large fluxes of reactive nitrogen to be present in water bodies that can cause many environmental issues. According to Sengupta, Nawaz and Beaudry (2015), there is an increase use of nitrogen and phosphorus fertilisers across the world which leads to the increase of the weight usage of synthetic fertilisers of approximately 15 Tg yearly. Although this increase in the nitrogenous and phosphorus-based fertilisers have aided to combat against the global issue of world hunger, the other issue of nitrogen leakage occurred. For instance, only 4 Tg out of 170 Tg of the applied reactive nitrogen accumulated for crop yield, while the rest is dissipated through air and water pathways. This have clearly shown the origin of nitrogen from the agricultural industry which brings adverse impacts on the human health, biodiversity as well as on the air and water quality.

According to Chan-Pacheco et al. (2021), there are several consequences for the pollution caused by nitrogen, which are the acidification and eutrophication of water bodies, imposing death of aquatic species which affects negatively on the biodiversity, the decreased quality of air leading to various respiratory diseases, potential emergence of carcinogenic compounds and more.

Eutrophication

One of the prominent examples of nitrogen pollution is the eutrophication phenomena as it continues to serve as a global ecological threat. This is because the necessary upgrading of wastewater industry on its unit operations in order to meet the constantly stricter discharge effluent is challenging (Zhou et al., 2021). According to the National Oceanica and Atmospheric Administration, also known as NOAA (2021), eutrophication is caused by the excessive discharge of nutrients such as nitrogen into the water bodies which leads to the substantial growth of algae on the surface of the water, also known as algal bloom. The presence of algal growth does not only consume the available dissolved oxygen present in the water bodies, but further blocks the sunlight from reaching the aquatic plants beneath which hinders the process of photosynthesis. Besides, severe decreased levels of available dissolved oxygen present in the water bodies can be found as the algae dies and undergoes the process of decay which takes up the already limited dissolved oxygen. Such a phenomena constitute to the death of many aquatic plants and animals of the water bodies. According to Edokpayi and Durowoju (2017), the formation of algal bloom in the water bodies leads to the increase of turbidity of the water, plant and animals biomass, sedimentation rate and the decrease of diversity of species. Possible anoxic condition of the water bodies can occur as well which causes fatality of aquatic plants and animals.

According to United States of Environmental Protection Agency, also known as US EPA (2021), the algal boom phenomena have created dead zone in the water bodies, which denotes the little or no oxygen present in the water. A condition known as hypoxia is also created in the water bodies as algal blooms consumes the oxygen and dies off later. A study conducted by US EPA (2021) have documented that there are over 166 dead zones globally, which includes waterbodies such as the Chesapeake Bay and the Gulf of Mexico US EPA, 2021).

Acidification of Water Bodies

Besides the occurrence of algal bloom, one of the related consequences of improper management of nitrogen in wastewater industry is the acidification of water bodies. When ammonia compounds are discharged into the water bodies, the formation of acidified streams will be produced. One of the most common forms of ammonia which is ammonium sulphate will release hydrogen ions into the water bodies during the process of nitrification (Kumar et al., 2020). The water streams is further acidified when the present nitric acid is formed through the nitrite ions present along with sulphate ions rendering the stream unapplicable to be used for drinking purposes.

Negative Health Impacts

According to Chen et al. (2017), it is known that nitrite ions are more toxic compared to nitrate. This may lead to serious health complications to human upon consumption of drinking water with nitrite such as liver damage, formation of various types of cancer and results in birth defects. Excess quantity of nitrite present in drinking water will also lead to the inability of red blood cells to transport the oxygen which causes the deoxygenation of the body. Besides, the exposure towards nitrite also causes the occurrence of “blue baby syndrome” on infants and young livestock, the formation of various tumours in the human body as well as causing thyroid-related diseases (Kumar et al., 2020).

Nitrous Oxide Emission

Nitrous oxide (N_2O) is a strong GHG as it was considered the most dominant ozone-depleting chemical compound in the 21st century (Law et al., 2012). According to US EPA (2021), the emission of nitrous oxide in the United States accounted for 7 % for the total GHGs as a result of anthropogenic activities which includes agriculture, combustion of fuel, industrial processes as well as the wastewater management process. The potent effect of the nitrous oxide can be demonstrated thought its ability to remain in the atmosphere for an average of 114 years before being eliminated completely through chemical reactions. Besides, warming effect of one unit mass of nitrous oxide GHG is equivalent to 300 times of carbon dioxide (US EPA, 2021). In the year 2019, the emission of nitrous oxide produced from the wastewater industry accounted for 6 % in the United States according to Figure 2.5.

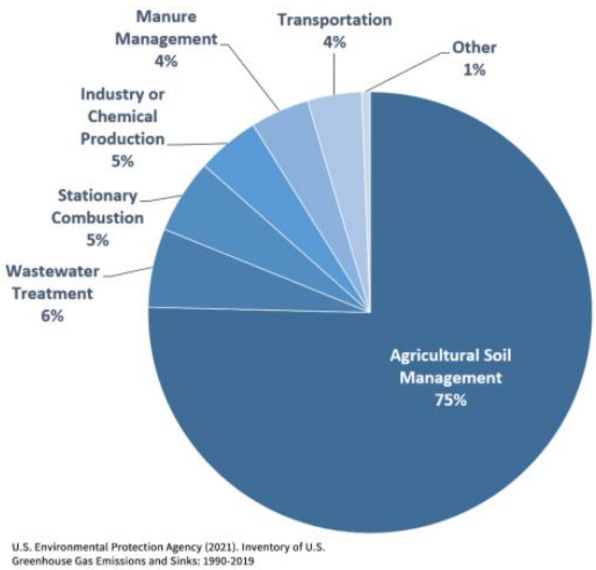

Figure 2.5: Emission Estimates of Nitrous Oxide from Various Sources in the United States (United States of Environmetnal Protection Agency, 2021).

The production of nitrous oxide from the wastewater industry is derived from the biological nutrient removal (BNR) processes which involved the aerobic nitrification and anaerobic denitrification pathways (Law et al., 2012). Although there are a variety of types of BNR plants available that are able to remove the high levels of nitrogen present in the wastewater, the emission of nitrogen oxide is still inevitable. According to Law et al. (2012), the rate of nitrous oxide emission from the wastewater industry is impacted by the loading rates of nitrogen in the reactors that may increase or decrease the rate of nitrification and denitrification. However, improvement on the process design and conditions on the wastewater industry poses opportunity to mitigate the challenges of nitrous oxide emission.

A research on the emission of nitrous oxide was conducted by Hwang, Bang and Zoh (2016) at Jungryang WWTP as one of the four major WWTPs industries located at Seoul, South Korea. A total flow of domestic wastewater with flowrate of 1,710,000 m^3/day was to be treated onsite with the activated sludge and anaerobic and anoxic processes which caters for approximately 3.22 million population in the vicinity. The emission factor (EF) of nitrous oxide on each of the wastewater treatment processes in the plant was shown in Table 2.9. It can be known that the aeration basin produced the most amount of nitrous oxide emission.

Another research was also conducted by Valkova et al. (2020) on the emission of nitrous oxide from ten full scale of selected municipal wastewater treatment plants (WWTPs) in Austria which utilises the most commonly used technology in the country. The EF of nitrous oxide on the ten WWTPs which have varying loading conditions, wastewater characteristics (TN-to-COD ratio) and its distribution of municipal and industrial wastewater was shown in Table 2.10. It can be seen that the nitrous oxide EF ranges from as low as 0.005 (G) to the highest of 1.52 (D). Thus, it is concluded that the nitrous oxide emission is affected by the different wastewater composition to be treated as well as its plant utilization capacity.

Besides that, Valkova et al. (2020) also proposed an estimation model that correlates the nitrous oxide emission factor to the total nitrogen removal rate of a plant which can be applied for other wastewater industries besides the region of Austria. The negative correlation of nitrous oxide with the degree of total nitrogen (TN) removal by assuming that the removal of nitrogen was achieved through the biologically nitrification and denitrification method as shown in Figure 2.6. Hence, it can deduced that the higher the TN removal rate of the wastewater, the lower the nitrous oxide EF which promotes a more greener pathway.

Table 2.9: Emission Factor of Nitrous Oxide for Different Wastewater Treatment Processes at Jungryang Sewage Treatment Plant (Hwang, Bang and Zoh, 2016)

Process Unit	**EF of Nitrous Oxide, N_2O (g N_2O/kg TN)**
The Activated-Sludge Process	
First settling basin	0.263 ± 0.03
Aeration basin	0.672 ± 0.37
Secondary settling basin	0.226 ± 0.01
Sludge thickener	0.195 ± 0.09
Total	1.256 ± 0.50
The Advanced Treatment	
Anaerobic reactor	0.182 ± 0.02
Anoxic reactor	0.249 ± 0.01
Aerobic reactor	1.174 ± 0.03
Total	1.605 ± 0.15
Sludge digestion tank	0.012 ± 0.01

Table 2.10: EF of Nitrous Oxide on Ten WWTPs in Austria with Varying Characteristics of Influent Wastewater (Valkova et al., 2020)

WWTPs	Plant Utilization (PE)	Type of Wastewater	Influent TN/COD ratio	TN Removal (%)	EF of Nitrous Oxide (%)
A	80,000	Mainly municipal	0.1	83.7	0.19
B	17,000	Mainly municipal, food manufacturing	0.065	90.1	0.13
C	14,500	Mainly municipal	0.08	92.0	0.012
D	30,000	Mainly municipal	0.08	68.7	1.52
E	725,000	Mainly with high inflow of industrial wastewater (steel, paper and chemical)	0.08	63.6	1.32
F	175,000	Municipal, paper and food industry	0.065	84.4	0.05
G	17,000	Mainly municipal	0.1	92.5	0.005
H	25,000	Municipal, winter tourism	0.09	72.7	0.99
I	61,500	Municipal, winter tourism	0.06	78.5	0.85
J	77,000	Municipal, winter tourism	0.09	67.4	1.03

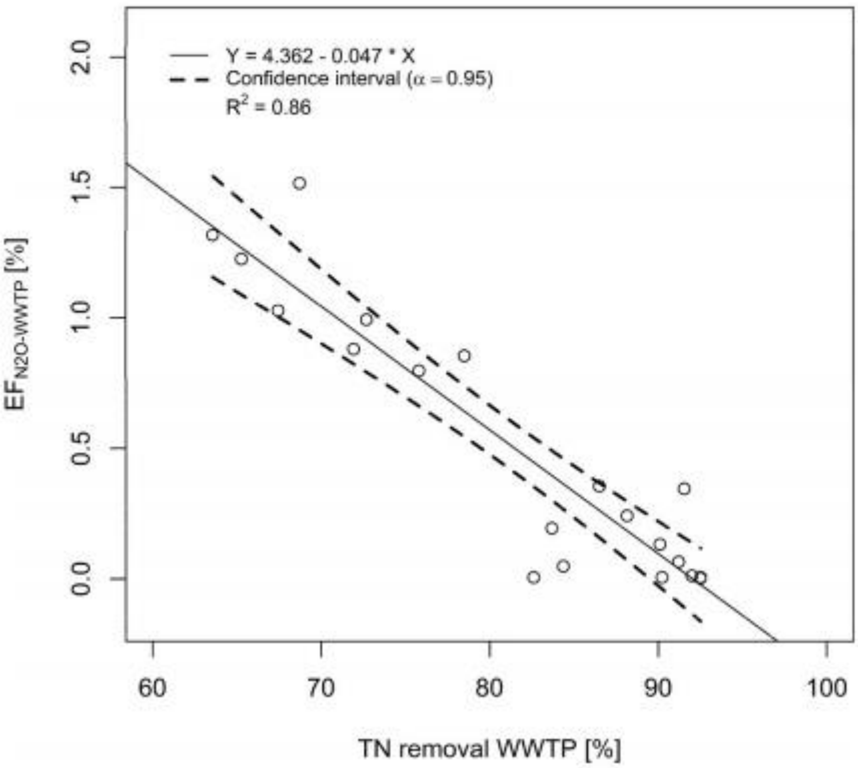

Figure 2.6: Scatter Plot of the Average Nitrous Oxide EF and the TN removal efficiency (Valkova et al., 2020)

The main pathways for the nitrous oxide formation consists of the two main pathways of autotrophic nitrification and heterotrophic denitrification which can be typically found in the activated sludge systems (de Haas and Ye, 2021). There is also a third uncommon chemical pathway for the formation of nitrous oxide. The details for each biochemical pathways were tabulated in Table 2.11 cited by de Haans and Ye (2021). It is important to know the biochemical pathway of the nitrous oxide emission in order that mitigation efforts can be done to curb the existing problem to further reduce nitrous oxide emission at decreased level

Table 2.11: Main Biochemical Pathways for the Formation of Nitrous Oxide in Activated Sludge Systems in Wastewater Industry (de Haas and Ye, 2021)

Pathway	**Sub-pathway**	**Process conditions favouring N_2O formation**	**Microorganisms involved**	**Biochemical mechanism**
Autotrophic nitrification	Nitrifier denitrification	Low concentration of dissolved oxygen (DO) and nitrite accumulation	AOB such as nitrifiers oxidising ammonia to nitrite	Ammonia is oxidised to nitrite by AOB and surviving under low DO levels and high nitrite concentration by means of reduction of nitrite to nitrous oxide. The emission of nitrous oxide is due to the lack nitrous oxide reductase enzyme which converts nitrous oxide into nitrogen gas as the final step.
	Hydroxylamine oxidation	High DO, high turnover rates of aerobic oxidising bacteria (AOB) and hydroxylamine accumulation		Nitrous oxide is emitted through the breakdown of nitrosyl radical which is formed through the oxidation of hydroxylamine as the intermediate product. Hydroxylamine acts as the intermediate product from the oxidation of ammonia into nitrite.

Heterotrophic denitrification	-	Anoxic condition, low pH, presence of hydrogen sulphite; nitric oxide and nitrite accumulation; lack of biodegradable organic carbon	Heterotrophic denitrifies	Inhibition of the nitrous oxide reductase which results in the accumulation of nitrous oxide in the four-step reduction chain of denitrification chain as follows: Nitrate > Nitrite > Nitrogen Monoxide > Nitrous oxide > Nitrogen gas
Chemical	-	High concentration of hydroxylamine and nitrite, very low pH levels	Abiotic	Chemical

2.5 Conventional and Current Nitrogen Management in Wastewater Industry

The current management of nitrogen is through its removal processes, which are the biological, chemical and physical treatment processes. According to Sandip and Kalyanraman (2017), the current method of nitrogen removal through the incorporation of the conventional biological nitrogen removal (BNR) processes is known as a highly energy and cost intensive practice to be used in new wastewater treatment facility. This is because huge amount of energy is required in order to provide an aerobic condition for the decomposition of organic matter through microbial actions. In the physical-chemical treatment method, it acts as an alternative to the commonly used biological process and is more robust according to the types of wastewaters it caters to and the various applications and configurations possible available in the wastewater industry (Boyer, 2014).

The need to remove excessive discharge of nitrogen from wastewater treatment is still an important consideration in nations across the world. Such as in the nation of India, it poses a huge challenge for industrial players to comply with the strict discharge regulation of total nitrogen less than 10 *mg/L* in sewage treatment plants for both private facilities as well as organizations. In the event that these organic pollutants are discharged in an improper manner, the environmental bodies will be affected through eutrophication and affecting many more living aquatic plants and animals (Sandip and Kalyanraman, 2017). Thus, the following section will highlight several pathways for the conventional and current methods of nitrogen removal in the biological, physical as well as chemical treatment processes.

2.5.1 Biological Treatment Methods

Nitrification-Denitrification

The conventional process for the management of nitrogen is the nitrification-denitrification pathway. Its pathway can be shown visually in Figure 2.7.

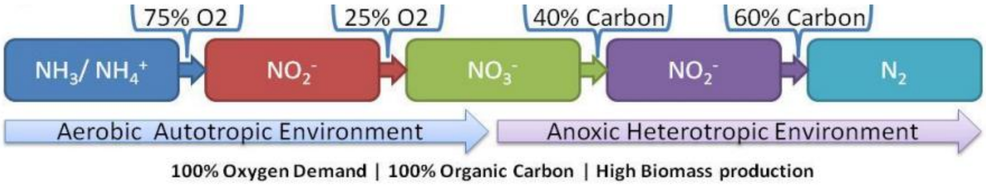

Figure 2.7: Nitrification-Denitrification Pathway (Sandip and Kalyanraman, 2017)

The nitrification-denitrification pathway can be broken down into both nitrification and denitrification processes. According to Rahimi, Modin and Mijakovic (2020), the nitrification process consists of two biological oxidation processes which are being carried out in sequence. The first step of the process involves the oxidation of NH_4^+ or NH_3 into NO_2^- while the second step involves the rapid conversion of the nitrite ion into nitrate ion, NO_3^-. Both of these processes involved the usage of biocatalytic enzymes such as ammonia monooxygenase (AMO) and hydroxylamine oxidoreductase (HAO) as well as bacteria such as nitrite-oxidising bacteria (NOB). This biological nitrogen removal process poses greater advantages to the physiochemical processes for wastewater treatment due to its cost-effectiveness. However, there are still drawbacks that remains in this process, which included the slow nitrification reactions, decreased activity of nitrification when ammonium and organic matter present were overloaded, the need to strict monitor and control of the oxygen content as well as need of two reactors instead of one for both aerobic nitrification and anaerobic denitrification processes. Besides, the use of large reactors is required for both processes which increases the hydraulic retention time which is necessary due to the low nitrification reaction rate. However, it gives the drawbacks of leading to the high operating cost in the long run.

In the denitrification process, it involves the complete removal of nitrate ions to produce the inert form of dinitrogen gas as the end product (Rahimi, Modin and Mijakovic, 2020). This process does not only generate low waste of brine, but it is available to treat various types of contaminants at once that resulted the waste disposal cost to decrease significantly. In this process, several factors need to be considered as deemed essential, such as the strict requirement of anoxic conditions, continuous sources of carbon as well as the need for post-treatments. Sources of carbon are required for this process as it functions as electron donors which are important for the growth of cells and the heterotrophic denitrification.

Despite the advantages it brings, several drawbacks can be found in this process which prevented the large-scale application of bio-denitrification (Rahimi, Modin and Mijakovic, 2020). This includes the formation of turbid substances due to the biomass and leftover carbon sources that further requires the need to undergo treatment process. Besides, there presents a risk for the production of nitrous oxide which is more potent than carbon dioxide. A continuous supply of carbon sources and the need to monitor its precise dosage also constitute a burden to the treatment process and to avoid the decreased effluent water quality through the excessive discharge of biomass and carbon sources. In addition to all these, the process of denitrification which is conducted in an anaerobic environment would be render ineffective due to the infiltration of oxygen into the process as it brings negative effects on the efficiency of nitrogen removal and further increases the concentration of nitrite ions in the treated water. One of the challenges of this process also includes the slow reaction rate, which is attributed to high start-up time and hydraulic retention time, the need for the adjustment of pH as well as the decreased productivity at low temperature condition. Thus, it can be concluded that the biological denitrification process may effectively remove nitrogen, it is not susceptible to the option of nitrogen recovery which poses a major drawback of this process (Rahimi, Modin and Mijakovic, 2020).

Aerobic Denitrification

According to Sandip and Kalyanraman (2017), the process of aerobic denitrification pathway constitute to an alternative to the conventional anaerobic denitrification pathway. This is because it is able to overcome the obstacle posed by the anaerobic denitrification process such as it is a more cost-effective and economical method for the removal of nitrogen as both nitrification and denitrification processes can now undergo under a single unit operation without segregating them (Rajta et al., 2020). This method is only discovered as early as in the 1985 by a researcher of the name of Roberston had reported the denitrification conducted in the presence of oxygen by the bacterium *Thiosphaera pantotopha* (Mulder et al., 1985). Due to the ability for both nitrification and denitrification to occur in a unit operation, it becomes more economically favourable as reduced usage of chemicals that is required to monitor the pH condition of the process because the alkalinity produces from the denitrification process is able to compensate for the alkalinity consumption in the nitrification process (Rajta et al., 2020).

Nitritation-Denitratation

The nitritation-denitratation process has similar characteristics to that of the conventional nitrification-denitrification pathway, but it avoids one of the process which involves the oxidation of nitrite into nitrate. The absence of this oxidation process by nitrite oxidising bacteria is replaced by the direct conversion of nitrite into dinitrogen gas by means of denitrification by heterotrophic bacteria according to Sandip and Kalyanraman (2017). The pathway for the nitritation-denitritation was shown in Figure 2.8.

Figure 2.8: Nitritation-Denitratation Pathway (Sandip and Kalyanraman, 2017)

According to Noutsopoulos et al. (2017), the nitritation-denitratation pathway, also known as the short-cut nitrification-denitrification pathway is used often to remove ammonia from sludge liquors produced from the treatment and handling of sewage sludge. The treated sewage sludge usually contains elevated concentration of ammonium and low COD to N ratio which will be channelled into the wastewater treatment line for further removal of the nitrogen content. It is used because of its prevalence of ammonia-oxidisng bacteria (AOB) and the inhibition of nitrite oxidising bacteria (NOB) which offers a reduction of 25 % aeration requirement as well as reducing as much as 40 % of the carbon source required for the denitrification stage in the conventional process. This process is able to undergo the treatment of reject water with satisfactory rates of nitritation and denitritation with an inlet temperature higher than 20 ℃ according to Noutsopoulos et al. (2017). However, the additional external carbon source with a high readily biodegradable content can achieve higher nitritation and denitritation rates which better boost the performance of wastewater treatment process.

Anaerobic Ammonium Oxidation (Anammox)

According to Weralupitiya et al. (2021), the emergence of an alternative pathway for the removal of nitrogen from wastewater industry is through the anaerobic ammonium oxidation process, also known as anammox. This pathway is only discovered in the early 1990s during a research performed in a fluidized bed reactor which acts as a revolutionary nitrogen removal pathway than the conventional nitrification-denitrification (Mulder et al., 1995). The transformation of nitrogen was being conducted in the anammox pathway as shown in Figure 2.9.

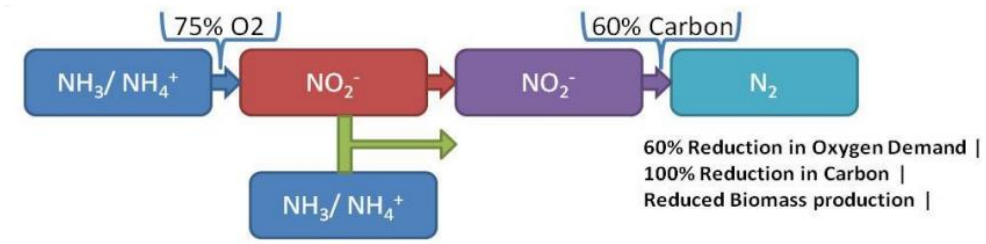

Figure 2.9: Anammox Pathway (Sandip and Kalyanraman, 2017)

The anammox pathway is considered to be unique due to its ability to aerobically oxidise the ammonia or ammonium ions by utilising nitrite ions as the final electron acceptor to produce the end product of dinitrogen gas, N_2. Anammox pathway is known for its novelty due to its promising benefits of low costs required compared to the conventional nitrification/denitrification pathway as it relies on bicarbonate or carbon dioxide to fulfil its carbon requirement rather than costly biomass (Gao et al., 2018). Besides, it is more advantageous towards the conventional process as it is an autotrophic process which does not utilise the carbon source present in the wastewater (Rahimi, Modin and Mijakovic, 2020a). In addition to these, the anammox pathway offers the enhancement of energy recovery from wastewater through the production of methane gas, reduction of the energy consumption due to lesser oxygen demand, reduction of 50 % on the amount of ammonium ion to be oxidised into nitrite instead of nitrate, higher rate of nitrogen removal, minimises surplus sludge production as well as decreasing the emission of GHGs as carbon dioxide is consumed and lower emission of nitrous oxide (Rahimi, Modin and Mijakovic, 2020a).

Despite many advantages that was offered by the anammox pathway as previously mentioned, the one crucial drawback is the overloading amount of nitrate to be found in the discharge effluent that requires additional denitrification treatment process. With the need for the denitrification treatment process comes with a price of increased demand of carbon sources, which ultimately leads to higher energy consumption, surplus activated sludge as well as the carbon dioxide emission. Besides, the high carbon to nitrogen ratio, low temperature as well as the poor discharge effluent quality pose great threat to the application of process of anammox in the wastewater industry (Rahimi, Modin and Mijakovic, 2020a). In addition to these, the presence of organic matter in the wastewater also affects the anammox bacteria as denitrifying bacteria acts to compete which results in decreased efficiency of ammonium removal. The presence of foreign organic substance such as methanol could also negatively affect the anammox activity by deactivating it partially or completely (Rahimi, Modin and Mijakovic, 2020a). Thus, it can be concluded that the anammox pathway is not feasible to be conducted for large-scale engineering application till this end due to its poor discharge effluent water quality which fails achieve the standard effluent discharge standard unless through the addition of post-treatment methods.

2.5.2 Chemical Treatment Methods

Break Point Chlorination

In the chemical treatment of breakpoint chlorination method, chlorine is utilised to remove ammonia compounds from the wastewater (Bock, 2016). The addition of chlorine into the wastewater treatment facility exists in several chemical compounds in the industry, such as chlorine gas, sodium hypochlorite, and calcium hypochlorite (Khawaga et al., 2021). The relationship between the total chlorine residual and ammonia nitrogen existing in the wastewater in relation to the dosage of chlorine was shown in Figure 2.10.

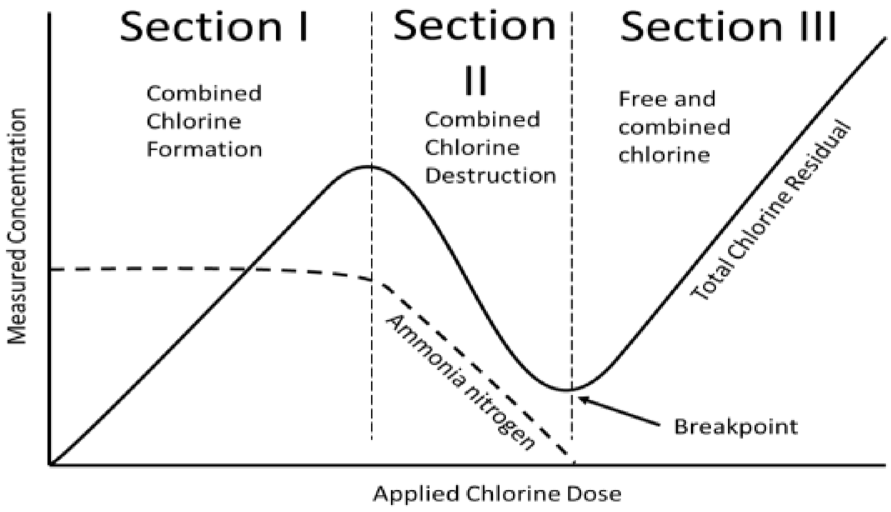

Figure 2.10: Breakpoint Chlorination Graph Showing Measured Concentration of Total Chlorine Residual and Ammonia Nitrogen versus Applied Chlorine Dose (Bock, 2016)

As seen in Figure 2.10, the addition of chlorine added in Section I showed no change in the ammonia concentration, and this added chlorine will start to combine with ammonia in the wastewater to form chloramines, which consists of monochloramines, dichloramine and trichloramine. The increasing dosage of chlorine added in Section II will cause the chloramines formed in Section II to decompose to produce stable products such as nitrogen gas, water, chloride ions, hydrogen ions and nitrate ions. Lastly, the free chlorine compounds will be accumulated once the dosage of chlorine exceeded that of the breakpoint which moves towards Section III. Thus, it can be known that suitable amount of chlorine at the breakpoint will lead to the theoretical total removal of ammonia nitrogen in the wastewater (Bock, 2016)

Electrochemical Oxidation

According to Bock (2016), electrochemical oxidation of ammonia, also known as ECOA is conducted when an ammonia-containing aqueous solution is passed through an electric current. This process is also similar to that of electrolysis and can be categorised into direct or indirect anodic oxidation methods. The direct anodic oxidation method involves the oxidation of ammonium present in the wastewater on the anode while the indirect anodic oxidation requires the addition of oxidising agents into the wastewater to react with the ammonium compounds. For the indirect oxidation method to proceed successfully the evolution of chlorine gas must occur in the process (Ghimire et al., 2019).

The application of ECOA has been widely practiced in various industries such as in the treatment of wastewater from semiconductor plant, lead smelting plant and aquaculture (Chung, Chung and Chung, 2020; Meng et al., 2020; Mook et al., 2012). This removal process is determined by a number of factors, such as the electrode composition, density of current, halogen ions concentration and the pH level of wastewater (Bock, 2016). There are two major factors that are more prominent in the usage of ECOA, which are the type of anode material used and the amount of electric voltage applied as it determines the overall operating costs and the efficiency of the removal process (Ghimire et al., 2019).

According to Ghimire et al. (2019), the electrochemical process for the removal of ammonium as well as other organic compounds in the wastewater industry has brought rewarding economic benefits as follows: high efficiency in operation, ability to operate with a wide operating conditions, require only small-size equipment, decreased generation of sludge and the rapid start-up process as compared to the biological removal process. However, there are also challenges to the application of this removal process such as the need for the reduction of energy consumption and cost for anode materials as well as improvement on the process stability and performance in the long run.

Photocatalytic Oxidation

According to Bock (2016), the usage of titanium oxide, TiO_2 photocatalyst is used for the photocatalytic oxidation of ammonium compounds present in wastewater. This is done through the activation of TiO_2 through exposing to ultraviolet (UV) radiation for the oxidation process. During the exposure of UV radiation on the photocatalyst, photons with sufficient energy will be absorbed resulting in a charge separation as an electron is elevated from the valence into the conduction band. A series of redox reactions will occur in the event that the negative charged electrons and positively charged holes do not recombine fast enough, which produces a charge transfer at the surface of the photocatalyst. These redox reactions generate either of the two pathways: direct oxidation of ammonia, or oxidation of chloride into active chloride and followed by the oxidation of ammonia.

There are a number of factors affecting the photocatalytic oxidation process, which are the concentration of oxygen, initial concentration of total nitrogen, pH levels, activity of various TiO_2 photocatalysts, UV radiation intensity, suspended catalyst concentration as well as salinity. These factors will ultimately determine the type of final products produced from the oxidation reaction and the conversion of nitrogenous compound from the initial concentration (Bock, 2016).

This process has received considerable amount of attention due to the advantages it brings such as its ability to maintain high oxidation rate in high saline concentration of wastewater as compared to the biological removal methods (Zhang, Wang and Sun, 2014). However, this process still poses its challenge due to its difficulty for the separation of suspended TiO_2 particles from the aqueous solution (Wang et al., 2014).

Ozonation

The oxidation reaction with ozone, also known as ozonation, is one of the advanced oxidation processes among all the established technologies for the removal of ammonium compounds from wastewater (Krisbiantoro et al., 2020). It is also known as one of the most promising removal technologies present due to its ability to operate at low temperature and at atmospheric pressure. This has allowed for the reduction of energy demand in place of energy intensive processes making it more applicable for industrial applications specifically in the wastewater industry (Krisbiantoro et al., 2020).

There are several reaction pathways for the ozonation of ammonium compounds which are developed over the years. The summary of the three reaction pathways, namely non-catalytic, homogeneous catalysis and heterogenous catalysis were shown in Figure 2.11.

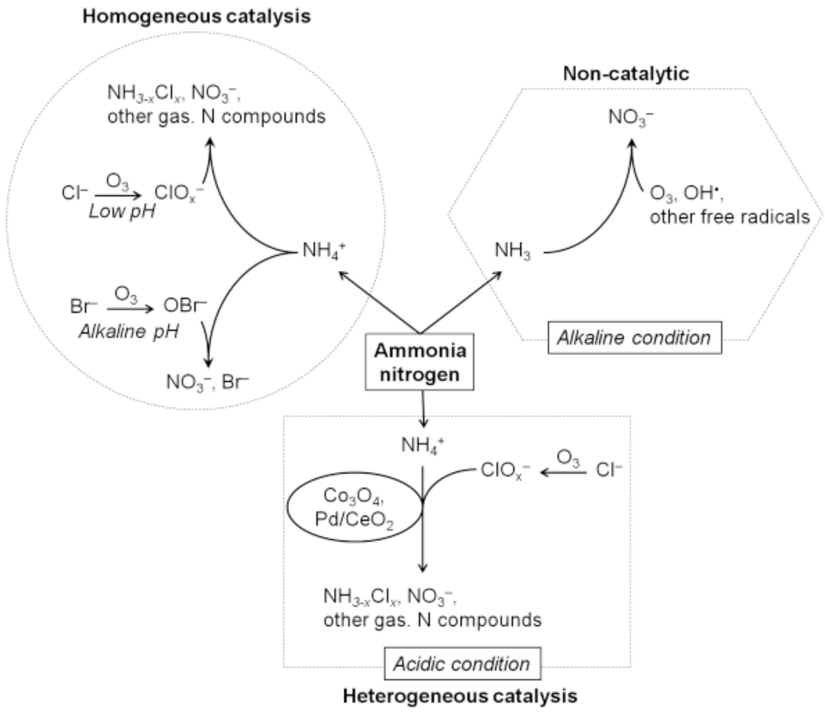

Figure 2.11: Three Reaction Pathways for the Oxidation of Ammonia Nitrogen Through Ozonation (Krisbiantoro et al., 2020)

In the homogenous catalysis process, three catalysts namely Cl^-, Br^- and Pd^{2+} is used while for heterogenous catalysis process utilises Co_3O_4, MgO or Pd/CeO_2 in the reaction. It can be concluded that the ammonia nitrogen can be directly oxidised through ozonation under milk alkaline condition and is oxidised with free radicals for non-catalytic processes, homogenous catalysis processes works under neutral to mild alkaline conditions with Cl^- only able to proceed under acidic condition and heterogenous catalysis is proceeded with Co_3O_4, MgO or Pd/CeO_2. It is known that the process of ozonation to oxidise ammonia nitrogen still requires much research in order for the successful implementation in the industrial world (Krisbiantoro et al., 2020).

Ultrasonic Irradiation

The usage of ultrasound technology in the wastewater industry is known as an advanced oxidation process (Ozturk and Bal, 2015). The mechanism involved in this process is in reference on sonochemical reactions, in which involved the pyrolytic reactions and cavitation. In the ultrasonic irradiation, the pyrolytic reactions achieved high temperature that ranges from 2000 K to 5000 K and pressure from 500 to 10,000 atm in the cavitation bubbles that are formed. The formation of free radicals is stimulated as a result of the ultrasonic irradiation which aids the oxidation of both organic and inorganic matters as well as the degradation of complex chemical compounds. The hot-spot theory is the more commonly used theory among the rest such as electrical, plasma discharge and supercriticial theories as it describes the three different zones in a homogenous liquid of sonochemical reactions, namely the cavitation of bubbles, the gas-liquid interface and bulk solutions (Ozturk and Bal, 2015).

Some of the findings by Ozturk and Baal (2015) on this process can be discovered for varying conditions of ammonia-nitrogen containing wastewaters: 8 to 64 % removal efficiencies of ammonia nitrogen depending on different initial conditions, pH values from 8.2 to 11 records the best removal efficiency. Besides, higher nitrogen removal efficiencies is also associated with higher power densities, higher periods of sonication and decreased levels of initial ammonia concentration. However, a research conducted by Le, Julcour-Lebigue and Delmas (2015) shows a completely opposite result of increasing ammonia nitrogen levels in the ultrasonic irradiation process. This is because the cells are lysed which results in the release of nitrogenous compound present in the cells.

Precipitation

The process of precipitation by transforming into solid compounds is a relevant application for the removal of nitrogenous compound in wastewater industry. In a specific case of wastewater industry from Li et al., (2012) states that the removal of ammonia nitrogen using the biological treatment method is difficult from the 7-Aminocephalosporanic acid (7-ACA) present in the wastewater due to high concentration of ammonium compounds, small quantity of cephalosporin and 7-ACA that prevents the growth of microorganisms. Thus, this method serves as an alternative for the removal of nitrogenous compound through the formation of struvite in the wastewater treatment. According to Bock (2016), the precipitation process begins with the formation of crystal structure through nucleation. After nucleation, layers of struvite will continue to be collected on the seed crystal which allows the growth of crystal. Chlorine can be used to decompose the struvite present in the process into Mg and PO_4. These compounds can be further recycled for the removal of ammonia in wastewater particularly in wastewater with high chemical oxygen demand (COD). Thus, it subsequently provides a more effective means for the ammonia removal as it lowers the demand for the oxidation process (Bock, 2016).

Synthetic Ion-Exchange using Zeolites

According to Bock (2016), the ion-exchange process denotes the exchange of ions from the liquid phase with that present in the insoluble resin phase with the equation as follows. $\bar{A}$ and $\bar{B}$ represent the insoluble phase while A and B represents that in the liquid phase.

$$\bar{B} + A \leftrightarrow B + \bar{A}$$

According to Jorgensen and Weatherley (2006), the process removal of ammonia from wastewater through ion exchange using zeolites such as clinoptilolite have gain much attention due to its outstanding relevant capacity, equilibrium and column breakthrough characteristics. The usage of ion-exchange in the wastewater industry can reduce the accidental discharge of nitrogenous compound limit from the biological removal method. Furthermore, it has the ability to adapt to shock loading of wastewater as compared to conventional method. It is able to perform optimally under pH and temperature levels where the conventional biological treatment method is unsuitable.

2.5.3 Physical Treatment Methods

Air Stripping

In the air stripping process, the ammonia-containing wastewater is stripped with air based on the principle of mass transfer (Kinidi et al., 2018). According to Kinidi et al. (2018), the conventional ammonia stripping process is conducted in a packed bed tower but poses several problems. However, much research has been conducted since then that attempts to overcome such challenges for the removal of ammonia. Compared to other methods, this stripping process has several advantages due to its simplicity in process and its cost-effectiveness in the wastewater industry. The ammonia from the stripping process can also be recovered which in turn for the production of other valuables. It is also a more suitable technology used to remove wastewater with high ammonia concentration and toxic compounds as the process is deemed stable. However, there are also environmental concerns for the release of waste gas into the atmosphere (Bock, 2016).

Membrane Distillation

The process of membrane distillation is used for the removal of volatile compounds which includes ammonia (Duong et al., 2013). As compared to the biological treatment and activated carbon adsorption technologies, membrane distillation provides a smaller footprint and guarantees low-cost treatment for heated waste source. This is because this technology utilises thermal energy as its driving force to transport the vapor molecules through micropores of hydrophobic membrane. This membrane acts as a barrier for the separation of ammonia compounds present in the wastewater.

According to Duong et al., (2013), there are several operating factors that would affect the removal efficiency of ammonia from the wastewater, such as the feed and gas flow rate and temperature. It is known that higher feed temperature, flowrate of feed and gas would lead to increased efficiency of the removal process and permeate flux. There are 4 common configurations available for the membrane distillation process, namely the direct contact MD, air gap MD, sweep gas MD and vacuum MD which can be shown in Figure 2.12. Each of the configurations have distinct characteristics, such as for vacuum MD demonstrates highest mass transfer coefficient but only able to achieve the lowest selectivity and sweep gas MD gives moderate selectivity while having the lowest mass transfer coefficient.

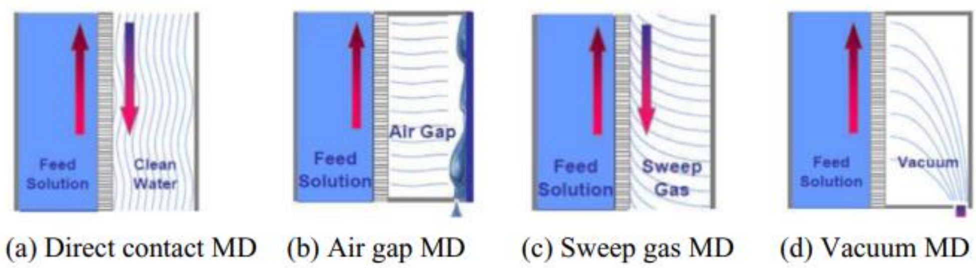

Figure 2.12: Four Common Configurations of Membrane Distillation Process (Duong et al., 2013)

Microwave Radiation

The use of microwave radiation on ammonia removal is based on the principle similar to that of evaporation where wastewater is heated for the release of ammonia into the atmosphere (Lin et al., 2009). This method is also utilised for the elimination of dyes, invasive organisms and other components found in the wastewater besides ammonia nitrogen. However, the full-scale application of this method is still limited as whether would it be effective for the removal of high concentration of ammonia nitrogen. A comparison between steam stripping and microwave radiation for the removal process is conducted by Lin et al. (2009) to found out that this method poses several advantages over it. They further added that this technology have successfully remove the high concentration of ammonia wastewater from the Coke company under a continuous pilot-scale. Nevertheless, the downside of this technology is still prominent which includes the high operating cost, and more research is yet to be done for its reduction (Lin et al., 2009).

2.6 Overview of Circular Economy

The concept of circular economy has been widely addressed in the 21st century in order to reduce the current environmental impacts and challenges brought forth from the linear economy model. According to Morganti and Coltelli (2021), the circular economy can be defined as the system of production and consumption with the objective of waste minimisation, resource optimization, regeneration of natural capital, creation of opportunities for jobs and entrepreneurship as well as the reshaping of production and consumption process in the life-cycle and recycling point of view. There are also other terms denoting the concept of circular economy, such as Cradle-to-Cradle, Zero Waste and Closed Loop Supply Chain (Morganti and Coltelli, 2021). The circular economy concept have revolutionised the production process of all industries in the world to replace the major current practice of linear and reuse economy. A clear illustration of the linear, reuse and circular economy models in terms of its material flow was shown in Figure 2.13.

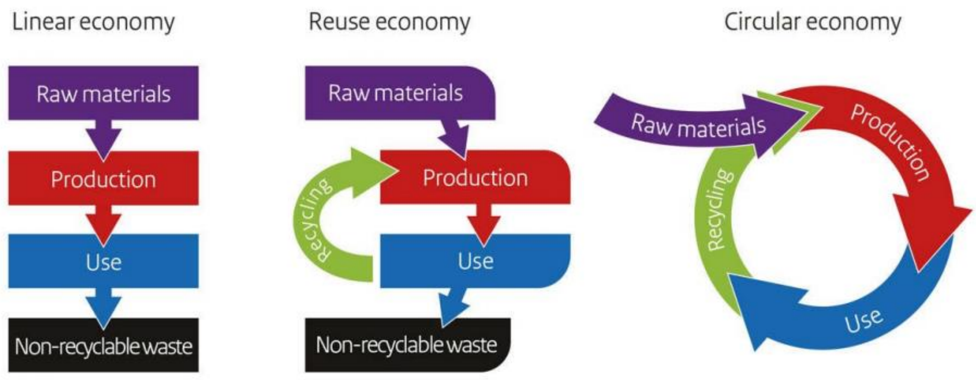

Figure 2.13: Linear, Reuse and Circular Models in Terms of Material Flow (Goverment of the Netherlands, 2021)

According to Wit et al. (2018), an approximately only 9 % of the world's economy have applied the circular economy concept. This could be attributed to the different challenges that many industries have to adapt to the new model as compared to the linear economy of take-make-dispose which is more convenient and easier but generating large amount of waste and unwanted pollution. Besides, the linear economy is able to thrive in the past as it promises high profit for different manufacturers and industries across the world and able to offer cheaper prices of products and services to consumers (Chatham House, 2021).

In the circular economy concept, there are two distinctive cycles, mainly the biological and technical. The biological cycle explains the generation of products for consumption purpose in a cyclic loop while the technical cycle demonstrates the production of products for service as part of the cradle-to-cradle concept. Figure 2.14 illustrated the two types of cycle in the circular economy model.

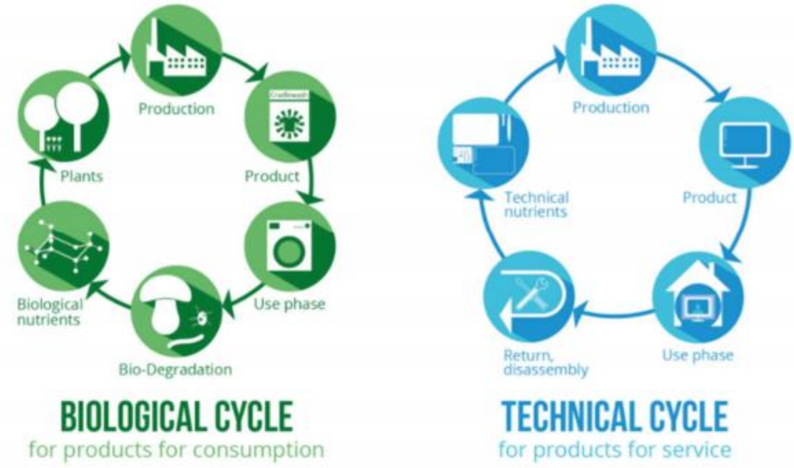

Figure 2.14: Biological and Technical Cycle in Circular Economy Model (Wautelet, 2018)

There are several benefits of the adoption of circular economy model into the current economy, not only does it mitigates the environmental issues at hand due to reduction in pollution and waste, but also provides benefits in terms of social and economic factor. According to Cavallo and Cencioni (2017), several advantages are highlighted when the circular economy is transited from the conventional linear economy, which are provides economic growth and collaboration between companies for a greater system, improvement of products and potential savings on cost associated with production, competition between business to be enhanced, creating more job opportunities and providing betterment to family as savings can be made.

2.6.1 Circular Economy in Wastewater Industry

Wastewater influent into the wastewater industry usually contains various nutrients and acts as a useful resource for many applications if it is recovered. These nutrient-enriched wastewater are mainly derived from various sources, such as the food and agricultural industries as well as municipal activities (Kurniawan et al., 2021). For example, the human sewage is nutrient-enriched in that the urine collected have contributed to 50 to 80 % of the concentration of phosphorus and 75 % accounted for the total nitrogen concentration which can be captured and reused for other applications (Saliu and Oladoja, 2020). In the current practice in the wastewater industry, most of these nutrients are removed through biological, chemical or physical treatment methods in order to comply with the discharge standard from regulatory bodies. Although these removal methods have granted promising removal efficiency of these nutrients, it is still limited by the amount of energy utilised and the need for trained personnel to handle such systems whose end goal is to comply with the discharge standard (Kurniawan et al., 2021).

2.6.2 Challenges of Circular Economy in Wastewater Industry

According to Zhang and Liu (2021), the current widely practice biological nitrogen removal (BNR) process for the removal of ammonium compounds from the wastewater is not suitable for the recovery of these nutrients in term of its design. Thus, the utilisation of BNR have departed from the circular economy approach which needs to be replaced with a more sustainable process. However, it is also known that the conventional BNR processes have been practiced for decades and a change of process would be challenging for all industrial players for the recovery of ammonium compounds from wastewater instead of its removal. Such a change from the linear economy to the circular economy will demand a change of thinking from all parties involved.

According to Flores et al. (2018), several wastewater treatment plant policy are also in place for the adoption of circular economy in Mexico. However, there are also several factors in place that continue to hinder its implementation, such as the perspective of top-down implementation, low availability of resources, spotlight short-term results due to short-sightedness into the overall benefits, enforcement of regulatory bodies on water pollution are not in place and the insistence of embracement of the linear economy model. Thus, such challenges needs to be overcome for the circular economy concept to come to pass.

Through the circular economy approach in the wastewater industry, the recovery of nutrients from wastewater can be converted into useful products which avoids wastage. This ultimately achieves the objective of the circular economy for protecting the environment as well as adding values to the wastewater industries all across the world (Kurniawan et al., 2021).

2.7 Summary

To this end, it is known that the management of nitrogen is crucial in the wastewater industry due to its intrinsic value which can be recovered. Therefore, the following section will further highlight the different potential technologies and methods that are able to manage nitrogenous compound in the wastewater industry from a circular economy approach which would revolutionise the wastewater industry to a more sustainable level.

CHAPTER 3

METHODOLOGY AND WORK PLAN

3.1 Introduction

The systematic literature review (SLR) methodology was used for the extensive findings on this the topic of this project, Nitrogen Management in Wastewater Industry from Circular Economy Perspective. According to Munaro, Tavares and Bragança (2020), the SLR approach founded upon deeply qualitative analysis which can be divided into descriptive and thematic analysis of the data. Such an approach have granted a more holistic view on the topic of the project in terms of the current nitrogen management methods, potential nitrogen recovery pathways in wastewater industry, as well as challenges for the implementation of circular economy in the wastewater industry through managing its nitrogen resources. Tranfield, Denyer and Smart (2003) also affirms that the SLR approach was a suitable method to be used for planning, synthesizing and accessing data from various journals available to establish the knowledge needed for a particular topic or field of study. The sequence of this report based on the SLR approach was being showed in Figure 3.1.

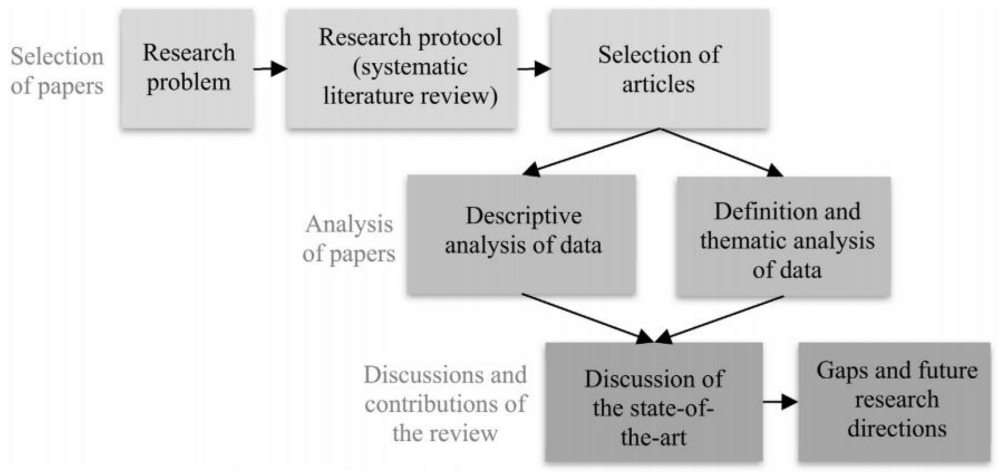

Figure 3.1: Overall Sequence of Report based on SLR Approach (Munaro, Tavares and Bragança, 2020)

3.2 Systematic Literature Review (SLR)

Figure 3.2 shows the necessary steps taken in the SLR approach based on the descriptive and thematic analysis as mentioned previously which consists of three steps: Planning, Processing and Analysis of Results (combination of Stages 3 and 4 from Figure 3.2).

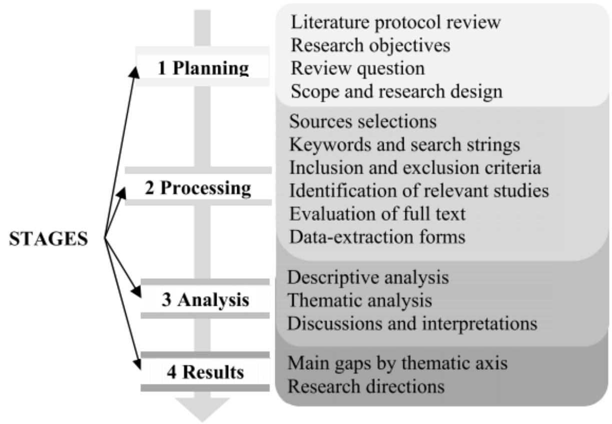

Figure 3.2: Steps Taken in the Systematic Literature Review Approach (Munaro, Tavares and Bragança, 2020)

Step 1: Planning

This was considered as one of the crucial stages for subsequent processes as it marked the beginning of SLR. It was important to review articles and journals related to the topic in an unbiased manner to achieve the aim and objectives of the project. This can be done by planning out a research protocol to promote impartiality and transparency of the research process. The extent of coverage for each objective such as the current nitrogen management, potential nitrogen recovery pathways, feasibility of nitrogen recovering technologies and the challenges of implementing circular economy on the nitrogen management in the wastewater industry are defined in order to set a clear pathway on the direction when reviewing related articles and journals. With the objectives in place, the scope and research design of the project were clearly established. Lastly, the progression of the report was in response to answer the review question of "How can the nitrogen management process in the wastewater industry be part of the concept of circular economy?".

Step 2: Processing

After defining the research scope and design, the process of research had begun by searching from reliable sources pertaining the topic, primarily from ScienceDirect, Scopus of Elsevier, MDPI and ResearchGate. These four sources had been the primary means of obtaining information due to its credibility and reliability of data as well as providing a wide range of research topics and database required for the topic of this project.

Several keywords and search string were employed during the process of reviewing suitable sources for the literature review in Chapter 2, such as "nitrogen management", "municipal wastewater industry", "nitrogen removal", "municipal wastewater", "circular economy in wastewater industry". In regard to the conventional nitrogen management for removing nitrogen from the wastewater industry, the term "nitrogen management" were replaced with "nitrogen removal" when searching for conventional nitrogen management in the wastewater industry. In researching for Chapter 4, the keyword of "nitrogen recovery in wastewater industry" was highlighted as pathways and technologies for nitrogen recovery can be achieved.

There are several inclusion and exclusion of criteria when reviewing sources. This included the date of publication from 2016 to the present day was reviewed for Chapter 4. Besides, only English-language published sources were considered, as well as only the full-text of journals were available. Lastly, specific nitrogen recovery pathways from a particular industry were not included as well as the focus was towards nitrogen recovery in the municipal wastewater industry which comprises of a mixture of municipal and industrial wastewater.

Step 3: Analysis of Results

The two types of result analysis which are the descriptive and thematic analysis provides different aspects of analysis as well as its criteria were being summarised in Table 3.1.

Table 3.1: Types, Aspect and Criteria of Analysis

Type of Analysis	Aspect of Analysis	Criteria of Analysis
Descriptive	Bibliometric data	▪ Title ▪ Year of publication ▪ Database ▪ Journal ▪ Article ▪ Author ▪ Keywords
	Research methodology	▪ Research objective ▪ Research method ▪ Research procedures ▪ Data source ▪ Data collection
Thematic	Thematic area	▪ Research purpose ▪ Research goals ▪ Definition of thematic area ▪ Research gaps in thematic area

The descriptive analysis is known as the quantitative approach which comprises of the bibliometric data and research methodology. This type of analysis governs the reliability and relevancy of the research that is to be reviewed. In the thematic analysis which covers the thematic area of purpose, goals, definition and research gaps, it focuses more on the qualitative approach to ensure the aim and objectives of the project is met and addressed appropriately without running out from the defined context. The thematic area of this study focuses on nitrogen management and recovery in wastewater industry, implementation and challenges from circular economy as well as pathways and technologies contributing to nitrogen recovery.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction to Nitrogen Recovery

By implementing the concept of circular economy of nitrogen management in the wastewater industry, the term “nitrogen recovery” is coined. It denoted that the nitrogenous compounds present in wastewater effluent was being recovered totally into other sectors such as the agriculture sector to omit the usage of fresh nitrogen feed normally produced through the Haber-Bosch process. There were many factors that leads to the transition from nitrogen removal to nitrogen recovery according to various sources, such as the high energy consumption and GHG emission involved in the nitrogen removal processes, the downside of Haber-Bosch process, the ability to resolve environmental issues, holistic usability of recovered nitrogen as well as the ability to help meet growing food demand as a result of increasing human population.

High Energy Consumption of Nitrogen Removal Processes

According to Ye et al. (2022), the existing nitrogen removal processes such as partial nitrification-anammox processes, nitrification-denitrification as well as other advanced nitrogen removal processes posed a challenge on the high energy consumption. They further stated that the energy range required for the removal of nutrients that includes nitrogen and phosphorus were from 0.39 to 3.74 kWh/m^3. This statement was further supported by a study conducted by Perera and Englehardt (2020) that stated that the conversion process into nitrogen gas through the nitrogen removal processes required high load of energy.

Production of GHG Gas from Nitrogen Removal Processes

According to Ye et al. (2021), GHGs such as nitrous oxide were proned to be generated during the nitrogen removal process which led to global warming occurence. The problem with the production of nitrous oxide gas was also because its exact causes, pathways and mechanism were not exactly understood, therefore causing the emission of nitrous oxide to persist. Besides, other researchers have also discovered the release of methane gas, which is another highly potent GHG to be generated in the removal processes (Ye et al., 2022; Vineyard et al., 2021) Given such negative outcomes it brought on an industrial-scale level, this have led to the inclination of nitrogen recovery pathways in the municipal WWTPs.

Downside of Haber-Bosch Process

The Haber-Bosch process is known as a highly energy-intensive process as it operates at a high temperature and pressure, ranging from 400 to 600 ℃ and 20 to 40 MPa respectively (Zhang and Liu, 2021a). The production of hydrogen gas as the raw material is mainly produced through steam reforming by using non-renewable fuel source such as natural gas with the production of carbon dioxide as the GHG. Thus, the Haber-Bosch process utilised 3 to 5% of natural gas produced globally, at the same time consuming approximately 1 to 2% of the global energy annually. In conjunction to the emission of carbon dioxide, there was an increasing adoption of carbon trading scheme in more countries to mitigate the increasing release of GHGs according to the Paris Agreement. All these information have led to the downside of the Haber-Bosch process and encourages the discovery of nitrogen recovery process.

Besides, Zhang and Liu (2021) had presented an estimated figure of €39 billion/year being spent on the production of ammonia, supposing that all of the nitrogeneous compounds removed were considered in the municipal wastewater treatment plant in a global level. Such have further reveal the economic necessity and the relevance in preserving the environment to recover ammonium from the wastewater industry. Furthermore, Ye et al. (2021) had also mentioned the recovering of nitrogen from the wastewater industry could serve to offset the energy consumption in the Haber-Bosch process. According to another research by Zhang and Liu (2021), a total of 50 TWh energy consumption can be saved when there was even a 5 % decline of the ammonia production through the Haber-Bosch process.

Production of Useful Nitrogen Recovered Products

According to Ye et al., (2021), the main objective for the implementation of nitrogen recovery pathway is its ability to exploit the ammonium compounds present in the wastewater to produce useful products such as fertilizer and food source for humans and animals. Besides, it can also be used to cultivate microalgae for the usage of biogas and biofuels industry. However, many studies have point to the production of green fertilizers as the main function of recovered nitrogen from the wastewater industry (Ye et al., 2021, 2022). By transitioning to the nitrogen recovery pathway, current wastewater treatment plants could be transformed globally to a resource factory.

Meet Needs of Growing Food Demand

Since the increase of food demand as a result of increasing human population, nitrogen-based fertilizers have become a normal commodity to the agricultural industry to produce large amount of crops. With the implementation of nitrogen recovery, Fang et al. (2018) suggested that it may represent a solution to resolving the crisis of the lack of nitrogen fertilizers for crop yield.

4.2 Nitrogen Recovery Technologies

There are various types of nitrogen recovery technologies that have been investigated and discovered by researchers. The list of recovery technologies includes the bio-membrane based integrated system (BMIS), through electrode capacitive deionisation (CDI), coupled aerobic-anoxic nitrous decomposition operation (CANDO) process, ammonium recovery through adsorbents, through chemical processes such as stripping and struvite precipitation and lastly through the usage of microalgae and cyanobacteria. The list of recovery technologies stated here were not exhaustive and will continue to develop over the years. However, here presented the commonly investigated technologies in the recent years for nitrogen recovery. In the following section, each proposed nitrogen recovery technologies were expounded in detailed in terms of its principle and mechanism for undergoing nitrogen recovery to better comprehend the nature of these technologies. Lastly, the comparison of several nitrogen recovery technologies listed earlier as well as not listed were being conducted to examined its rate of nitrogen recovery.

4.2.1 Bio-Membrane Based Integrated System (BMIS)

The application of BMIS is considered as one of the nitrogen recovery technolgoies due to its special ability to recover nitrogen efficiently and sustainably from the wastewater. The usage of membrane technology promises minimal energy consumption and without the addition of chemicals which further added to its benefits in the recovery process, including the separation and enrichment operation involved (Ye et al., 2021). Furthermore, they also stated the incorporation of the biological process with membrane technology also contributes to the removal of foreign substance such as organics materials and heavy metals. Such phenomenon grants several benefits, such as (1) it helps reduce membrane fouling risk and (2) able to transform organic ammonium compounds into recoverable and reusable ammonia nitrogen for its enrichment. Hence, the BMIS here acted as a concentrative technology for the encirchment of ammonium compounds in the diluted municipal wastewater. Since the BMIS alone is insufficient for its direct application in the agricultural industry, additional steps such as struvite precipitation and adsorption may be required to produce the final applicable bioproducts (Ye et al., 2022).

According to Ye et al. (2022), the BMIS can be categorised into two broad categories depending on the location of the membrane in or out of the bioreactor as known as (1) side-stream, and (2) submerged configuration. Table 4.1 summarized the two main types of BMISs. Comparing both side and submerged configuration, the latter configuration is favoured as it required lesser infrastructure and lower energy consumption and costs.

Table 4.1: Two Main Types of BMISs, Its Characteristics and Examples

Types of BMISs	**Characteristics**	**Examples**
Side Stream	Membrane module existed as a separate entity outside the bioreactor. Influent wastewater flow successively through the membrane module first, followed by bioreactor.	• Electrodialysis (ED) • Membrane Distillation (MD) • Forward Osmosis (FO) • Reverse Osmosis (RO) • Nanofiltration (NF)
Submerged	Module membrane within the bioreactor.	• Osmotic Membrane Bioreactor (OMBR) • Bio-Electrochemical System (BES) • Membrane Photobioreactor (MPBR) • Anaerobic Membrane Bioreactor (AnMBR)

Therefore, the following section first highlighted the side stream configuration with the different membrane modules and then followed by the submerged configuration. Figure 4.1 depicted the graphical illustration of the BMISs in achieving nitrogen recovery in the wastewater industry through a circular bio-economy model.

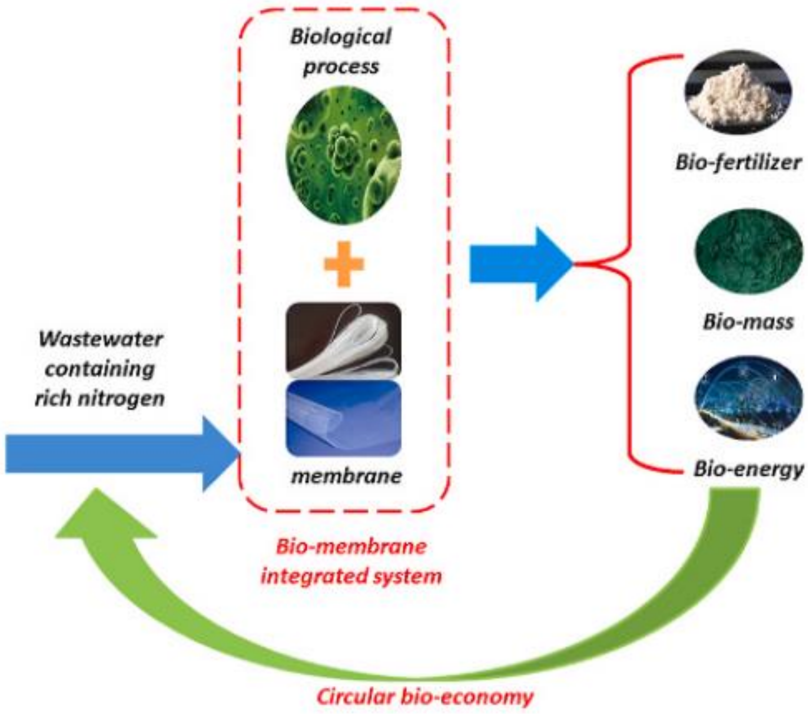

Figure 4.1: Graphical Illustration of Nitrogen Recovery From BMISs Technologies (Ye et al., 2022)

4.2.1.1 Side Stream Configuration

Electrodialysis (ED)

One of the sidestream BMIS is the usage of electrodialysis (ED). It is a separation system powered by electricity with the presence of ion-exchange membranes (IEMs) stacks. The IEMs stack in ED is normally situated between the concentrated and dilute chambers of a multi-chamber cell, and is made up of anion and cation exchange membranes (AEMs and CEMs) placed alternately (Ye et al., 2022). Besides, each compartment is also equipped with an inert electrode. Figure 4.2 below showed the visual illustration of the conventional ED diagram.

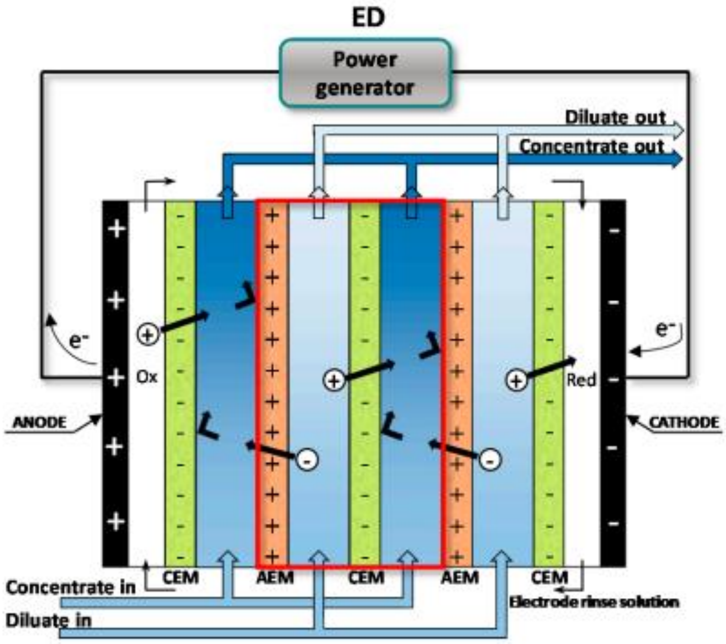

Figure 4.2: Conventional Electrodialysis Schematic Diagram (Gurreri et al., 2020)

According to Vineyard et al. (2021), the predicted lifetime of the ED membranes was 10 years. Thus, the ED technology was considered as a mature separation technology that relied on the presence of electrical field to move ions present in the wastewater influent to pass by IEMs and subsequently allowing ammonium ions to be concentreated at the cathode chamber across CEM. The ammonium ions can then be further recovered through a variety of methods such as chemical precipitation, air stripping or adsorption according to Ye et al. (2022). Due to the complete reliance of electricity by ED, the migration of ammonium ions is primarily dependent on the amount of electrical current supplied. In fact, the multiplication of voltage and electric current supplied to the ED stack made up the total energy consumption, and there existed a complex relationship between the stack conditions and the resultant voltage according to Vineyard et al. (2021). They have also conducted a research and found out that there was a 26 % reduction of energy consumption in the ED as compared to the conventional nitrification-denitrification reactor in the nitrogen removal process.

However when compared to the anammox reactor, ED consumed 64 % more electricity. Besides, Vineyard et al. (2020) also predicted the energy consumption of 2.36 kWh per kg of nitrogen recovered given an economically optimal ED stack. It is also important to note that the performance of ED can be negatively affected by the high electrical resistance of wastewater influent. Hence, the process of in-situ pH control of the wastewater influent is paramount for the application of ED, which can be resolved through the application of bipolar membranes (Ye et al., 2022).

Membrane Distillation (MD)

The membrane distillation (MD) is a temperature dependent process where the difference in temperature between the feed and permeate acted as the catalyst in which vapour molecules migrate and eventually diffuses through the hydrophobic or gas permeable membrane (Ye et al., 2022).Volatile nitrogenous compounds such as ammonia at the wastewater influent can pass through the membrane to the permeate side even under low pH condition and subsequently put into use the condensed ammonium solution. In the MD process, there were two important parameters that would affect its functionality, which are the temperature and pH of the wastewater feed containing nitrogenous compounds according to Ye et al. (2022). A study was conducted by Noriega-Hevia, et al. (2020) which revealed that the pH of the solution in MD would be the most crucial factor for concentrating the nitrogen. A slightly alkali pH value of 9.26 and above was found beneficial to the formation of ammonia according to Munasinghe-Arachchige, et al. (2021). It is also known that the MD process had lower energy requirements (0.22 – 1.2 kWh/kg·N) as compared to conventional acid absorption and stripping of ammonia (23.6 – 49.6 kWh/kg·N). Figure 4.3 showed the graphical illustration of the direct-contact MD configuration.

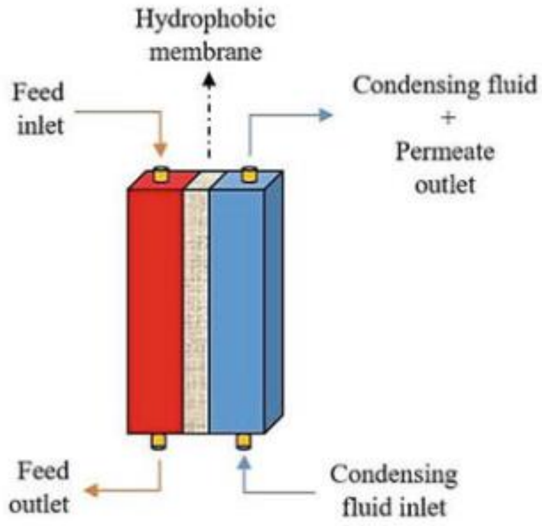

Figure 4.3: Direct-Contact MD Configuration (Rahimpour, 2020)

Forward Osmosis (FO)

The forward osmosis (FO) process is unlike reverse osmosis (RO). In FO, the driving force lies at the osmotic pressure between two different concentration of solution; while in RO, it relied on the hydraulic pressure. The FO process functions by having feed and draw solution with different chemical potential to drive ions across the membrane from a low concentration of feed solution to a high concentration of draw solution as opposed to RO. According to Ye et al. (2022), the recovery of nitrogen from wastewater influent through FO process was conducted by concentrating ammonium ions at the feed side by the transportation of water from feed into the draw solution through the FO membrane with an appropriate amount of draw solute present in the draw solution. According to a research conducted by Engelhardt et al. (2020) that utilised aquaporin-based membranes in forward osmosis for the treatment of urea, it was recorded that approximately 100% of the ammonium ions in the urea can be rejected after some pretreatment strategy. Nonetheless, this study by Engelhardt et al. (2020) have shown that the process of forward osmosis with a suitable type of membrane can be used to concentrate and retained the ammonium ions at the feed side, provided that pretreatment of wastewater influent may be necessary.

Besides, the recovery of nitrogen through FO process is more environmentally friendly and cost effective when compared to other side-stream technologies according to Ye et al. (2022). However, a major setback in the FO process would be its high energy consumption for the regeneration of draw solutes from the diluted draw solution This posed a huge limiting factor for the commercialization of this process in many industries, one of which is in the wastewater industry. Hence, research should be focusing on how to regenerate draw solutes in the FO process in an economical feasible way. Chekli et al. (2016) have proposed the feasibility of integrating FO with MD as the solution by stating that MD as a post-treatment was capable of recovering fresh water from the diluted draw solution from the initial FO process. With this, the draw solute can be regenerated and so fulfilling the concept of circular economy. An example of the application of FO process in the wastewater industry was conducted by Singh et al. (2019), where 66 % of ammonia can be recovered within 24 hours from the sewage by using divalent magnesium chloride as the draw solution. Besides, regular clean-up were being conducted of the used biomimetic aquaporin membranes to allow for the performance restoration of original membrane after every 24 hours cycle.

Reverse Osmosis (RO) and Nanofiltration (NF)

Both reverse osmosis (RO) and nanofiltration (NF) can be categorized into one type of side-stream BMIS due to its similarity of having high rejection rates of salts and ions, and both were considered high pressure membrane processes (Ye et al., 2022). Besides, several studies have proven that both of these technologies were used frequently in the recovery of ammonium from urine (Patel, Mungray and Mungray, 2020; Ray, Perreault and Boyer, 2020). Such as an example, Ray, Perreault and Boyer (2020) have discovered that a total percentage of 64 % and 90 % of unionised ammonia can be recovered from urine that has been hydrolysed through RO and NF processes respectively. pH optimization is the key factor in ensuring all of the ammonium ions can be enriched through the RO membrane as the dominant form of ammonium-nitrogen exists as NH_4^+ form at pH less than 7 (Ye et al., 2022).

However, there are still some differences between RO and NF. NF is characterized of its ability to separate molecules ranging from 1- 10 nm due to its pore size ranges from 1 to 5 nm which contributes to the rejection of very minule size of molecules. Besides, NF requires lower pressure and energy input as compared to RO. The very small pores sizes of NF render its ability to reject almost all micro pollutants such as phosphate, and at the same time allowing nitrogenous compound to enrich by permeating through the membrane.

4.2.1.2 Submerged Configuration

Osmotic Membrane Bioreactors (OMBR)

Osmotic membrane bioreactors (OMBRs) consists of the integration of biological process with forward osmosis (FO) membrane module. When compared to conventional and traditional approach of MBRs that utilises microfiltration (MF) or ultrafiltration (UF), OMBRs poses more advantages such as showing superior water quality, ability for reversible fouling, lower susceptibility of membrane fouling and decreased energy requirements. Such is the case due to the following two reasons: (1) it operates at ambient temperature and (2) it requires significantly lower or no hydraulic pressure as replaced by the use of osmotic pressure. A major factor contributing to the use of OMBRs is due to the replacement from hydraulic to osmotic pressure that enhanced the removal of various contaminants as well as rendering the washing process of the fouling layer on the membrane with greater ease. These have contributed to the reduction of costs of cleaning and increment of permeate flux (Ye et al., 2021).

The principle of nitrogen recovery in the OMBR is that the FO membrane acts to reject the ammonium compounds in the influent municipal wastewater, thus causing it to accumulate on the feed side and coupled with struvite precipitation for the eventual recovery of usable ammonium compounds by means of increasing the pH or through the addition of chemicals if necessary (Ye et al., 2022). Ye et al. (2021) have discovered a study conducted by Qiu and Ting where as high as 97 % of the ammonium ions can be recovered and enriched from the influent wastewater, alongside with other compounds such as phosphate and magnesium ions although some of the ammonium compounds are consumed for microbial activity in the bioreactor. Therefore, approximately 80 % of the ammonium compounds are then recovered through struvite precipitation when the pH range is being adjusted between 8.0 to 9.5 (Ye et al., 2021, 2022)

It is known that further improvement of the OMBRs in terms of technical and economical feasibility is possible through the addition of UF/MF membrane with the FO membrane as shown in Figure 4.4. Besides, the addition of a fixed bed biofilm at the feed side could also significantly removed suspended solids that aided the reduction of risk for FO membrane fouling. Lastly, RO and MD can also be applied to OMBRs to mitigate the issue of recovering water from the draw solution, thus regenerating the draw solute and increases overall feasibility of OMBRs (Chang et al., 2017). Several factors that affected the efficiency of OMBR includes (1) the type of membrane, (2) pH value, (3) sludge retention time, and lastly (4) temperature according to Ye et al. (2021).

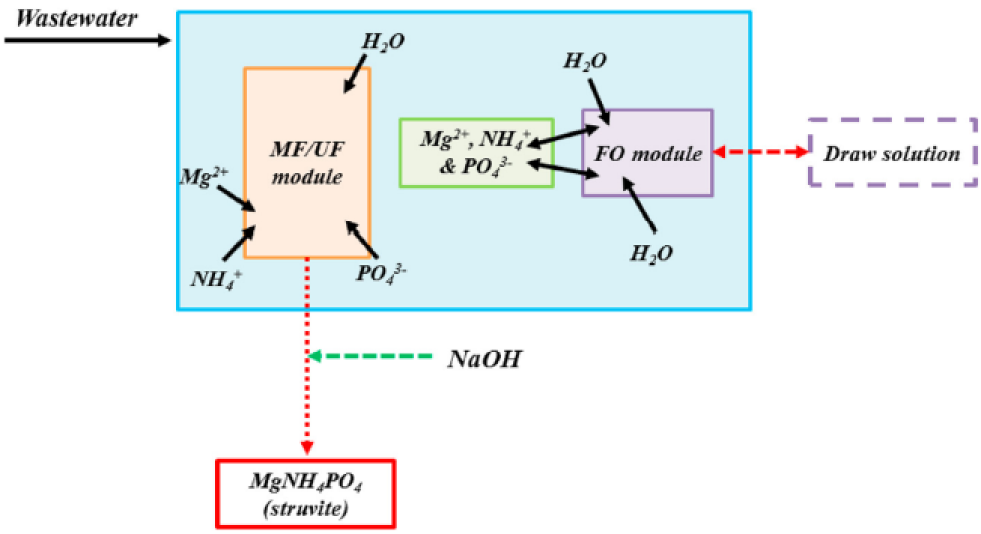

Figure 4.4: Schematic Diagram of Osmotic Membrane BioReactor (OMBR) (Ye et al., 2021)

Bioeletrochemical System (BES)

According to Ye et al. (2021), the bioelectrochemical system (BES) consists of two chambers divided by a cation-exchange membrane (CEM). In the BES, the process of chemical energy found in the waste organics residue in wastewater influent is being converted into electrical energy by electrochemically active bacteria at the anode chamber by means of catalyzed reactions (Sun et al., 2019).

The mechanism involved in the process is that while heterotrophic microbes oxideses the organic waste, it donates free moving electrons to the anode, in which it would be transported through an external circuit and finally reaching the cathode. Once the electrons have reached the cathode chamber, it is being reduced by electron acceptors such as air. Figure 4.5 illustrated the overall schematic diagram of a BES for ammonium recovery. According to Ye et al. (2022), BES had the ability to not only allowed recovery of ammonium from wastewater, but also simultaneously undergo water purification and recover energy. The BES can be categorized mainly into: (1) microbial electrochemical cell (MEC), (2) microbial fuel cell (MFC), and (3) microbial desalination cells (MDC).

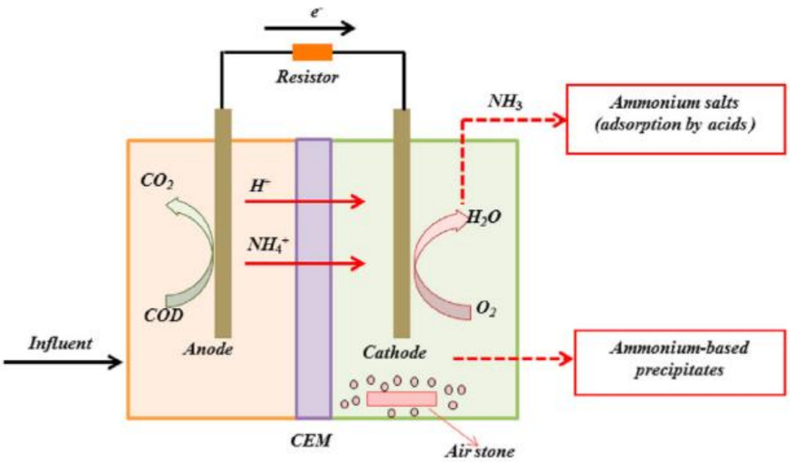

Figure 4.5: Schematic Diagram of BES for recovery of ammonium (Ye et al., 2021)

There are differences between the three main types of BES as mentioned, with its different thermodynamic reaction favourability being the major factor for choosing amongst the three. According to Zhang and Liu (2021b), the presence of applied voltage as the driving force in MECs grants better transfer of ammonium ions to the chathode chamber which aids in increasing the ammonium concentration as compared to MFCs. However, another point of view from Ye et al. (2021) mentioned that MECs is thermodynamically unfavourable, that is unlike MFCs whose uses a thermodynamically favourable reaction, which results in extra external electricity required to operate MECs. In the context of nitrogen recovery, MECs would stand out to be a better option compared to MFCs due to the additional voltage power supplied for the more efficient transport of ammonium ions from the anode to the cathode (Ye et al., 2022). This ultimately contributes to the ammonium enrichement for further recovery process through stripping or chemical precipitation.

As for the application of MDCs, it is mostly used in the wastewater treatment with the main objective to recover ammonium and also simultaneously undergoes recovery of resources, desalination and purification of wastewater that includes landfill leachate and anaerobic digester liquor (Ye et al., 2022). Zhang and Liu (2021b) also stated that the removal of ammonium from the wastewater influent at the anode is determined by a number of factors: (1) concentration of ammonium compounds in wastewater, (2) removal efficiency of ammonium from the cathode head space by means of stripping action, (3) the amount of current applied, (4) pH of electrolye in cathode, (5) the properties of ion exchange membrane, and lastly (6) the state of equilibrium of other ions at both side of anode and cathode chamber. Through the application of BES, energy recovery in the form of hydrogen or production of electricity aids in enchaning the market potential of BES in the process of recovering ammonium. Although it is known that the problem of CEM fouling persists, it is being offset by the generation of electricity which could alleviate the issue.

Membrane Photobioreactor (MPBR)

The usage of MPBR acts as a phototrophic system in which it utilises the energy stored in the sunlight to reduce the amount of nitrogenous compound present during the wastewater treatment process. One of the major class of phototrophic organisms used alongside in MPBR is microalgae. They can be found in waterbodies such as in freshwater and marine systems and are characterised with its quick growth rate and ability to adapt to harsh environments. The current use of photobioreactor (PBR) to cultivate the microalgaes in wastewater treatment systems is used to perform oxygenic photosynthesis and the decomposition of organic wastes into simple inorganic nutrients (Ye et al., 2021). Furthermore, the photo-autotrophical growth of these microalgae relies water as the only electron donor and at the same time requires constant presence of light. The downside of the currently practiced PBR is that solid separation is a huge challenge due to the poor settlement capacity of microalgae that makes the separation of solids difficult. Therefore, the invention of MPBR that incorporates the membrane technology serves to be a new solution to not only mitigate the solid separation process to make it less energy-intensive, but also provides more compact reactor footprints and improves the accumulation and growth of biomass in the MPBR (Abouhend et al., 2018). Besides, MPBR is also a more environmental friendly approach as compared to the conventional nitrogen removal processes. Ye et al. (2022) further mentioned that MPBR has the advantages of reducing energy consumption and chemical usage due to the simpler operation processes, simple to scale up and capable for excellent fractionation. Figure 4.6 portrayed the schematic diagram of microalgae MPBR.

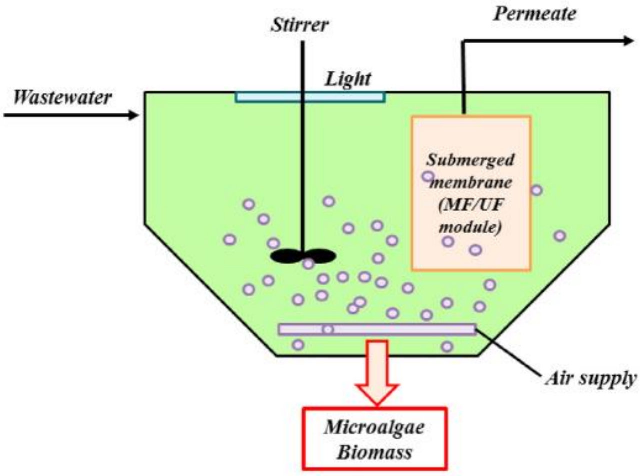

Figure 4.6: Schematic Diagram of Miroalgae MPBR (Ye, et al., 2021)

Due to the substantial amount of phosphorus and nitrogen present in municipal wastewater influent, the usage of MPBR help promotes the fast growing microalgae. According to Baral et al. (2020) and Liao et al. (2018), the advantage of using MPBR was that operational parameters of the bioactivity of microalgae can be controlled which includes the aeration, temperature, frequency of light, spectral composition, power and water and nutrient resources. With the usage of MPBR, ammonia can be assimilated by microalgae for the conversion into biomass for better application as compared to conventional process of releasing nitrogen gas back into the atmosphere. A study conducted by Chang et al. (2019) found that a high-efficiency of 74.31% of nitrogen can be recovered from landfill leachate by using a scalable MPBR. Nguyen et al. (2021) also conducted a research where a high nitrogen recovery rate of TN 90.5 mg/L·d can be achieved from urine for the cultivation of microalgae in MPBR.

Anaerobic Membrane Bioreactor (AnMBR)

AnMBR is the combined anaeroabic reaction with membrane separation in the submerged mode. It grants more advantages as compared to conventional MBR processes, such as having higher COD capture efficiency with minimised waste sludge generation, decreased demand of energy due to the absence of aeration, generation of clean energy from biogas as well as producing permeate that is free of solids and rich in nutrients (Zhang and Liu, 2021a). AnMBR has currently been used widely in the municipal wastewater treatment due to its ability to directly reuse the ammonium present in wastewater influent for irrigation purposes. This does not only aid economically but also contributes to a cost-effective municipal wastewater reclamation process due to the practically feasible engineering option. Such as an example, the AnMBR have shown its ability to achieve removal rate of COD up to 90% and almost 100 % of the solids with the minimum consumption standard levels of soluble ammonium and phosphate according to Wu et al. (2017) and Gu et al. (2019). Since it is able to produce high quality of clean permeate, it can be used for direct agricultural irrigation. Nevertheless, the transportation of permeate to the agricultural lands still is a concern for practical applications.

In terms of AnMBR potential for nitrogen recovery from wastewater influent, Grossman et al. (2021) have conducted a research in the application of AnMBR in the food processing wastewater. It is found that a nitrogen recovery percentage of 77 % is discovered and 57 % of the total organic carbon was also being recovered by means of methane production. According to an estimation conducted from Zhang and Liu (2021a), approximately 20 millions tons of ammonium in AnMBR permeate can be recovered and be directly used for irrigation purposes without the need for further purification as a substitute for chemical fertilizers if and only AnMBR is applied globally in the municipal wastewater treatment systems. Therefore, the permeate from AnMBR that contains soluble nutrients should be considered for the usage of chemical fertilizers as this will only only lead to a drastic decrease usages of chemical synthetic fertilizers, but also supplying freshwater for irrigation application (Zhang and Liu, 2021a).

4.2.2 Electrode Capacitive Deionization (CDI)

According to Fang et al. (2018), there are many present pre-concentration technologies to produce ammonia-rich solution as the first step towards nitrogen recovery from wastewater. However, the process of recovering nitrogen from low concentration municipal wastewater is almost impractical due to high cost and energy incurred. For instance, the utilization of ion-exchange technologies for nitrogen recovery requires complex regeneration and secondary waste is needed to be processed. Besides, the RO technology requires high energy consumption and operational cost with a poor rate of water recovery. Therefore, the emergence of capcacitive deionization (CDI) technology, also known as electrosorption is in place to alleviate the constraints of high cost and energy requirement as it brings the following advantages: (1) low cost, (2) energy efficiency, (3) environmental friendly, and (4) increased academic interest for future research and development. Its working mechanism is through the adsorption of charged ions (ammonium ions) from the wastewater into the electric double layers (EDLs) which led to its formation at the electrode and solution interface by the existence of potential differences across anode and cathode (Fang et al., 2018).

There is a type of CDI that has been explored, known as the membrane capacitive deionization (MCDI) cell. It is constructed through the usage of CEM and AEM assembled in front of the electrodes. Though MCDI was capable of blocking approximately all of the co-ions present and have increased performance of deionization and lower energy consumption, the solid electrodes present have limit the adsorption capacity by the longitudinal dimensions of the electrodes. Such a phenomena caused the low adsorption capacity of ammonium ions by the MCDI cell. To mitigate these shotcomings, the invention of flow-electrode capacitive deionization (FCDI) technology equipped with suspended carbon materials was able to operate continuous and shows high salt removal efficiency of up to 95 % when tested in a concentrated NaCl solution at 32.1 g/L. Its ion-desorption mechanism is conducted by mixing the flow of cathode and anode that allows the recovery of ammonium ions adsorbed on the surface of activated carbon (AC). As compared to previous MCDI and conventional CDI, FCDI the following advantages: (1) larger desalination capacity, (2) allowing energy storage and recovery, (3) selective ion separation, (4) ability for cell design for scaling up and (5) more optimized flow-electrode. Despite the following potential advantages, there are only few investigations focused on the enrichment of ammonia nitrogen from the wastewater with a FCDI unit. Figure 4.7 and 4.8 have illustrated the module structure and the setup of the FCDI technology respectively.

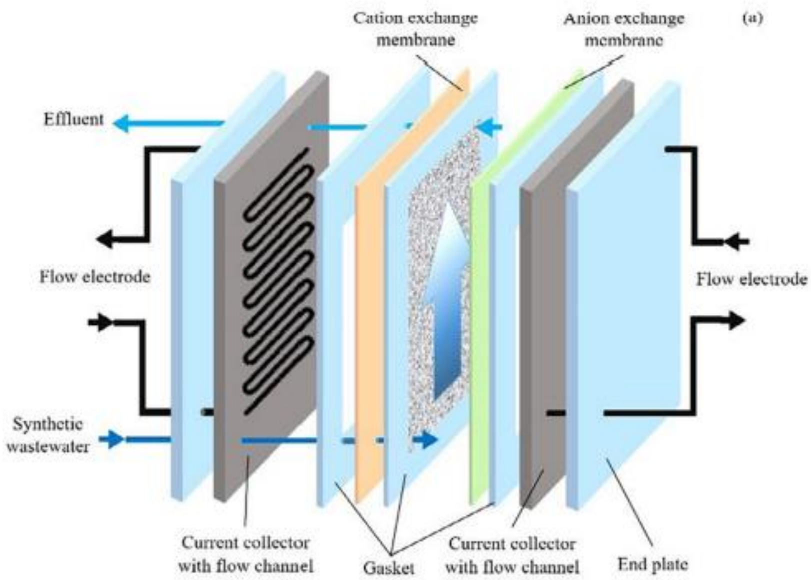

Figure 4.7: Flow-Electrode Capacitive Deionization (FCDI) Cell (Fang et al., 2018)

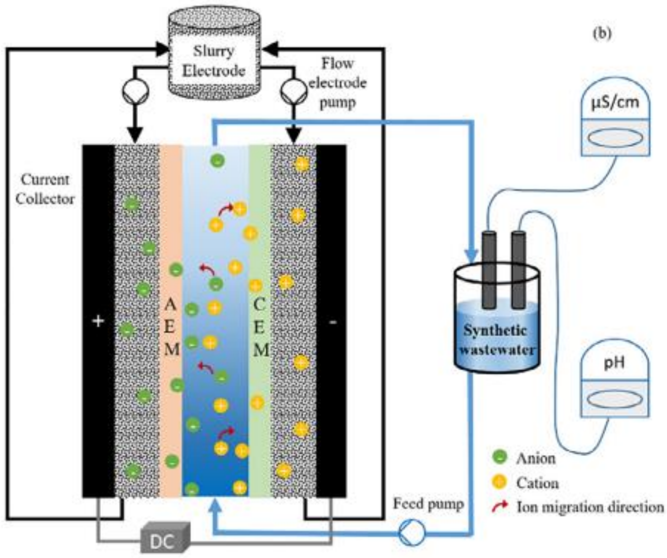

Figure 4.8: Operation Setup of Flow-Electrode Capacitive Deionization (FCDI) Cell (Fang et al., 2018)

4.2.3 Coupled Aerobic-Anoxic Decomposition Operation (CANDO) Process

In the CANDO process, it is aimed to recover the potent GHG of nitrous oxide (N_2O) from the municipal wastewater influent to be used as useful fuel oxidizer according to Zhang and Liu (2021a). From previous discussion it was known that N_2O gas carries strong greenhouse effect and is a form of reactive nitrogen with a certain amount of chemical energy. Through the CANDO process, the production of N_2O is increased by means of anaerobic digestion of liquor with the following biological reactions taking place among different types of nitrogen species: ammonium to nitite, followed by nitrite into nitrous oxide gas. The final reduction process of nitrite into nitrous oxide can be carried out in high efficiencies through the aid of accumulated poly-hydroxybutyrate as the oxidising agent generated by the decoupled strategy substrate addition strategy of the CANDO process (Zhang and Liu, 2021a). The final step for the oxidization of nitrous oxide as fuel was to be subjected to co-combustion with methane gas for the generation of energy, which resulted in 30 % more energy production compared to oxygen given the equal amount of mole of methane and oxygen being combusted.

From this CANDO process, 60 to 80 % of the nitrite produced could be transformed into nitrous oxide to be used as fuel oxidizer. However, due to the small amount of recoverable nitrogen ammonium compounds present in the wastewater influent, there placed a limit on the amount of nitrous oxide that can be produced from the municipal wastewater industry. Apart from generating nitrous oxide through the CANDO process, Wang et al. (2021) stated that the inhibiton of cytochrome proteins in biological wastewater treatment processes also aids in the production of nitrous oxide. However, this method of recovering nitrogen from the municipal wastewater influent required more comprehensive assessment due to its environmental sustainability and economic viability for the recovery of nitrous oxide (Zhang, Gu and Liu, 2019). A visual illustration on the overall CANDO process through anaerobic and anoxic processes has been granted in Figure 4.9.

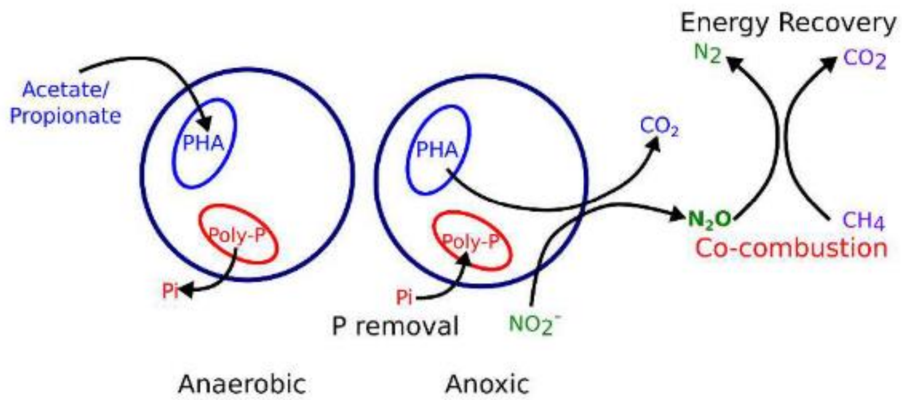

Figure 4.9: Overall CANDO Process With Anaerobic and Anoxic Processes (Gao et al., 2017)

4.2.4 Ammonium Recovery Through Adsorbents

According to Zhang, Zhang and Liu (2020), the simplistic design configuration in the process of adsorption has the potential for the recovery or removal of ammonium compounds from municipal wastewater. There are a variety of common adsorbents materials available for the recovery of ammonium compounds with different adsorption capacity under varying contact time and pH values as listed in Table 4.2. These were compiled by Zhang and Liu (2021a) from various researchers. It is known that from Zhang and Liu (2021a) that the ammonium recovery through adsorption is highly in demand in the municipal wastewater. Therefore, it is of paramount importance to choose a novel and suitable process of adsorption to ensure maximum recovery of ammonium compounds. There were three different types of adsorbents highlighted by Zhang and Liu (2021a), which are the (1) natural and modified zeolite/inorganic adsorbents, (2) biochar, and (3) ion exchange resins and others. The following section briefly summarised the three types of adsorbents for ammonium recovery.

Table 4.2: List of Adsorbent Materials for Recovery of Ammonium Compounds from Wastewater (Zhang and Liu, 2021a)

Adsorbent Materials	Ammonium concentration (mg N/L)	Adsorption Capacity (mg/g)	Contact Time (min)	pH
Natural Iranian zeolite	40	8.51 – 10.3	30	7.0
$NaNO_3$ modified zeolite	1 - 20	16.9	400	5.0 – 8.0
Bentonite/Chitosan	10 - 40	15.9	180	6.0
Synthetic zeolite from fly ash	50	23.9	75	8.0
Modified cornhob-biochar	40	12.1	120	-
Resin C150H	25-150	28.2	90	6.5
Polyacrylic acid-based hydrogel	50 - 180	8.8 – 32.2	240	7.1

Natural and Modified Zeolite/Inorganic Adsorbents

There are a variety of adsorbents such as natural zeolite, clay, bentonite and more that are being used in to recover ammonium from wastewater. It is a type of nagetively-charged porous aluminosilicate mineral which gives a very huge inner surface area. It functions by utilising the electrostatic interaction and relying on the structure of the aluminosilicate material in providing the strong cation exchange capability for the zeolite. However, natural zeolite is limited in terms of recovering ammonium from the municipal wastewater because of its low adsorption capacity at approximately or under 10 mg/g.

Therefore, various modficiations techniques of the natural zaolites is essential such as through surface coating and heat treatment methods or artificial synthesis by acid, alkali, salt and microwave. The resulted modified zeolites have demonstrated improved adsorption capacity. For instance, Liu et al. (2018) have discovered that zeolite that have undergone astificial synthesis from fly ash without utilizing solvents have showed increased adsorption capacity of

18.4 mg ammonium/g at a relatively high 75 mg/L of ammonium concentration. Hence, further modifications of the natural zeolites is needed to increase the adsorption capacity of the weak-ammonium-affinity natural adsorbents in the municipal wastewater. However, a thorough economic and environmental analysis of the long term usage of modified zeolites should be considered (Zhang and Liu, 2021a).

Biochar

According to Yuan et al. (2019), biochar was made by thermally decomposing carbon-rich biomasses under decreased oxygen level. The ammonium adsorption mechanism associated with biochar is based on the interaction between acid groups on the surface of biochar and ammonium through amine salts formation, and at the same time cations on the biochar’s surface can be exchanged with the ammonium ions in the wastewater.

The physicochemical properties of biochar is affected by the types of raw materials that are being thermally decomposed and the condition of pyrolysis. For instance, biochar that is derived from wood possesses highly crystalline structure with more pores and micropores. On the other hand, biochar derived from rice and coconut husk does not possess crystallaine structure as it is more amorphous, and lesser pores structure. However, some biochar were found to have low ammonium adsorption capacity, which suggests that modification of biochar is essential for the increased ammonium adsorption capacity in treating municipal wastewater. Hence, two methods have been proposed to increase its adsorption capacity which are the calcination of raw materials with oxygen-containing functional group at different temperatures, and pretreatment of biochar using alkali or acid to generate more negative charges on the surface of biochar for increased ammonium adsorption capacity according to Yuan et al. (2019).

Ion Exchange Resins and Others

The usage of cation exchange resin relies on its networking structure with the functional groups that could be exchanged with ammonium ions in the wastewater. Yuan et al. (2019) also quoted that sulfonated polystyrene type resin is able to achieve an adsorption capacity of 10.4 to 26.6 mg ammonium/g for the removal of ammonium specifically from the municipal wastewater. However, as compared to the previous two types of adsorbents (zeolite and biochar), ion exchange resins are more costly due to its increased adsorption capacity amongst. Such as an example is the cost of 1 kg of Dowex 50WX8. resins is US$ 390 (Zhang and Liu, 2021a).

Other types of adsorbents such as the hydrophilic hydrogels was found to be able to bind ammonium ions through complexation, covalent bonding, electrostatic interations as well as physical adhesion according to Cruz et al. (2019). Therefore, this rendered the hydrogels to be equipped with fast and high adsorption rate for ammonium removal/recovery. Besides, it was discovered that commercial polyacrylic acid (PAA)-based hydrogel with the initial adsorption capacity of 8.8 mg-ammonium/g at municipal wastewater with 50 mg/L of ammonium concentration can be further increased to 30 mg-ammonium/g after modification with chitosan through the studies conducted by Zheng and Wang (2009) and Cruz et al. (2019). Lastly, nanohydrogel as the ammonium adsorbent was also explored and proposed where it was able to promise a maximum capacity of ammonium adsorption as high as 57.6 mg/g with astounding characterizations (Wang et al., 2014b; Zhang and Liu, 2021c). However, this technology was still in its early developmental stage and more researches and studies needs to be conducted before applying in a large scale implementation.

4.2.5 Stripping and Struvite Precipitation

The stripping and struvite precipitation technology are mature technologies used for the recovery of nitrogen from the wastewater influent (Rahimi, Modin and Mijakovic, 2020b). It can be used for the removal as well as the recovery of ammonium ions. As for the stripping technology, ammonium ions are recovered to ammonia gas by means of forcing of air or other gases into the wastewater. There are four steps involved in the ammonia stripping process, which are as follows: (1) Ammonium ions being converted to ammonia gas, (2) Diffusion of ammonia gas to the air-water interface, (3) Release of ammonia gas at the interface, and lastly (4) Diffusion of ammonia from interface to air above it. Several parameters in the ammonia stripping process governs its effectiveness, such as pH, temperature and the area available for mass transfer to take place. According to Limoli, Langone and Andreottola (2016), the usage of air, steam and biogas were available for the release of ammonium ions from the wastewater influent. Besides, the process of ammonia stripping is also available to be conducted in continuous and batch mode. As ammonia is being recovered from the wastewater, several chemical reagents can be added into the wastewater to absorb the emitted ammonia such as phospric acid and sulphuric acid according to stoichiometric concentration (Shen et al., 2017; Rahimi, Modin and Mijakovic, 2020a). With the ammonium ions being recovered, it can be used to convert into fertilizers with 40 to 60 % ammonium fertilizer solution with low organic contamination after the neutralization process. Therefore, the ammonia stripping process is an effective nitrogen recovery method in the wastewater industry to support the agriculture industry by means of supply of organic fertilizers.

Struvite precipitation is one of the effective means for nitrogen recovery from the nitrogen-rich municipal wastewater. Such recovery method is highly effective, simple and environmental friendly as the nitrogen can be recovered as fertilizers (Rahimi, Modin and Mijakovic, 2020a). Struvite is a while crystalline solid and acts as a valuable fertilizer formed through the following chemical reaction:

$$Mg^{2+} + NH_4^+ + PO_3^{2-} + 6H_2O \rightarrow MgNH_4PO_46H_2O$$

However, this process is affected mainly by two factors, which are the molar ratio of Mg:NH4:P and pH whose optimum pH range is 9 to 10 according to Cao et al. (2019). Successful struvite precipitation for the removal of 95 % of ammonium ions, the Mg:NH4:P ratio should be set to 1:1:1 and struvite crystals are precipitated when the pH increases (Rahimi, Modin and Mijakovic, 2020a). However, this type of technique faces the challenge of treating wastewater with high ammonium and phosphorus content as this would require external source of magnesium for its precipitation.

4.2.6 Usage of Microalgae and Cyanobacteria

According to Rahimi, Modin and Mijakovic (2020a), microalgae and cyanobacteria are unicellular species of microscopic photosynthetic microorganisms found in most of the water bodies. Due to their ability to withstand under harsh environment, they can be considered to be used in the wastewater industry in the biological wastewater treatment. Their presence in the wastewater helped produce oxygen through photosynthesis, in which bacterias in the wastewater can utilise the oxygen generated to decompose organic waste into simple inorganic nutrients. Furthermore, they are often used in the tertiary treatment process for the removal of inorganic nutrients before wastewater is being discharged into the environment. These microbs fed on the nitrogen and phosphorus present in the municipal wastewater for its growth and development which render it useful in the recovery of nutrients which in this specific context refers to nitrogen. With these microbs, nitrate and ammonia can be assimilated to convert into biomass rather than the conventional way of releasing nitrogen gas back into the atmosphere. The factors that influenced the nitrogen recovery rate through these microbes is dependent on the ammonification and assimilatory reduction of nitrite into ammonium (Rahimi, Modin and Mijakovic, 2020a).

4.2.7 Comparison of Nitrogen Recovery Technologies

In this section, the different nitrogen recovery technologies were being compared in treating different wastewater sources. The parameters involved were its source of wastewater, pH, initial total ammonium concentration, percentage of nitrogen recovery, form of recovered nitrogen and its experimental stage. This section aimed to highlight the percentage of recoverable nitrogen with the given source of wastewater for treatment. The source of wastewater were narrowed down to municipal wastewater sources as of the target of this project. The technologies involved here consist of those mentioned earlier, as well as those not listed. The detailed comparison of the nitrogen recovery techniques in treating different wastewater sources were tabulated in Table 4.3.

Table 4.3: Comparison of Nitrogen Recovery Techniques in Treating Different Wastewater Sources (Beckinghausen et al., 2020a)

Nitrogen Recovery Techniques	**Source of Wastewater**	**pH**	**NH⁴₊ (mg/L)**	**TN (mg/L)**	**N Recovery (%)**	**Form of Recovered N**	**Experimental Stage**
Vacuum thermal stripping/acid absorption	Municipal sludge digestate	-	1093	-	100	Ammonium sulphate crystal	Pilot
Microbial Electrolytic Cell	Municipal reject water	-	1000	-	100	Ammonium chloride	Lab
Electrolytic Cell with Stripping/absorption	Undiluted urine	9.2	5100	-	53 - 77	Ammonium sulphate solution	Lab
Hydrogen Recycling Electrochemical System and Transmembrane ChemiSorption	Preteated human urine	9	-	3400	100	Ammonium sulphate solution	Lab
Stripping/acid absorption	Undiluted urine	9.6	-	4500	100	Ammonium sulphate solution	Lab
Enlarged Microbial Nutrient Recovery Cell	Raw domestic wastewater	-	47.4	-	62	Struvite	Lab

Bio-electrodialysis	Domestic wastewater primary clarifier effluent	7.8	70	-	80	Ammonia in boric acid	Lab
Electrodeionization	Synthetic domestic wastewater	-	28,700	-	100	Solution	Lab
Microbial Electrolytic Cell – Forward Osmosis	Synthetic side stream centrate	-	1,000	-	100	Struvite and ammonium sulphate	Lab
Resource Recovery Microbial Fuel Cell	Wastewater containing urine	-	3.89	-	42	Solution	Lab
Microbial Nutrient Recovery Cell	Domestic wastewater	-	23.8	-	24	Solution	Lab

Based on Table 4.2, it can be deduced that the percentage of recoverable nitrogen was proportional to the concentration of nitrogen in the wastewater source. The major concern for nitrogen recovery from municipal wastewater lies on the low-strength wastewater, which can be mitigated by means of incorporating the process of preconcentration of ammonium compounds, also known as ammonification before it can be recovered by struvite precipitation or stripping coupled with acid adsorption. With the six nitrogen recovery techniques mentioned in this subsection, the BMISs and FCDI both possessed the potential for the recovery of nitrogen. As of BMISs, the integration of both membrane process for ammonification followed by the biological process to remove unwanted organic, inorganic matters and heavy metals contributed to the holistic recovery of nitrogen. The FCDI method also shows excellent recovery of ammonium compounds from diluted municipal wastewater, such as its ability to preconcentrate multiple times of 20 compared to the initial concentration of low-strength wastewater up to 322.06 mg N/L (Fang et al., 2018).

4.3 Recovered Bioproducts in Nitrogen Recovery

Bioproducts can be recovered from the previously mentioned nitrogen recovery technologies. In the following section, each of the contribution of potential bioproducts will be expounded in nitrogen recovery towards bioeconomy, which consists of biofertilizer, microalgae biomass and bioenergy.

4.3.1 Biofertilizer

Biofertilizers derived from wastewater industry have been actively sought out by researchers to replace the conventional chemical fertlizers due to its severe consequences it has towards the environment, plants and soil. According to Dineshkumar et al. (2018), the usage of synthetic fertilizer have causes elevated soil erosion issue, while the improper use of nutrient-based fertilizer have caused the reduction of plants and crop yield that leads to negative consequences to the environment. In light of these issues, biofertilizers were sought out as an alternative source as it is more resource-efficient and eco-friendlier choice for its usage in the agriculture industry. There are four types of biofertilizers, namely struvite, ammonium sulphate, biochar and microalgae-based fertilizer which are described as follows.

Struvite

The generation of struvite as biofertilizers in the nitrogen recovery system is able to replace conventional fertilizers in the growth of plants and crops yield as it contains the important nutrients such as nitrogen and phosphorus. Song et al. (2011) mentioned that one of the characteristic of struvite was its slow release of nutrient that made it a more advantageous choice over other fertilizers. Besides, the presence of magnesium in the struvite also render it an effective fertilizer for grasses since magnesium is an important element in chlorophyll. Lastly, struvite contains lower content of heavy metal ions as compared to conventional fertilizers which poses less harm towards the plant roots.

Although there were varying research outcomes on the effectiveness of struvite as compared to conventional fertilizers in which one claimed to have the same effectiveness while the other observed a lower crop yield treated by struvite fertilizers, it can be concluded that the combined addition of struvite and other fertilizers can satisfy the requirement of nitrogen for the healthy development of plants. This was because excess struvite application will lead to increase of soil pH and negatively affects the nutrient uptake and availability of the soil. Hence, Rahman et al. (2014) concluded that the application of struvite acted as an intermediary strategy between global nutrient transfer and recycling of nutrient in agriculture land. Therefore, the large-scale execution of nitrogen recovery of wastewater treatment plants can aid the existing expensive production of industrial ammonium through the Haber-Bosch process.

Ammonium Sulphate

Although the downside of this type of biofertilizers is that it increases soil electrical conductivity which may cause lower yield, the usage of recovered ammonium sulphate can still produce a yield amount to 10 % due to its contribution in decreasing the soil pH and also elevating nitrogen level in leaves. Some examples of ammonium sulphate application is that it granted a more efficient growth and development of highbush blueberry plant in the initial five years as compared to other nitrogen fertilizers sources according to Ye et al. (2022). They also stated that the presence of sulphur in the ammonium sulphate fertilizers is important to the soil which very often is lacked in other types of fertilizers.

Biochar

According to Pathy, Ray and Paramasivan (2021), nitrogen being recovered from the wastewater can be adsorbed by biochar where it is being applied in the agriculture to increase crop yield as a slow-releasing fertilizer as a result of the aromatic group attached on it. Besides, the usage of biochar contributes to the sequestering of carbon in the soil where biochar is being applied, aids in the reduction of nitrogen loss, as well as enhancing productivity of soil according to Ye et al. (2022). On top of this, a research also showed that the application of nitrogenous biochar also brings helps improved soil pH level and its cation exchange capacity, and polymer matrix biochar composite was also being utilised for the extraction of nitrogen from urine sources and then being applied to cotton plant (Ye et al., 2022). The addition of nitrogen-loaded biochar to the cotton plant have granted positive effects such as increased nitrogen usage efficiency and decreased leaching of nitrogen. A study conducted by Xu et al. cited by Ye et al. (2022) also showed similar result where application of magnesium modified biochar loaded with ammonium from urine to ryegrass and maize plants have increase in their plant height.

Microalgae-based Fertilizer

The usage of microalgae-based fertilizers can be used in many application, such as for growth of plants, solubilizing phosphate and nitrogen, providing vitamins needed for plant healthy sustenance, and providing many other nutrients. In the agriculture industry, low doses of microalgae-based fertilizers sufficed to improve the overall physiological processes in a plant. According to Ronga et al. (2019), this type of fertilizers was also able to prolong the productivity, shelf-life, quality of crops and plants as well as increasing crop tolerance towards abiotic stresses and nutrient adsorption. According to a study conducted by Coppens et al. cited by Ye et al. (2022), the quality of fruit was enhanced as microalgae-based fertilizers was being applied in tomato cultivars, such that it contained 44 and 70 % more carotenoids treated by organic and inorganic fertilizer respectively.

4.3.2 Microalgae Biomass

The production of microalgae is usually conducted through the photoautotrophic cultivation in which biomass can be obtained in a large-scale through sunlight as a renewable source of energy. According to Ye et al. (2022), current studies showed that MPBRs were being examined for its nitrogen recovery efficiency and its production of biomass from various types of wastewater, such as from agricultural land, wastewater slurry as well as commonly produced sewage. There are many functionalities of microalgae biomass such as its usage as raw material in feed stream as well as in the food industry as it provides valuable health and nutritional content (Morais Junior, et al., 2020). Another study from Vermaas cited by Ye et al. (2022) indicated that microalgae biomass was also used as supplements for feed such as for poultry feed in which it provided conventional proteins to improve the colour of egg yolk and skin of broiler. The focus of future research could be to consider the unique biochemical properties of various species of microalgae before utilising them as supplements in animal diets.

Besides, microalgae biomass/extracts have a list of functions such as to reduce cholesterol and offers many health benefits, used as medication against bacterial infections in fish or shrimp, in producing cosmetic products such as anti-aging creams as well as sun and hair protection products. Due to its potential of antimicrobial which helps anti-aging effects, they have contributed to a wide range of available cosmetic products we use today (Mourelle, Gómez and Legido, 2017). Apart from this, microalgae biomass aids in the sequestration of carbon dioxide in the ethanol synthesis factory due to its ability to capture carbon dioxide gas, which contributed in cost saving for chemical removal processes according to Rosenberg, et al. cited by Ye et al. (2022). Lastly, microalgae biomass was also being explored as a material for the production of biofuels, biofertilizers, bioplastics and neutraceutics according to various researchers (Ye et al., 2022).

4.3.3 Bioenergy

Bioenergy is a renewable source of energy that is highly in demand due to the challenges posed by the conventional use of non-renewable fossil fuels. Besides, the combustion of fossil fuels also produces undesirable GHGs such as carbon dioxide which has brought negative consequences to the global climate. Therefore, bioenergy was sought out where biological or natural resources such as flora, fauna and their by-products could be used as raw materials. From the nitrogen recovery process in the wastewater industry, the generation of bioenergy can be derived from microalge biomass and through BMISs.

According to Raheem et al. (2018), the synthesis of bioethanol and biodiesel were made possible from the conversion of microalgae biomass. The simple structure of microalge biomass have further aided in the effective conversion of energy five times more than most of the terrestrial plants (Yap et al., 2021). Besides, it has elevated growth rates compared to traditional food crops in which they are 5 to 10 times faster. The advantage of microalgae biomass-derived energy is that modifications of existing fuel engines is not required as these biofuels are compatible in nature. In the context of lipid productivity, microalgae biomass is 15 to 300 folds larger than the common oil crop, rendering it feasible to be used as vegetable oils (Ye et al., 2022). The oil yield is also significantly higher than that of other crops such as rapeseed, soybean and palm oil which are commonly used (Paniagua-michel, Olmos-soto and Morales-guerrero, 2015). In spite of these advantages, the highlight is that the productivity of microalgae biomass is 10 to 20 fold more than other biofuel crops (Ye et al., 2022). Nevertheless, the harvesting and managening of large volumes and high production of biomas remained the challenges for application as biofuel. Secondly, the proposed BMISs also functioned to produce bioenergy in forms such as biogas generated from methanogens' activity in anaerobic conditions as well as electricity from BES-based systems.

4.4 Feasibility and Challenges of Nitrogen Recovery Pathways

Figure 4.10 below illustrated the nitrogen recovery pathway that can be demonstrated in the wastewater industry (Ye et al., 2018). In summary, the beginning of the cycle starts when nitrogenous compound in the form of ammonium ions are excreted by human and animals into the municipal wastewater. After that, these ammonium ions in the wastewater are required to undergo pre-concentration process where the BMISs technologies are in place to enrich the ammonium concentration before being recovered by means of ammonium precipitation or stripping and adsorption technique. The next process is that the recovered ammonium ions in the form of struvite or ammonium salts are being produced, where it can be used as ammonium-based fertilizers for agricultural purposes to substitute the conventional synthetic fertilizers for the growing of food and crops. Lastly, the concept of bio-circular economy is achieved when human and animals ingested the produced crops which eventually excrete ammonium ions in the wastewater, or through the leaching of ammonium-based fertilizers from the agricultural land into the wastewater. Thus, the circular bio-economy of nitrogen management in the wastewater industry can be achieved.

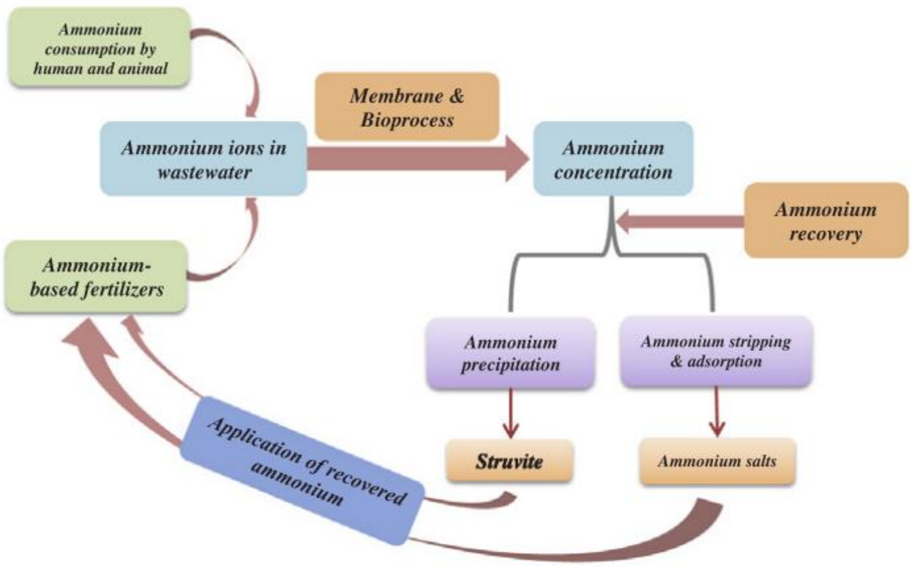

Figure 4.10: Graphical Illustration of Overall Nitrogen Recovery in Municipal Wastewater Industry (Ye et al., 2018)

With that being said, though the above-mentioned recovery of nitrogen in the municipal wastewater industry can be theoretically feasible and achievable given the advantages it offered, nevertheless it still posed many challenges and therefore in-depth feasibility study was very much needed. In the following section, several challenges on the proposed nitrogen recovery pathway were expounded in terms of the economic and technical prespective to determine its feasibility in the large-scale application in the municipal wastewater industry.

4.4.1 Economic Analysis

To embrace the concept of nitrogen recovery, the cost-effectivenes of the proposed pathway needs to be thoroughly analysed as it acted as one of the main driving incentives for its development on a commercial scale. Therefore, the proposed nitrogen recovery technologies needs to be further analysed in terms of its rate of energy consumption, efficiency of reaction and its operating parameters to evaluate its applicability in the municipal wastewater industry. In the case of BMISs, it is important to ensure that it is economical for its proper execution in comparison to the Haber-Bosch process to ensure the energy consumption per unit of nitrogen recovered in the BMISs is lower than that of Haber-Bosch process. According to Ye et al. (2022), they found out that although there were ample studies and researches on the economic analysis of BMISs in the recovery of nitrogen, most of the datas obtained were only limited to lab-scale reactors for short-term experiments, which leaves the commercial execution of the BMISs processes questionable. This was because several factors such as its durability and sustainability in the long run, possible high cost of component material that included membrane and electrode, and only pilot-scale operations being conducted act as the limitations to fully comprehend the economical feasibility in a commercial scale. Hence, the trustworthiness of the current techno-economic analysis was still in doubt for most of the industrial players to venture into the nitrogen recovery pathway in the municipal wastewater industry.

Apart from the lack of solid evidence of executing in a commercial scale, Ye et al. (2018) highlighted the energy and cost consumption associated with the nitrogen recovery pathways. The first process highlighted was struvite precipitation, that though posed advantages such as ability to recover phosphate simultaneously and being a safe and effective slow release fertilizer suitable for direct application in agricultural land, it still has its disadvantages of requiring a huge amount of extra chemicals such as alkali sources for pH elevation and magnesium as part of the building block of struvite precipitation.

The next process highlighted was stripping coupled with adsorption where this pathway does not depend on the concentration of wastewater feed, and therefore indicated its suitability for treating a wide-range of wastewater sources. Nevertheless, the challenges to this process were the selection of acid solutions for the adsorption process in ammonia that could affect the economic feasibility and the ammonia stripping process is only economical viable for the treatment of wastewater with 1000 to 1500 mg N/L total ammonium nitrogen. Besides, the cost incurred to this process is dependent on the aeration and the additional chemicals needed such as alkali to maintain the required alkaline pH for the formation of gaseous ammonia and acid solutions for the ammonia adsorption to form ammonium salt. Therefore, these additional elements may increase the overall costs because of the inclusion of purchase cost of raw material and energy consumption from blowers and heaters for aeration purposes (Ye et al., 2018).

Lastly, Ye et al. (2018) highlighted the economical feasibility of BES system where it is considered advantageous as there was no need for pH elevation for the recovery of ammonium ions in the vapor phase as compared to previous methods. The energy consumption used in this process included aeration, ammonia adsorption by sulphuric acid, additional power consumption in the case of MEC, and generation of energy by MFC. The analysis of the energy balance for the recovery of ammonium ions by several researches according to Ye et al. (2018) were being tabulated in Table 4.4. As shown in Table 4.4, MFC demonstrated a positive net energy yield while the conventional ammonia stripping technologies was recorded with the highest energy consumption amongst MFC and MEC. MEC had recorded a higher energy consumption than MFC as it required an external power source to function. Besides, ammonia stripping produced the highest net energy yield as it does not produce energy in the process as compared to MFC.

Table 4.4: Energy Balance Comparison Between MFC, MEC and Conventional Ammonia Stripping Process (Ye et al., 2018)

Parameters	MFC	MEC	Ammonia Stripping
Recovery rate of Ammonia (g N/(d.m^2)	3.29	7.59	-
Energy Consumption (kJ/g N)	10.93	18.36	26.3
Net energy yield (kJ/g N)	3.46	-18.36	-32.5

In terms of the quality of the recovered ammonium from the wastewater, it is deemed economical feasible as it is expected to promise great contributions to the agricultural industry. The characteristics of struvite render it a good choice as fertilizers, such as its low leaching and rate of release of nutrients during the season where plants grow as compared to conventional fused super-phosphate, readily soluble and bioavailable at broader pH range and soil types (Ye et al., 2018). Besides, the recovered ammonium salts and liquid ammonia has the great potential to be used directly as agricultural fertilizers. Amongst these, ammonium sulphate could also be utilized in the nitrogen polymer manufacturing industry and in the food production process. Lastly, pure ammonium sulphate crystals produced from the adsorbed ammonia during pre-saturation of sulphuric acid solution can also be used as laboratory chemicals and fertilizers, thus further adding to its economic viability.

4.4.2 Technical Analysis

As for BMISs, due to the involvement of microbal activity, there is a high possibility for more problem occurrences, including undesired consumption of substrate by mircrobes due to competing metabolic processes, growth of unwanted biomass as well as non-complete biodegradation of substrates negative affected the amount of recoverable nitrogen (Ye et al., 2022). Another common problem for BMISs were the potential for membrane fouling which will contaminate the permeate and causes deterioration on the efficiency of membrane over the long haul, rendering it unsustainable. Next, the scaling up of membrane in the large-scale setting still posed a challenge as only lab-scale or pilot plant study was conducted to date. Lastly, the cost of membranes needs to be considered when applying in a large-scale municipal wastewater industry, especially to keep the cost of membrane synthesis economical whilst having its properties improved.

Amongst most of the BMISs technologies, Yang et al. (2021) claimed that the OMBR integrated system has unique advantages such as its reduced energy input and potential for membrane fouling. Despite its advantages, the OMBR integrated system was still hindered for its commercial execution due to the problem posed from the recovery of draw solute from the FO process. The conventional method of draw solute recovery through ED, MD and RO requires high consumption of energy, thus increasing the overall cost and rendering it economically unfeasible.

Another BMISs is the BES where it was capable of converting chemical energy of waste in the wastewater into energy and chemicals to curb the problem of high energy requirement and pollution related issues. The BES has the potential for commercial application due to its ability to rapidly degrade organic wastes in the anode chamber, thus rendering this process to fulfil the circular bioeconomy model. However, challenges still existed in the BES, such as the need for close and effective monitoring strategies of the anode chamber. This was because the generation of electricity of the BES will be negetively affected by the anode deactivation due to undesired side reactions such as organic wastes accumulation on anode surface. Besides, the selection of suitable electrodes and membrane materials was needed for the technical feasibility of the process as the efficiency of the BES will be affected. However, there existed a dilemma of choosing a suitable electrode and the cost incurred. For example, Wang et al. (2017) stated the traditional use of Pt-supported cathode electrodes in the BES acts as a hindrance for commercialisation due to the high cost it incurred, despite the fact that this type of cathode aided in the oxidation-reduction potential. The same analogy could be applied to membranes, where its quality could potentially be affected by the condition of wastewater, contamination and accumulation of mircobes as well as the leakage of gas between the anode and cathode chambers (Ye et al., 2022). Besides, the MPBR integrated systems have its challenges derived from the efficiency of photosynthetic activity, amount of dissolved gas, fluctuation of pH solution, mixing, temperature, nutrient solubility and irradiance, of which the greatest challenges lies at the sunlight irradiance (Ye et al., 2022).

For CANDO process, despite the potential for the recovery of nitrous oxide for useful application, it appeared that such practice of recovery was debatable in terms of its environmental sustainability and economic feasibility due to the challenges posed from the process of harvesting, nitrous oxide purification and the emission of residual dissolved nitrous oxide according to Zhang and Liu (2021a). Such challenges were expected to complicate the process configuration, leading to high operational cost and generation of insignificant recoverable energy compared to the total energy consumption in the plant.

As for the recovery of ammonium through adsorbents, it appeared to be technically unfeasible in the long haul as regeneration of adsorbents causes the reduction of ammonium adsorption capacity. Besides, Zhang and Liu (2021a) also added that using low adsorption capacity of adsorbents rendered it not environmentally friendly and economically inviable. Moving on to the use of microalgae and cyanobacteria, the concern of requiring a vast land for ponds installation served as a limitation. Besides, the varying characteristics of the wastewater possibly hindered the reliance on microalgae and cyanobacteria for nitrogen recovery due to varying nutrients, toxic substances and environmental factors such as pH, light, temperature as well as oxygen and carbon dioxide levels (Rahimi, Modin and Mijakovic, 2020a).

4.5 Future Perspective of Nitrogen Recovery Pathway in Municipal Wastewater Industry

In the following section, the future prespectives of each proposed nitrogen recovery pathways in the municipal wastewater industry were reviewed in the order of BMISs, CDI, CANDO process, ammonium recovery through adsorbents, stripping and struvite precipitation, and lastly the usage of microalgae and cyanobacteria. Besides, an overall perspectives of the future of nitrogen recovery pathways were highlighted at the end of this section.

4.5.1 Future Perspectives of Nitrogen Recovery Technologies

BMISs

According to Ye et al. (2021), the BMISs should consider the following in future research:

1. The potential for membrane fouling was still a major hindrance for nitrogen recovery as the filtration efficiency can be severely affected and causing the reduction of membrane lifetime. Possible research direction suggested were to optimize operating parameters, decreasing mixed liquor salinity and conduct cleaning through chemical or mechanical means. Besides, other hindrances such as the high cost, energy and chemicals requirements to prevent membrane fouling were the limiting factors. Hence, comprehensive understanding on membrane fouling is deemed crucial for the development of BMISs, such as on the aspects of membrane properties, sludge characteristics and the operation conditions.
2. The final product of the nitrogen recovery process should be considered, such as analysing its actual market of the fertilizers products generated. From there, it was found possible to establish connection between local farmers and wastewater treatment plants for the supply of fertilizers. The various end products generated from the nitrogen recovery processes were being shown in Figure 4.11.

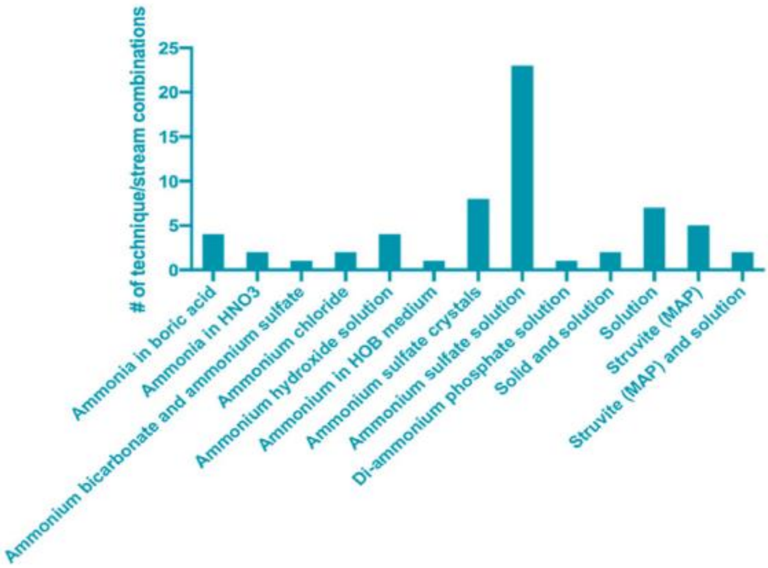

Figure 4.11: Final Products Generated from Nitrogen Recovery Pathways (Beckinghausen, et al., 2020)

3. Parameters of microbial activity needed more attention due to its uncertainties. These includes the consumption of substrates in among other metabolic proceses involved, undesirable growth of biomass and incomplete biodegradation of substrates that decreases the rate of performance. Therefore, economic analysis should be conducted to determine the most technical and economical feasible nitrogen recovery pathway. Besides, avoidance and minimization of the occurrence of nitrogen removal techniques should be conducted to maximise the execution of nitrogen recovery techniques.
4. Full-scale implementation of BMISs to treat real wastewater should be prioritised for future research as there were limited confined only to lab-scale.
5. Optimization studies were needed to be conducted such as the effects of assembling communities, the types of material of electrodes, varying nutrients concentration, internal resistance and configuration of reactor. In addition, other parameters such as enrichment of ammonium ions and transport for membrane processes, process upsets due to the presence of contaminants and competing ions needed to be focused for future research.

6. Integration of nitrogen recovering technologies were proposed to minimise energy consumption while maximising amount of recoverable nitrogen. Amongst other BMISs, the BES and FO-based integration system should be prioritised due to its promising advantages of low energy consumption and reduced membrane fouling potential.
7. Research be conducted on ammonium and energy recovery in BES as the existing process promises high ammonium recovery with low energy recovery and vice versa. Therefore, future research on developing the BES configuration is needed to achieve both energy and ammonium recovery in an optimized manner.

CDI

According to Fang et al. (2018), the FCDI technology can be considered a promising pathway for the recovery of ammonia from low concentration of municipal wastewater. In addition, it can be applied for an extensive range of wastewater influent qualities. Therefore, future research on the optimization of new electrode materials, as well as forming a theroretical framework were needed for the successful implementation of the recovering of ammonium ions from low influent concentration.

CANDO Process

According to Zhang and Liu (2021b), this process required a more thorough assessment on the nitrous oxide recovery pathways from the municipal wastewater, as well as on the aspects of anaerobic digestion of liquor.

Ammonium Recovery through Adsorbents

According to Zhang and Liu (2021b), the usage of low-ammonium adsorption capacity of the adsorbents acted as the limiting factor for its long term sustainability. Therefore, it is crucial to find alternative technology to replace the use of adsorbents without the need for its regeneration. There were two integrated technologies that were proposed by the authors, which consists of the: (1) integration of anaerobic fixed-film MBR-biochar adsorption with RO, and (2) anaerobic AnNFMBR-RO coupled with electrodialysis removal (EDR) and ozonation process. These two integrated systems were explored and concluded its potential for not limited to nutrients such as nitrogen, but also high-grade product water and energy.

Stripping and Struvite Precipitation

These methods were established technologies proven to be used for the generation of biofertlizers. Hence, its integration with other technologies mentioned previously can be researched for optimal nitrogen recovery. However, research efforts can be directed to find inexpensive magnesium sources for struvite precipitation as large amount of magnesium is needed (Rahimi, Modin and Mijakovic, 2020a).

Use of Microalage and Cyanobacteria

According to Rahimi, Modin and Mijakovic (2020b), the future perspective of this technology on the reliance of microalgae and cyanobacteria was to conduct investigation on the possible negative environmental impacts of land use change and the changes in carbon content in soil. Besides, research efforts on the optimization of characteristics of wastewater and the environmental parameters should be conducted as the growth of these microorganisms will be affected leading to inefficient treatment of wastewater.

4.5.2 Overall Future Perspetives

Apart from the specific future perspectives of each nitrogen recovery pathways, it was important to highlight the general future perspective according to several researchers as follows (Beckinghausen et al., 2020b; Ye et al., 2018):

1. Introduction of governmental policies and regulations if economic incentives from nitrogen recovery in municipal wastewater industry was insufficient for industrial players.
2. More research efforts on proper treatment of wastewater after the nitrogen recovery process as nitrogen residues may still be present in treated wastewater above the discharge limit of nitrogen in wastewater.

3. Life cycle assessment (LCA) on the sustainability of ammonium recovery should be conducted, which includes the aspects of economics, environment and society.
4. The recovery of phosphate element should be considered when ammonium was being recovered as phosphate can supplement the usage of fertilizers and lower the risk of euthrophication.
5. Research efforts on the recovery of ammonium to not only limited to wastewater, but also wastewater sludge as 3 to 4 % of nitrogen can be found in the dry weight of sludge (Ye et al., 2018).
6. Intense research on integration of different nitrogen recovery technologies should be conducted for maximization of ammonium recovery while minimizing energy consumption.
7. Engagement of researchers with stakeholders from the agricultural sector or chemical sector was needed. Thus, inputs on the type of bioproducts (fertilizers, chemicals, etc.) that will be needed for the successful implementation of biocircular economy in the municipal wastewater industry can be known.
8. Relationship between local farmers scientists should be cultivated to make adjustment on the current nitrogen recovery technologies to meet the local soil and field demands.
9. Full-scale economic and technical analysis should be conducted for pathways under consideration, such as BMISs, FCDI and struvite and chemical precipitation. This includes cost-and-benefit analysis for potential investment in this processes in the future.
10. The local market of the bioproducts from the recovery processes needs to be considered and investigated.
11. The perception for the adoption of bio-circular economy concept in the municipal wastewater industry by stakeholders and consumers should be considered.

In short, all of the discussed technologies, except for CANDO process, should be analysed thoroughly to ensure its economic and technical feasibility on the long haul. Despite the shortcomings of each proposed technologies presented, it can be deduced that the integration of several types of technologies would be deemed helpful for the successful implementation of nitrogen recovery at a municipal wastewater industry. Since not all regions possesses the same maturity of nitrogen recovery technologies due to varying resources availability, it is made possible for industrial players in this field to gather as much pilot/full-scale data available online as possible on the integration of these technologies to achieve a cohesive nitrogen recovery pathway in their own specific wastewater treatment plant. Amongst these, the BMISs, FCDI, and struvite precipitation and stripping should be considered first for its integration as they have granted solid evidence of its potential for nitrogen recovery in a pilot-scale setting. Since there are many available altrnatives within the BMISs, BES-FO can be given priority for its full-scale implementation of treating real municipal wastewater as previously mentioned by Ye et al. (2021).

Therefore, the future prospect for the municipal wastewater industry lies on the recovery of nutrients such as nitrogen as well as others nutrients such as phosphorus and magnesium. The main reason nitrogen was being emphasised in this project was due to its devastation towards the environment (acidification of water bodies, euthrophication, etc.) and its potential to be recycled to be mainly used for biofertilizers to meet increasing food demand as mentioned in Chapter 2. To embrace the paradigm shift from linear economy to the prospective circular bioeconomy model of the wastewater industry, both the government and industrial players play a crucial role in ensuring the eventual successful implementation. Although most, if not all, of the proposed nitrogen recovery technologies proposed earlier were confined in lab or pilot-scale in treating the wastewater, investment towards research of the full-scale implementation of potential recovery pathways should be conducted as these pathways have demonstrated excellent nitrogen recovery rate in the lab or pilot-scale. Further research effort on the techno-ecnomical datas on these potential pathways could be conducted to convince the parties involved in revolutionising the wastewater industry as a resource hub of nutrient.

As important is it to consider the techno-economic feasibility, the social aspects needs to be considered, such as the relationship between local farmer scientists, researchers with stakeholders, and the perception of end-consumers on such implementation as previously mentioned. The long term sustainability of such practices needs to be evaluated by means of LCA or other similar methods to ensure all aspects were considered before its full-scale implementation.

CHAPTER 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

The current linear economy model of nitrogen management in the municipal wastewater industry ought to be replaced with the circular economy model due to the drawbacks that were currently presented. These drawbacks included the deterioration of environment through euthrophication and acidification of water bodies and the negative impacts it brought about to living organism. The current nitrogen management practices mainly revolved the BNR processes which includes nitrification-denitrification, aerobic denitrification, nitritation-denitritation, and aerobic ammonium oxidation (Annamox). Although the widely practiced BNR processes being utilized is the nitrification-denitrification, there were also other chemical and physical treatment methods for removing nitrogen from the municipal wastewater.

Since these valuable nitrogeneous compounds were not being utilized into its full potential but merely being discharged into water bodies and atmosphere, the circular bioeconomy concept was proposed to be implemented to produce useful products such as biofertilizers, biomass and bioenergy. Hence, a list of nitrogen recovery pathways were being investigated, in which all were conducted either in lab or pilot scale such as BMISs, CANDO process, FCDI, ammonium recovery through adsorbents, usage of microalgae and syanobacteria, and struvite precipitation and stripping process. Amongst these, the BMISs, FCDI and struvite precipitation and stripping technology can potentially be used to treat real municipal wastewater conducted in a full-scale municipal WWTP.

Although these pathways have demonstrated the potential for nitrogen recovery in municipal wastewater, there were still many feasibility uncertainties from the technical and economic aspects. The technical aspects mainly included the challenge of recovering nitrogeneous compounds from low-strength municipal wastewater that requires the preconcentration of ammonium compounds prior to its recovery, while the economic aspects present were mainly on maintaining the balance of maximizing ammonium recovery and minimizing energy expenditure and usage of additional chemicals that incurred higher costs. Several challenges in embracing the circular economy model in municipal wastewater industry also included the insistence of old thinking of embracing the linear economy model, low resources availability, only spot-light short-term results, and lack of enforcement of regulatory bodies.

5.2 Recommendations for Future Work

Thus, ample researches on the nitrogen recovery pathways and its future perspective were being investigated. Nonetheless, several constraints were in place in completing this project, such as the non-expertise of student undergoing the study and limitation of study/research time. Hence, several recommendation for future work to implement the biocircular economy model of nitrogen management in the municipal wastewater industry were proposed as follows:

1. Explore integration of feasible nitrogen recovery pathways (BMISs, FCDI, struvite precipitation and stripping coupled with adsorption),
2. Conduct further study on techno-economic on feasible nitrogen recovery pathways for long-term sustainability,
3. Research on maximization of ammonium recovery and minimization of energy expenditure and usage of chemicals in low strength municipal wastewater on feasible nitrogen recovery technologies,
4. Adoption of potential strategies or incentives by local authorities to improve the current policies or legislation for the implementation of nitrogen recovery pathways in municipal wastewater industry, and
5. Perform optimization studies on feasible nitrogen recovery pathways on pilot or large-scale in the treatment of real municipal wastewater.

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