Ali Saud

# SUSTAINABLE RECOVERY OF NITROGEN FROM SEWAGE SLUDGE

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Dissertation for the degree of Doctor of Science (Technology) to be presented with due permission for public examination and criticism in the Auditorium 1314 at Lappeenranta-Lahti University of Technology LUT, Lappeenranta, Finland on the 18th of December 2023, at noon.

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# Abstract

The growing population and urbanization have increased the need for wastewater treatment alongside concerns about the disposal of the resulting sewage sludge. On the other hand, food scarcity and the reduction of fossil fuel-based fertilizers have increased the need for the recovery of nutrients from sewage sludge. This dissertation presents a comprehensive investigation into the feasibility of resource recovery from sewage sludge. Mostly focusing on nitrogen recovery, also taking into account phosphorus (P) and energy. The challenge posed by potential hazardous materials in sludge such as pathogens, pharmaceutical residues and microplastic has led to the selection of thermal treatments of sewage sludge. In thermal treatment, pyrolysis and combustion are used as the primary treatments while adsorption and stripping and scrubbing are often selected as post treatments to recover nitrogen from reject water, condensate, and the drying fumes resulting from the thermal drying of sewage sludge. This study examines the economic and environmental implications of these approaches and provides valuable insights for sustainable waste management practices.

The aim and objectives of the dissertation are achieved by process modelling, carrying out mass and energy calculations and by using a life cycle assessment (LCA) methodology. The research objectives of this dissertation are i) the comparative technical possibilities for recovering ammonia ($NH_3$) from exhaust fumes of thermal drying, ii) the optimal integration approach for maximizing the $NH_3$ recovery in wastewater treatment plants, sewage sludge treatment, and exhaust gases treatment processes, and iii) the assessment of environmental impacts which are associated with different $NH_3$ recovery technologies, and strategies which are the most effectively minimize their environmental footprint.

In the light of the aims and objectives, four studies were conducted to address them. In Publication I, $NH_3$ recovery from drying fumes during the thermal drying of sewage sludge is explored using packed bed scrubbers. The process is modelled for different ammonia concentrations (75 and 100 ppm) and a drying fumes inlet flow rate of 1,000 $m^3$/h. The optimized parameters for scale-up are determined for 7,700 t/a sewage sludge treatment in Lappeenranta. The results indicate that a single scrubber with an inlet gas flow rate of 24,000 $m^3$/h, 75 ppm ammonia concentration, 1.5 liquid to gas ratio, 100 °C liquid acid temperature, and pH of 3 achieves an impressive efficiency of more than 99%, reducing the outlet stream ammonia to 0.2 ppm. However, the initial economic analysis

suggests that producing commercial-grade ammonium sulfate (AS) fertilizer from the recovered ammonia may be economically challenging.

Publication II focuses on heat and nutrient recovery through pyrolysis and combustion with gas scrubbing. Mass and energy balance calculations are made for a wastewater treatment plant (WWTP) with a capacity of 65,000 t/a of mechanically dewatered digestate (29% total solids). The nitrogen and phosphorus recovery from the digestate streams is evaluated, and the scenarios show potential for generating 3,500 t/a of ammonium sulfate (AS) fertilizer, along with producing 120 GWh/a of district heat and 9,700 t/a of ash with 500 t/a phosphorus in the combustion scenario, and 12,000 t/a of biochar with 500 t/a of phosphorus in the pyrolysis scenario. The nitrogen recovery requires additional electricity using a stripper and scrubber, and economic estimates reveal yearly investment expenses of 2–4 M €/a, as well as 2–3 M €/a for combustion and pyrolysis, respectively, with projected product revenues of 3–5 M €/a and 3–3.5 M €/a.

In Publication III, an LCA is used to evaluate composting, combustion, and pyrolysis options for dewatered sewage sludge digestate. Sewage sludge digestate combustion and composting outperformed pyrolysis in most effect categories. The pyrolysis of sewage sludge is currently under research, so additional data is needed to judge its performance. Publication IV used an LCA to investigate the environmental impact of nitrogen recovery for fertilizer from sewage sludge treatment in a municipal wastewater treatment plant (WWTP). Nitrogen was recovered from ammonium-rich reject streams from mechanical dewatering and thermal drying of anaerobically digested sewage sludge using air stripping or a pyrolysis-derived biochar adsorbent. The results varied by scenario and impact category. The global warming potential of nitrogen recovery based on biochar was the lowest, with net negative greenhouse gas (GHG) emissions of 22.5 kt $CO_2$ eq./FU. Total GHG emissions were 2 kt $CO_2$ eq./FU when $NH_3$ was captured by air stripping and were 0.2 kt $CO_2$ eq./FU in the base case without nitrogen recovery. The study also analyzed the potential environmental and health benefits of wastewater systems that incorporate integrated resource recovery.

This study promotes multifunctional wastewater systems with integrated resource recovery for environmental and health benefits. In conclusion, this dissertation contributes significant insights into the potential and challenges of resource recovery from sewage sludge, promoting sustainable and environmentally conscious wastewater treatment practices. It also highlights the utility of recovered products such as ammonium sulfate, biochar, and district heat. Biochar has demonstrated its significance as a valuable product to utilize as an adsorbent and to capture carbon. The integration of different nitrogen recovery technologies can enhance resource efficiency, reduce environmental impacts, and improve the circular economy in sewage sludge treatment with the development of sustainable solutions to handle sewage sludge volumes safely.

Keywords: Sewage sludge, nitrogen recovery, nutrient recovery, process modelling, life cycle assessment, LCA, waste to energy, sewage sludge utilization, waste recovery

# Acknowledgements

*To my parents, my wife, my family and my Minha*

# Contents

**Abstract**

**Acknowledgements**

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# List of publications

This dissertation is based on the following papers. The rights have been granted by publishers to include the papers in dissertation.

I. Saud, A., Havukainen, J., Mänttäri, M., & Horttanainen, M. (2020). Evaluation and techno-economic analysis of packed bed scrubber for ammonia recovery from drying fumes produced during the thermal drying of sewage sludge. E3S Web of Conferences, 191, 03001. https://doi.org/10.1051/E3SCONF/202019103001

II. Saud, A., Havukainen, J., Peltola, P., & Horttanainen, M. (2021). Integrating Pyrolysis or Combustion with Scrubbing to Maximize the Nutrient and Energy Recovery from Municipal Sewage Sludge. Recycling 2021, Vol. 6, Page 52, 6(3), 52. https://doi.org/10.3390/RECYCLING6030052

III. Havukainen, J., Saud, A., Astrup, T. F., Peltola, P., & Horttanainen, M. (2022). Environmental performance of dewatered sewage sludge digestate utilization based on life cycle assessment. Waste Management, 137, 210–221. https://doi.org/10.1016/J.WASMAN.2021.11.005

IV. Saud, A., Havukainen, J., Peltola, P., & Horttanainen, M. (2023). Environmental Performance of Nitrogen Recovery from Reject Water of Sewage Sludge Treatment Based on Life Cycle Assessment. Recycling 2023, Vol. 8, Page 43, 8(2), 43. https://doi.org/10.3390/RECYCLING8020043

## Author's contribution

Ali Saud is the principal author and investigator in Papers I–II and IV. In Paper III, Dr. Jouni Havukainen was the corresponding author and principal investigator and Ali Saud contributed to writing, collecting, and analysing data and making calculations for stripping and scrubbing.

# Nomenclature

**Symbol**

| | |
|---|---|
| % | percent |
| $°C$ | Celsius |
| $A$ | area |
| eq. | equivalent |
| wt% | weight percent |
| $\pi$ | (pi) usually reserved for mathematical value $\pi = 3.14159...$ |
| $\rho$ | (rho) density |

**Abbreviations**

| | |
|---|---|
| AS | Ammonium sulfate |
| ENRTL | Electrolyte non-random two liquid |
| FU | Functional unit |
| GHG | Greenhouse gas |
| HETP | Height equivalent to the theoretical plate |
| HSY | Helsinki region environmental services authority |
| L/G | Liquid-to-gas ratio |
| $l_{25}$ | Heat of vaporization of water |
| LCA | Life cycle assessment |
| LCI | Life cycle inventory |
| LCIA | Life cycle impact assessment |
| LHVar | Lower heating value as-received basis |
| $LHV_{dry}$ | Lower heating value for total solids dry basis |
| max | Maximum |
| min | Minimum |
| NRTL | Non-random two liquid |
| tot | Total |
| WWTP | Wastewater treatment plant |

**Chemical compounds**

| | |
|---|---|
| $(NH_4)_2SO_4$ | Ammonium sulfate |
| $CO_2$ | Carbon dioxide |
| K | Potassium |
| N | Nitrogen |
| $N_2$ | Nitrogen gas |
| $PO_4^{3-}$ | Phosphate ion |
| $NH_3$ | Ammonia |
| $NH_4^+$ | Ammonium |

| | |
|---|---|
| NO | Nitric oxide |
| NO$_3$–N | Nitrate nitrogen |
| P | Phosphorus |

# 1 Introduction

## 1.1 Background

In the presence of wastewater, health problems for humans and environmental risks, basic engineering has attempted to eliminate contaminants from wastewater treatment plants (WWTPs) to deliver safe effluent (Taddeo et al., 2018). However, water treatment requires substantial energy and resources, including land and infrastructure. Because of rising greenhouse gas (GHG) concentrations and human population growth, proper organic waste management has become an enormous logistics, environmental, and economic problem. The numerous international, national, and regional environmental control measures put in place to prevent or reduce the dumping of these residues in landfills indicate the seriousness of the matter (Vaneeckhaute et al., 2014). In addition, the projected increase in the global population to ten billion people by 2050 (UN,2022) will significantly strain food supplies. P and N are critical nutrients for plant growth. Therefore, increased food production will increase N and P requirements (van der Hoek et al., 2018).

N is an essential ingredient for all living things. Because fixed N sources are insufficient to meet an expanding population's demands, atmospheric nitrogen must be transformed into reactive nitrogen in the form of ammonia ($NH_3$) or nitrate ($NO_3$) (Erisman et al., 2015). N is abundant in the atmosphere as nitrogen gas ($N_2$), but most plants cannot use it due to the nonreactivity of molecular nitrogen. Due to biological N fixing, some plants and vegetables can absorb $N_2$ as ammonium ($NH_4^+$) and nitric oxide (NO). However, this process is insufficient to meet the $NH_3$ needs of all plants and crops. As a result, the Haber Bosch process has been modified to manufacture N fertilizers to meet such demands (Ye et al., 2018). The Haber Bosch process converts 120 million tons of atmospheric nitrogen into reactive nitrogen for fertilizer manufacture yearly. The Haber-Bosch method was developed to generate low-cost fertilizers. However, the high temperature and pressure necessary for reactions consume significant energy: 1kg of liquid ammonia consumes 42 MJ of energy and releases 1.9 kg of $CO_2$ (Razon, 2014). A considerable share of natural gas (50%), oil (31%), and coal (19%) is also used for $NH_3$ production (Smith et al., 2020). This reactive N, however, eventually returns to the ecosystem as a substantial pollutant (Rockström et al., 2009). In addition, food production has increased due to Haber Bosch N fertilizers, while nitrogen is disposed of in the form of urea and $NH_4^+$ due to human defecation and ends up in sewage (Paredes et al., 2007). As a result, the high flow of these nutrients causes eutrophication. (Kahiluoto et al., 2014). Another cause of eutrophication is the overuse of N fertilizers in fields, which causes additional nutrients to be flushed into the water system.

$NH_3$ is an integral component of the global N cycle and serves an essential purpose. Over 40% of the world's current food output would not be possible without the manufacturing of synthetic $NH_3$ to produce N fertilizers (Pfromm, 2017). However, due to high N intake and inefficiencies in the food production, processing, and consumption chain, a large

amount of N fertilizer used for food production is lost to the environment in various ways, including $NH_3$ emissions into the atmosphere. The release of $NH_3$ into the atmosphere harms human health, the environment, and the climate. The reaction of $NH_3$ with the acidic compounds in air such as sulfur dioxide ($SO_2$) and nitrogen oxide ($NO_x$), results in the formation of $NH_4^+$ aerosols which affect air quality (Wang et al., 2015).

Furthermore, the eventual return of the released $NH_3$ to the earth's surface via N deposition could result in soil acidification, water eutrophication, and species extinction (Ti et al., 2018). The necessity of N, P, and potassium (K) fertilizers for intensive agriculture raises concerns regarding long-term availability and production costs. This is notably valid for P and K, predominantly produced from mineral reserves in specific geographic regions (Batstone et al., 2015). Given all these considerations, it is critical to consider new environmentally friendly nitrogen transport routes, which include improvements to the current (anthropogenic) nitrogen cycle, such as direct N recovery and recycling from wastewater (van der Hoek et al., 2018).

To solve pollution concerns posed by wastewater, wastewater treatment is a required technique. However, this process produces sewage sludge, which must be managed and disposed of properly. Sewage sludge is the residual or semi-solid formed while treating industrial or municipal wastewater. While wastewater treatment addresses one issue, the growing amount of sewage generated by urban communities demands appropriate regulations and innovative technologies for safely handling, disposal, and treatment of sewage and its remaining sludge (Mateo-Sagasta et al., 2015). As a result, controlling sewage sludge necessitates specific tactics and attention to avoid any adverse effects on the environment and public health. (Syed-Hassan et al., 2017).

Sludge management of WWTPs is strictly regulated, and typical techniques include utilization in agriculture, landfill disposal, anaerobic digestion, and incineration. The high-water content of sludge or sewage sludge digestate makes disposal and transportation expensive. Therefore, removing water from sewage sludge, is critical for cost savings. However, the presence of hydrophilic organic materials in the sludge makes dewatering difficult and inefficient. To improve sludge dewatering performance, wastewater treatment facilities often use a pre-treatment step, followed by mechanical dewatering techniques such as pressure filtration and centrifugation for deep dewatering (Zhang et al., 2022). The produced reject water requires treatment, and one option is to recycle it from the start of the process in the WWTP plant.

Sewage sludge can be directed to anaerobic digestion with or without dewatering. In case of the AD of sewage sludge that has not been dewatered, the resulting digestate can be dewatered to produce dewatered digestate and reject water. Reject water has a high N and P concentration, ranging from 750 to 1,500 mg N/L and up to 130 mg P/L. Despite accounting for only 2% of the total raw sewage flow, reject water considerably adds to the N and P load of the activated sludge tank, ranging from 10% to 30% and 10% to 80%, respectively (Guo et al., 2010). However, due to the changeable pH and high nutrient concentration, which might lead to eutrophication, this water cannot be disposed of in

surface waterways. While recycling reject water back into the wastewater treatment process is typical, this might lead to process instability and other difficulties (Solon, 2015).

The recycling of reject water into influent wastewater introduces a reactive N load accounting for 15% to 25% of the total nitrogen load of WWTP (Fux and Siegrist, 2004). This reject water is high in ammonium nitrogen, with up to 2.5 g $NH_4^+$-N/L possible. Therefore, it is necessary to treat the reject water to lower the reactive N load in the wastewater treatment facility. The recovery of ammonium nitrogen from reject water is an effective technique that not only treats the water but also eliminates the addition of reactive N loads to the wastewater treatment plant (Nancharaiah et al., 2016). As a result, it is recommended that reject water be treated from the initial influent to guarantee proper disposal.

However, after mechanical dewatering, sewage sludge or sewage sludge digestate still has a high moisture content of 73–84 wt.%, which could be reduced to an average of 5.6 wt.% using thermal drying processes to reduce the moisture content for further thermochemical treatments (Chan and Wang, 2016). Thermal drying is often done at temperatures of 85–115°C, which can increase sewage sludge dewatering properties and liberate bound water (Schnell et al., 2020). This method works well for lowering the moisture content of sewage sludge (Kelessidis and Stasinakis, 2012).

Recycling N from waste and wastewater streams with high nitrogen levels, such as municipal wastewater, manure, and sewage sludge, could be a promising solution to address excessive nitrogen fixation. It is important to consider the unexplored opportunities for N recovery from sewage sludge, which has a high nitrogen content. Although the direct application of sewage sludge or compost on land is not favoured due to environmental risks associated with heavy metals, pathogens, microplastics, and pharmaceuticals, many alternative methods can be used to safely and effectively to manage sewage sludge (Nkoa, 2014). For example, many developed countries have found thermal drying and thermal treatments of sewage sludge to be suitable disposal options due to the tightening legislation on landfilling and concerns about the direct land application of sewage sludge (Deviatkin et al., 2019).

The management of sewage sludge is a complex problem for wastewater treatment plants. (WWTPs). Thermal transformation techniques, such as incineration, pyrolysis, and gasification, can, on the other hand, convert troublesome sewage sludge into valuable energy and raw resources such as biochar, bio-oil, and gases.

Pyrolysis is a thermal chemical process that converts sewage sludge and biosolids into biochar, bio-oil, and non-condensable gas (Patel et al., 2020). This is a promising process for converting waste into valuable resources. The procedure entails heating organic substances at high temperatures in an inert atmosphere (Bridgwater et al., 1999). Bio-oil is utilized as a fuel or processed into specialty chemicals, while biochar could be used as a soil amendment, carbon-neutral fuel, adsorbent, or carbon black alternative. The shares

of the products their quality can be changed by changing pyrolysis process parameters such as reaction temperature, biomass heating rate, and vapor residence periods (Barry et al., 2019). Pyrolysis is a non-oxygenated endothermic process that reduces the volume of biosolids by up to 70%, stabilizes organic content, and wraps heavy metals in pyrolytic char (Fonts et al., 2012). Biochar from sewage sludge pyrolysis is pathogen-free and nutrient-dense (S. Singh et al., 2020). Pyrolysis sequesters carbon in biochar, and pyrolysis gas produces heat and electricity. Pyrolysis has numerous advantages, including safe sludge disposal, climate change mitigation, and the generation of valuable materials (Tripathi et al., 2016).

Pyrolyzing wet dewatered sewage sludge creates a steam-rich atmosphere in the reactor, increasing the water content in the liquid product, complicating purification, and promoting gas production due to in-situ steam reforming. Consequently, obtaining liquid fuel from wet sludge pyrolysis may not be suitable (Zhang et al., 2011). The main obstacles to applying pyrolysis are its economic viability and the complexity of processing equipment. However, these challenges could be overcome by increasing oil yields and effectively producing high-value products from pyrolysis chars, significantly improving its economic feasibility (Bridle and Skrypski-Mantele, 2004). Due to these issues, thermal drying is also required for pyrolysis as a pretreatment of sewage sludge (Domínguez et al., 2006).

In oxygen-limited settings, biochar made from biomass feedstocks has the potential to be an economic and effective adsorbent for removing $NH_4^+$. However, the pyrolysis temperature, oxygen-containing functional groups on the biochar surface, surface area, and pH of the solution are just a few of the variables that can affect the $NH_4^+$ adsorption capacity of biochar. Higher $NH_4^+$ adsorption capabilities may result from these modifications, enhancing biochar's overall structure and surface characteristics (Han et al., 2021).

Combustion is currently the most prevalent thermal treatment option for sludge energy valorization in Europe and other industrialized nations. Fluidized bed incinerators, for example, offer benefits including reduced sludge volume, thermal pathogen and odour annihilation, as well as renewable energy recovery. Sewage sludge is subjected to thermal decomposition through combustion at high temperatures ranging from 850 to 1,300 °C, generating heat and electricity for various uses (Schnell et al., 2020). In addition, the resulting bottom ash is extracted from the bottom of the furnace, while fly ash can be captured using bag filters or wet scrubbers (Syed-Hassan et al., 2017). The leftover ash necessitates additional costs for processing and disposal, and there are worries about hazardous emissions, among other disadvantages. Alternative methods of disposing of sludge include co-combusting sewage sludge with existing coal/lignite combustion units for power generation and using sewage sludge as a substitute fuel in cement manufacturing facilities with the integration of ash into the finished product. According to numerous research, these methods could reduce GHG emissions while increasing public acceptance and overall sustainability (Samolada and Zabaniotou, 2014).

Considering sewage sludge as a potential source of high-value nitrogen, as well as the embedded energy potential, and the reduction of sludge handling costs, disposal by thermal treatment is clearly advantageous. Furthermore, thermal treatment is suitable for destroying organic hazardous substances in the sludge, but N is usually lost in that context to the atmosphere as $N_2$ or $NO_x$. Our earlier article (Horttanainen et al., 2017) shows that at least part of the N is in a reactive form in the gases produced during thermal drying. Additionally, in reject water treatment N removal has been studied earlier (Guo et al., 2010), but its recovery has not been studied much. This dissertation bridges the gap from N removal to N recovery.

## 1.2    **Aim and objectives**

This research aims to evaluate $NH_3$ recovery from wastewater treatment plants and sewage sludge treatment and the integration of different recovery technologies and treatments. This research will also discuss why implementing various technologies to recover $NH_3$ is not feasible and what steps should be taken to make it feasible. The following research questions (RQs) were formulated to describe the aim of this dissertation and the articles addressing these research questions have been listed in Table 1.

**RQ1:** What are the comparative technical possibilities for recovering $NH_3$ from exhaust fumes of thermal drying?

**RQ2:** What is the optimal integration approach for maximizing $NH_3$ recovery in wastewater treatment plants, sewage sludge treatment, and exhaust gas treatment processes?

**RQ3:** What environmental impacts are associated with different $NH_3$ recovery technologies, and which strategies most effectively minimize their environmental footprint?

Table 1. Article publications in relation to research questions

|  | Publications | RQ1 | RQ2 | RQ3 |
|---|---|---|---|---|
| I | Techno-economic analysis of nitrogen recovery from exhaust fumes of the thermal dryer | ✓ |  |  |
| II | Integration of WWTP with pyrolysis and incineration to maximize the nitrogen recovery and waste-to-energy conversion |  | ✓ |  |
| III | Environmental impact assessment of composting, pyrolysis, and combustion technologies |  |  | ✓ |
| IV | Environmental impact assessment of nitrogen recovery from reject water of dewatering and condensate of thermal drying exhaust fumes |  |  | ✓ |

## 1.3    Scope and limitations of the current research

This dissertation's focus areas align with the research questions mentioned above, which define the dissertation's scope. The research is carried out with the help of data from different companies related to wastewater treatment and ammonia recovery.

The idea behind Publication I is the evaluation of stripping and scrubbing technologies for the recovery of N from the exhaust fumes of the thermal dryer. These technologies are selected based on their maturity and a literature review for their ability to recover low N concentrations in the gas phase. The idea behind Publication II are the mass and energy calculations needed to recover N from all possible escape routes of a WWTP with the integration of thermochemical treatments such as pyrolysis and combustion. Publication III and Publication IV both include environmental assessments of the studied technologies. However, the difference is that in Publication III, three technologies to treat sewage sludge are studied (combustion, pyrolysis and composting), whereas in Publication IV, N recovery from the reject water form dewatering and from condensate from thermal dryer fumes were studied, keeping the focus on two technologies, which are stripping and scrubbing together and adsorption on biochar.

The limitation of this research work is that it is location- and case-based. This dissertation focuses only on Finland, and the data used in the research was mainly obtained from Finland. However, the presented method can be used globally. The process modeling in Publication I is based on previous experimental data (Horttanainen et al., 2017) specifically for exhaust fumes from thermal drying. In Publication II, HSY Viikinmäki WWTP and Endev Oy data were used in the study, which provides information about Helsinki, its adjoining areas, and Rovaniemi. The LCA software used for quantifying the potential environmental impacts in Publication III was EASTECH, whereas in Publication IV the Gabi software program was used. ReCiPe 2016 v1.1 (midpoint hierarchical timeframe) was used in Publication III and IV because of its extensive environmental impact categories and endpoint features.

## 1.4    Research process and dissertation structure

Publication I is part of the SAWE—Safe Water for All—research platform of LUT University which aimed to develop water purification and treatment technologies to ensure the reutilization and availability of water for everyone.

Publications II, III and IV were accomplished under the Academy of Finland's funded project NITRO (decision number 315051).

**Section 1:** This section provides a concise overview of the research context, goals, scope, and constraints. It also outlines the research methodology and structure of the dissertation. Next, the background of the study topic is briefly introduced, followed by the objectives and research questions. Next, the scope of the study is clarified, and the research process

employed is described. Finally, the structure of the dissertation is outlined to provide a clear framework for the reader to follow.

**Section 2:** This section aims to present an in-depth overview of the research on techniques for recovering N from wastewater and its impact on the environment. Studies on nitrogen recovery using stripping and scrubbing technologies, thermal treatments of sewage sludge, and life cycle assessments (LCA) of these technologies will all be reviewed in this part. In addition, it also offers insights into the current state of research in this area and its applicability to the scope of this dissertation by summarizing the findings of these investigations.

**Section 3:** Materials and Methods - This section explains the materials and methods in Publications I, II, III, and IV and discusses the fundamentals of process modeling, calculations, and LCA methodology. It provides information on the data, process modeling, techno-economic analysis, the basis of mass and energy calculations, life cycle inventories, life cycle impact analyses, functional units, system boundaries, and research aims.

**Section 4:** Results and Discussions - This section offers the research's key findings and discusses them in more detail by summarizing the key findings in each publication and summarizing the results.

**Section 5:** Conclusions - In this section, the findings of the dissertation are outlined to form conclusions. It gives an overview of the key conclusions and contributions of the research work.

# 2 State of the art

## 2.1 Overview of the research

Nitrogen recovery from wastewater has achieved undeniable recognition in the last decade. The need to conserve natural resources and an efficient food supply for the growing global population are associated with each other.

The dissertation deals with the following core issues:

I. Nitrogen recovery from sewage sludge for utilization purposes.
II. Integration of N recovery from different material flows of sewage sludge treatment.
III. Evaluation of the economic feasibility of N recovery from sewage sludge.
IV. Evaluation of the environmental impacts of N recovery and integrated recovery from the treatment of sewage sludge.

The following chapters summarize the N recovery from municipal waste, manure, and sewage sludge to highlight the overview of the research work and target accomplished in nutrient recovery.

## 2.2 Nitrogen recovery from sewage sludge for utilization purposes

Part of the nitrogen can be recovered from the drying fumes of mechanically dewatered sludge before combustion (Deviatkin et al., 2018). It is also advantageous to recycle this $NH_3$ to meet the shortfall in the large amounts of fertilizers required to produce food (Sengupta et al., 2015).

Scrubbers are a widely developed technology that control odour from poultry and animal farms (Melse and Ogink, 2005). Two standard wet scrubbers are used for this purpose: packed bed scrubbers and spray scrubbers. The basic working principle of wet scrubbers is that a liquid, usually water or acid, is sprayed from the top while gas enters from the bottom. The packing material in a packed bed scrubber is soaked with liquid to prevent any dry spots where the gas can escape. It allows the maximum contact between the gas and liquid, facilitating absorption phenomena that result in a product. In a counter current approach, the gas and liquid flow in opposite directions, which enhances the mass transfer between them. The gas and liquid come into contact with each other, allowing the absorption of $NH_3$ from the gas into the liquid. This contact is maximized through the packing material in packed bed scrubbers or the spray of liquid in spray scrubbers (Hadlocon et al., 2015b; Van der Heyden et al., 2015). This ensures the efficient removal of $NH_3$ from the exhaust air in animal facilities, leading to high recovery rates with 91-99% efficiency (Melse and Ogink, 2005).

NH$_3$ scrubbing has been investigated to mitigate gas emissions in various animal facilities, such as poultry and swine farms. Hadlocon & Zhao (2015) studied the ammonia recovery from poultry and swine farms with the production of (NH$_4$)$_2$SO$_4$ Chiumenti and da Borso (2022) worked on N recovery via the acid scrubbing of exhaust air from the thermal drying of sewage sludge. Khakharia et al. (2014) studied design calculations and parameters to recover NH$_3$ by acid scrubbing,

The design of scrubbers has been extensively studied and optimized in numerous studies, with further details provided in the references (Hadlocon and Zhao, 2015; Hadlocon et al., 2015a; Jafari et al., 2018). However, there has been limited research on utilizing ammonia absorption to produce commercial products, such as ammonium sulfate fertilizer, from drying fumes generated during the thermal drying of sewage sludge.

## 2.3 Integration of N recovery from different material flows of sewage sludge treatment

### 2.3.1 Combustion of sewage sludge

Sewage sludge has a high moisture content, which could go up to 98%, and needs to be dewatered before utilizing in thermal processing (Chan and Wang, 2016). Wet sewage sludge can cause several combustion problems when burned directly, including temperature fluctuations, delayed ignition, and reduced furnace temperature (Schnell et al., 2020). In a typical sewage sludge combustion procedure, the wet sewage sludge is mechanically dewatered before being transferred to a dryer to stabilize the combustion process. Wet sewage sludge can be burned directly in a fluidized bed using auxiliary fuel. The combustion products must be appropriately handled and cleaned to ensure safe emissions (Liang et al., 2021). The selection of an incinerator is also crucial depending on the moisture of the sewage sludge, but commonly a fluidized bed, multiple hearth and rotary kiln are used (Gao et al., 2020). A fluidized bed combustor is better regarding environmental impacts than others (Abuşoğlu et al., 2017).

During the combustion of sewage sludge, the nitrogen present in the sludge transforms into exhaust fumes, leading to their subsequent release into the environment and this minimizes the chance of recovering the nitrogen. Furthermore, nitrogen does not end up in ash and P is the only nutrient which is available. In this context, thermal drying is implied prior to combustion (Horttanainen et al., 2017). The practice of thermal drying of sewage sludge followed by its subsequent combustion has gained popularity in numerous advanced nations and is anticipated to witness further growth in the future. This process results in the recovery of nitrogen and mitigates the moisture content within the sewage sludge (Deviatkin et al., 2018).

Liang et al. (2021) review research on municipal sewage sludge combustion and pollution control. Municipal solid waste incinerators, coal-fired power stations, and cement kilns can co-combust sludge without requiring new incineration and flue gas treatment

equipment. Co-combustion can also be environmentally friendly. Staged air combustion, low oxygen dilution combustion, and other approaches can improve sludge combustion and reduce nitrogen oxide emissions. Their results indicated that calcium (Ca)-based compounds remove sulfur well. Staged air combustion and low-oxygen dilution combustion can control sewage sludge combustion NOx.

The combustion of sewage sludge occurs in six steps: drying, devolatilization and auto-gasification, combustion of volatiles, melting of ash, burning of char, and agglomeration of ash. Devolatilization can start at a lower particle temperature and happen concurrently with or after the drying phase, depending on the combustor's design and the sludge's properties (Cui et al., 2005).

Wastewater sludge contains concentrated P and is a promising source for P recovery since it comprises surplus activated sludge and chemical sludge from P removal by coagulation. However, the sludge cannot be used directly as fertilizer due to its high-water content, organic matter, pathogens, and heavy metals (Lim and Kim, 2017). On the other hand, sewage sludge ash (SSA) from sludge combustion plants has a substantially higher P concentration than wastewater sludge. (Liu et al., 2010).

Herzel et al. (2016) has studied the utilization of sewage sludge ash, as a rich phosphorus source. According to their study, approximately 19,000 t of P/year could be recovered in Germany by thermochemically treating sewage sludge ash with sodium and potassium additives. This would convert the residual ash into marketable fertilizer by removing heavy metals. A similar approach to using sewage sludge ash as fertilizer has been studied by removing heavy metals and improving P bioavailability (Jeon and Kim, 2018). Magnesium chloride and calcium chloride have been used as to remove heavy metals. Tonini et al. (2019) worked on comparing the consequential impacts of thermochemical treatment of sewage sludge in context with P recovery. The study determined that the societal costs which include internal cost such as equipment and material costs, external costs such as economic influence through environmental impacts and benefits such as recovery of energy or resource substitutions were lower from rock phosphate.

P can be recovered from ash in various ways, including leaching with acidic or alkaline solutions or utilizing a thermochemical process to produce a P-fertilizer raw material or finished fertilizer. The ash from sewage sludge mono combustion has a high phosphorus content, whereas thermochemically treated sewage sludge ash can be used as a raw material for making P fertilizers or multi-nutrient fertilizers. However, ash from sewage sludge co-combustion is generally unsuitable for phosphorus recovery due to presence of heavy metals (Havukainen et al., 2016).

Although combustion is a common method of treating sewage sludge, the process initially aimed to reduce the waste volume and eliminate harmful elements rather than to recover energy. However, it is possible to recover energy through combustion, but the current use of this process is still primarily focused on waste management purposes (Oladejo et al., 2018; Syed-Hassan et al., 2017).

Singh et al. (2020) compared sewage sludge combustion and anaerobic digestion in terms of electricity production and found that 555–1,068 kWh/t by combustion and 315–608 kWh/t by anaerobic digestion can be produced from sewage sludge (dry basis). Đurđević et al. (2019) found that the combustion of sewage sludge and generation of biogas from anaerobic digestion could fulfil 60% of the requirement of electricity and 100% of the requirement for thermal energy for WWTP processes. Bianchini et al. (2015) worked on integrating a waste-to-energy power (WTE) plant and the thermal drying of sewage sludge. In their study, the waste heat of WTE was used as a source for fulfilling the energy demand of thermal drying of sewage sludge.

Combustion can efficiently convert large amounts of sewage sludge into electricity and/or heat. Therefore, there is an opportunity for cooperation between wastewater treatment plants (WWTPs) and other industrial sectors, including power plants or incinerators. However, a key element in determining the environmental impact of this strategy will be the level of energy recovery attained.

### 2.3.2    Pyrolysis of sewage sludge

In pyrolysis, organic material is thermally degraded at a high temperatures without oxygen. As a result, bio-oil, biochar, and non-condensable gases are produced. Three subgroups of pyrolysis can be defined based on the heating rate and residence period. While fast and flash pyrolysis maximizes the production of liquid bio-oil, slow pyrolysis maximizes bio-char production. In boilers, furnaces, combustors, diesel engines, and gas turbines, bio-oils have undergone extensive testing as potential combustion fuels for generating heat and electricity. These can also be converted into syngas or used as a chemical material. Fast pyrolysis can produce up to 50–70 wt.% of liquid, whereas flash pyrolysis can provide up to 75–80 wt.% of bio-oil (Kan et al., 2016).

The temperature, residence time, heating rate, ambient gases, pressure, and properties of the raw materials used in the pyrolysis process are some of the variables that affect the product's quality. These process variables change depending on the required quality of the finished product. Pyrolysis temperatures commonly vary from 350 to 1,000 °C. Sewage sludge is pyrolyzed to produce biochar, bio-oil, syngas, and water vapor (Fan et al., 2016). Karaca et al. (2018) studied the high-temperature (850 °C) pyrolysis of sewage sludge to recover energy in the form of syngas and tar to be used as a source of energy. Ghodke et al. (2021) studied the pyrolysis of sewage sludge and its potential applications. They analyzed the pyrolysis process, including the influence of temperature on product yields, the characterization of feedstock and reaction products, and the determination of kinetic parameters. This study also found that biochar obtained from sewage sludge pyrolysis can be utilized as organic manure in the agriculture sector due to its nutrient content, such as carbon, nitrogen, phosphorus, and potassium.

Among the many benefits of biochar, it can enhance soil enzymatic activity and firmly bind soil contaminants (Paz-Ferreiro et al., 2018), it can be used as a soil fertilizer to increase crop production (Shang et al., 2018; Taghizadeh-Toosi et al., 2012), and it has

the capability to be used on acidic soils (Šimanský et al., 2018). Ghezzehei et al. (2014) studied the adsorption of $NH_3$ and phosphate on biomass-derived biochar and found it was able to capture 20–43% $NH_4^+$ and $PO_4^{3-}$ from dairy manure during the recovery of N and P. Similarly, Takaya et al. (2016) analyzed biochars from oak wood, press cake from anaerobic digestate, municipal waste, and greenhouse waste for the adsorption capacity to adsorb $NH_4^+$ and $PO_4^{3-}$. Zhang and Wang (2016) also analyzed the adsorption capacity for $NH_4^+$ using a co-pyrolyzed biochar of spent grain from brewers and sewage sludge. Fawzy et al. (2021) reviewed the potential of biochar carbon sequestration, and it was estimated that 0.3–2 Gt $CO_2$ per year would be sequestered by 2050. Sun et al. (2022) worked on the pyrolysis of sewage sludge and minimizing environmental impacts. The results showed that 0.101 tonnes of direct $CO_2$ equivalent ($CO_2$ eq.) (from N-containing organic compounds in sewage sludge) and 1.506 tonnes of indirect $CO_2$ eq. (from energy consumption and dewatering agents) were emitted for each tonne of dry sewage sludge treated during its dewatering and pyrolysis processes, and at least 0.252 tonnes were sequestered as stable carbon in sewage sludge biochar, depending on its final application route. Under similar scenarios, typical sewage sludge treatments emitted 2.432 tonnes of $CO_2$ eq. per tonne of dry sewage sludge.

Barry et al., (2019) studied the slow and fast pyrolysis of sewage sludge to produce biochar and bio-oil along with the leachability of heavy metals in land use. The yields of biochar decreased with the increase of temperature from 300–500 °C in slow pyrolysis and 400–500 °C in fast pyrolysis. Figueiredo et al., (2021) studied the pyrolysis of sewage sludge with different temperatures to estimate biochar production and phosphorus bioavailability to replace P fertilizers. Ghodke et al. (2021) worked on analyzing bio-oil, biogas, and biochar to utilize as engine oil, fuel for spark engines, and organic manure.

### 2.3.3 Nitrogen recovery from reject water of sewage sludge dewatering and condensate of thermal drying fumes

A sizable amount of nitrogen is released into the drying fumes during thermal drying. Condensate is the nitrogen-rich liquid stream that is created when the fumes condense. The nitrogen can be recovered from this condensate by processing it with the water that mechanical dewatering has rejected as studied in (Horttanainen et al., 2017). It has also been found that 78–99% of $NH_4^+$ released exhaust fumes of thermal drying which could be recovered. Mustonen et al. (2017) studied the potential of N recovery from biosludge of the pulp and paper industry and found that 56–74% of $NH_4^+$ released in sludge during thermal drying could be recovered by subsequent stripping and scrubbing of exhaust fumes. Similarly, Deviatkin et al. (2018) studied the potential of nitrogen recovery from the condensate of exhaust fumes originating from thermal drying of sewage sludge.

Reject water, which is formed from the dewatering of sewage sludge, contains up to 15–25% of the total N load of WWTP (Nancharaiah et al., 2016). Many studies have been conducted on reject water sewage sludge dewatering. Koskue et al. (2021) studied the N recovery from reject water from the dewatering of sewage sludge in a 3-chamber bioelectric concentration cell and concluded that N could be recovered with an efficiency

of 75% with synthetic reject water and 53% with real reject water. Guo et al. (2010) studied nitrogen recovery from dewatering reject water using ferric and nitrate bio reductions. Karwowska et al. (2014) researched the characteristics of reject water and condensate from drying to find the BOD, COD, nutrients, and heavy metals levels.

### 2.3.4    **Composting of sewage sludge**

The bioavailability of nitrogen in the soil is increased by composting sewage sludge, which also reduces undesirable pollutants to acceptable levels (Zbytniewski & Buszewski, 2005) and allows for the reutilization of nutrients (Tarrasón et al., 2008). The process of composting shifts organic waste into a stable and effective substance that can be used as organic fertilizer. The organic material in sewage sludge is primarily broken down by bacteria, producing a hygienic and dry end product.

Malińska et al. (2014) found that adding biochar reduces ammonia emissions and nitrogen loss when added as an amendment to composting sewage sludge and wood chip mixtures. Additionally, the presence of biochar led to higher temperatures and extended the thermophilic phase during the composting process. Sánchez-Monedero et al. (2001) investigated N forms during the composting of various organic materials, including sewage sludge, municipal solid waste, brewery sludge, sorghum bagasse, cotton waste, and pine bark., revealing the highest ammonium levels during the early weeks (decreasing to <0.04%), and highest $NO_3$–N (nitrate nitrogen) concentrations at maturation (0.12% to 0.53%). Nitrogen losses varied based on the materials and the pH, with lignocellulose-rich mixtures showing a <25% loss and municipal solid waste mixtures losing >40%. Significant correlations were found between the $NO_3$–N concentration, pH, and electrical conductivity, confirming the role of nitrification. The $NH_4$–N to NO3–N ratio served as a maturity indicator, with final values (0.08, 0.04, 0.16, and 0.11) indicating maturity levels below the established maximum.

Composting, however, can result in foul odours that are increasingly viewed as a type of pollution. The emission chemicals' composition can determine the composting process quality and status. The types of chemicals emitted are directly related to the composting process (Hort et al., 2009). Compost quality is determined mainly by the amount of nitrogen in the decomposed materials. Nitrogen decomposition causes ammonia emission, which is seen as a major issue during the thermophilic stage of composting. The carbon-to-nitrogen ratio, moisture content, substrate composition, temperature, pH, bulking agents, and aeration rate affect how nitrogen is transformed during composting (Malińska et al., 2014). Peat is added to composting sewage sludge to reduce the odour and to make a compost-effective soil product. Tarrasón et al. (2008) suggested that composting digested sludge has a favourable impact on increasing the N availability in the soil.

## 2.4 Evaluation of the economic feasibility of N recovery from sewage sludge

Hadlocon and Zhao (2015) found that based on production rates, swine and poultry scrubber operations cost US\$5.74 and US\$1.11 per kg of pure ammonium sulfate, respectively. Dizaji et al. (2022) examined the techno-economic feasibility of incineration in new combustion facilities to recover energy and P fertilizer from sewage sludge. The results demonstrated a strong dependence on energy and fertilizer market prices and sewage sludge gate fee support. Heat was favoured over CHP, which was only viable at 100 MW and 8,000 h/y. Co-combustion costs 19–30 €/MWh less than mono-combustion (29–66 €/MWh) due to 25–35% lower energy demand and 17–25% higher fuel heating value.

Ahmed et al. (2016) studied that the cost of various biochar feedstocks, such as sewage sludge, sphagnum peat/coconut coir, yard waste, greenwood, compost, and others, and found that the cost ranged from 6.71 to 110 US\$ per ton. This cost was found to be lower than the overall production cost of biochar, which ranged from 51 to 5,668 US\$ per ton. Capital, storage, utility, labor, and other plant-related expenditures are included in the production cost. Callegari and Capodaglio (2018) found that despite biochar having several times higher production costs than feed costs, the predicted market price for sewage sludge biochar is 246 US\$/ton, which is six times lower than the price of 1,500 US\$/ton for commercially activated carbon.

Tomei et al. (2016) conducted a feasibility assessment of two upgrading alternatives for sewage sludge stabilization, aiming to reduce sludge production and improve its quality for either agricultural use or combustion. The cost analysis revealed that the upgrading alternatives are generally more cost-effective than a conventional plant, with the economic advantage varying based on local conditions. The post-aerobic stabilization stage showed the highest potential for cost savings, exceeding 6.00 €/ (person equivalent (PE) year) for both disposal options. However, uncertainties remain, especially regarding capital costs for upgrading existing plants with new equipment, and accurate evaluation of sludge disposal cost, and an income from the sale of electric energy would be crucial for real-world applicability.

## 2.5 Evaluation of the environmental impacts of N recovery and integrated recovery from treatment of sewage sludge

An LCA of sewage sludge treatment has been studied in various ways. Gievers et al. (2021) studied the LCA of sewage sludge pyrolysis and made a comparison with incineration of sewage sludge. The results showed that the usage of biochar in horticulture was the best environmentally friendly option regarding the global warming potential (GWP) with net emissions of 2 $CO_2$ eq./kg sewage sludge and 78% fewer $CO_2$ eq. emissions in comparison to the mono incineration of sewage sludge. Huang et al. (2022) studied the feasibility of pyrolysis scenarios for sewage sludge treatment through an LCA,

and examined the energy consumption, carbon emissions, and carried out economic benefit analyses, considering circular economy principles and comparing pyrolysis scenarios for activated carbon and biochar. Bio-oil and biochar pyrolysis scenarios with 1,000kg of dried sewage sludge as a functional unit (FU) showed significant environmental improvements (-0.31 kg $CO_2$ eq./kg and -0.05 kg $CO_2$ eq./kg FU, respectively) compared to conventional and current methods. Activated carbon scenarios had a higher toxicity but lower carbon emissions (1.50-1.70 kg $CO_2$ eq./kg FU) than current activated carbon production processes. The examined pyrolysis scenarios were potentially profitable, with bio-oil and biochar achieving net energy recovery through product applications, whereas activated carbon processes required energy input.

Xiao et al. (2022) examined the environmental, energy, and economic effects of sewage sludge management methods by utilizing an LCA, and examining the cumulative energy demand (CED), and life cycle cost (LCC). The study looked at scenarios involving sewage sludge disposal by mono-incineration, co-incineration in a municipal solid waste (MSW) incineration plant, a coal-fired power plant, and cement kiln. Global warming is the main environmental impact of sludge incineration. After that, eutrophication, marine aquatic ecotoxicity, and human toxicity potential are important in all four scenarios. LCA, CED, and (LCC) values of 5.41 x $10^{-9}$, 1736 MJ, and 253k US\$, respectively, showed that the mono-incineration scenario had the highest environmental impact and required the most energy and economic resources. Co-incineration in cement kilns had the lowest CED (368 MJ), LCC (81k US\$) and environmental burden (1.02 x $10^{-9}$). Morsink-Georgali et al. (2022) conducted a comparison of composting in windrow facilities and biogas production by anaerobic digestion. It concluded that in the production of biogas from sewage sludge, 27,060 MJ/ t of sewage sludge energy saved, and 697 kg/t of digested sludge were avoided carbon emissions as compared to composting of sewage sludge. Deviatkin et al. (2019) investigated nitrogen release during the thermal drying of sewage sludge and its recovery for fertilization, including the possibility of nitrogen recovery from condensate. The proposed nitrogen recovery system had a 28% lower global warming potential than conventional fertilizer production (4.1 kg $CO_2$ eq. $kg^{-1}$ N vs. 5.7 kg $CO_2$ eq. $kg^{-1}$N). However, a sensitivity analysis suggested that under specific process parameters, the global warming potential could increase to 6.2 kg $CO_2$ eq. during nitrogen recovery.

Kar et al. (2023) studied the feasibility of ammonia recovery from WWTPs through air-stripping for ammonium sulfate (AS) for production as fertilizer. A life cycle assessment showed significantly lower GHG emissions (0.2-0.5 kg $CO_2$ eq./kg AS) compared to the conventional Haber-Bosch process (2.5 kg $CO_2$ eq./kg AS). A techno-economic analysis suggested a break-even selling price of 0.046 US\$/kg of AS, making air-stripping an environmentally and economically attractive option for nitrogen recovery and AS production at WWTPs. van Zelm et al. (2020) evaluated the environmental impacts of the Nijhuis Ammonia Recovery system (AECO-NAR) for ammonia removal and ammonium sulfate (AS) production, compared to a combined SHARON-anammox (Single reactor system for High activity Ammonium Removal Over Nitrite) plant for nitrogen removal. Integrated water treatment and AS production in the AECO-NAR system resulted in

negative impact scores, avoiding the impacts associated with conventional AS production. The total impacts of the AECO-NAR approach decreased with higher ammonia inflow concentrations, and the system outperforms separate ammonia recovery and wastewater treatment processes. Renewable energy use and sustainable cleaning chemicals further improved the system's environmental performance. Lam et al. (2020) provided details on the adaptability and application of life cycle assessments (LCAs) in evaluating the environmental impact of nutrient recycling from wastewater for agricultural use. Their study analyzed 65 LCA studies and found that wastewater-based nutrient recycling generally had good environmental results when minimizing chemical inputs and implementing source separation of human excreta. The review emphasized the need for methodological consistency, transparency in inventory and methods, consideration of uncertainty, and integration of interdisciplinary knowledge to optimize wastewater-based nutrient recycling decisions at various scales. Opportunities include performing LCA on recycled nutrient products, integrating LCA with other systems approaches, and assessing emerging nutrient recovery technologies and integrated resource recovery systems. In all the studied literature, it was found that there is a gap in the LCA of reject water from dewatering processes and condensate from the fumes of thermal drying for nitrogen recovery using biochar as an adsorbent and a comparison with other nitrogen recovery technologies.

# 3 Materials and methods

This dissertation used three methods to evaluate nitrogen recovery from municipal sewage sludge as follows:

**i)**   ASPEN Plus modeling for stripping and scrubbing units
**ii)**  Mass and energy calculations for sewage sludge recovery technologies and their integration with WWTP
**iii)** LCA for environmental impact assessment

In Publication I, stripping and scrubbing units were designed to estimate the nitrogen recovery from the drying fumes from the thermal drying of sewage sludge. ASPEN Plus, a flowsheet simulation software program, was used to model chemical processes from raw materials to the final product. A steady-state flowsheet model for the modeling of the scrubber was used. In Publication II, mass and energy calculations were performed to assess the nitrogen recovery from WWTP by integrating combustion and pyrolysis with scrubbing with acidic media. In Publication III, the environmental performance of stripping and scrubbing units was analyzed by integrating pyrolysis and combustion in LCA modeling. Finally, in publication IV, LCA modeling was performed to calculate the environmental impacts of nitrogen recovery from reject water from dewatering and condensate from the drying fumes of thermal drying of sewage sludge using stripping and scrubbing technology and adsorption on biochar in comparison to WWTP.

## 3.1 ASPEN Plus

ASPEN provides a model library with many different unit operations, including splitters, heat exchangers, reactors, and distillation columns, allowing for the precise simulation of large-scale processes. By linking these subprocesses, engineers can generate process flow diagrams that the program then solves with the help of Fortran codes. To further aid in the reduction of input requirements and the minimization of human error throughout the solution process, ASPEN includes a comprehensive thermodynamics and physical characteristics database. It also provides a graphical user interface that operates independently from the simulation engine.

ASPEN Plus, one of the available bundles, is optimized for steady-state simulation of chemical, petrochemical, and refinery processes. ASPEN Plus uses a modular sequential simulation approach, but users can switch to an equation-based approach for some models. The program can also be used to model processes in batches (ASPENTech).

The following are the basic steps for simulating in ASPEN Plus.

1) Specify or introduce chemical components:
   - Obtain the chemical components from ASPEN Plus databanks or introduce them manually.
   - Specify the chemical compounds or substances involved in the process.
2) Select thermodynamic models:
   - Choose appropriate thermodynamic models to represent the physical properties of components and mixtures.
   - ASPEN Plus provides a range of built-in thermodynamic models for accurate representation.
3) Define the process flowsheet:
   - Identify and define the unit operations required in the process (e.g., reactors, separators, heat exchangers).
   - Specify the process streams that enter and exit each unit operation.
   - Utilize the models available in the ASPEN Plus Model Library to describe each unit operation or chemical synthesis.
   - Place the selected models onto the process flowsheet, representing each unit operation.
   - Label each unit operation model (block) and establish connections between them using process streams.
4) Specify feed stream conditions:
   - Determine the flow rates of each component in the process streams.
   - Define the thermodynamic conditions such as temperature, pressure, and composition for all feed streams.
5) Set operating conditions for unit operation models:
   - Specify the desired operating conditions (e.g., temperature, pressure, residence time) for each unit operation model.
   - Adjust parameters and variables within the models to accurately represent the desired behaviour (Al-Malah and Kamal I. M., 2017).

## 3.2  LCA: Methodology and principles

The life cycle assessment (LCA) studies carried out for this dissertation follow the guidelines and framework provided in ISO 14040 and ISO 14044, which offer a standardized methodology for carrying out life cycle assessments. Through the LCA process, these criteria ensure dependability and transparency. While ISO 14044 outlines the prerequisites and best practices for carrying out LCA investigations, ISO 14040 outlines the LCA's guiding principles and framework.

During the goal and scope phase, the purpose and motivations of the study are established, along with other relevant details. The product system to be studied, functional unit, system boundary, impact categories, impact assessment methodology, interpretation,

assumptions, limitations, and data quality requirements are also defined. This phase ensures consistency and adequacy in addressing the objectives (ISO 14040, 2006). Figure 1 illustrates the LCA framework.

The selection of impact categories in LCA studies should encompass all relevant environmental issues related to the assessed product system. The specific impact categories chosen may vary depending on the applied impact assessment methodology. However, the International Reference Life Cycle Data System (ILCD) handbook recommends including midpoint impact categories such as human toxicity, radiation, carcinogens, respiratory inorganics, climate change, ozone layer depletion, acidification, eutrophication, ecotoxicity, summer smog, land use, and resource depletion (EC-JRC, 2010). The areas of protection (AoPs) should cover human health, the natural environment, and natural resources. It should be noted that the selection of impact categories depends on the goals, scope, and LCI of the study, which may lead to certain limitations and deviations from the full implementation of the ILCD recommendations (ISO 14040, 2006).
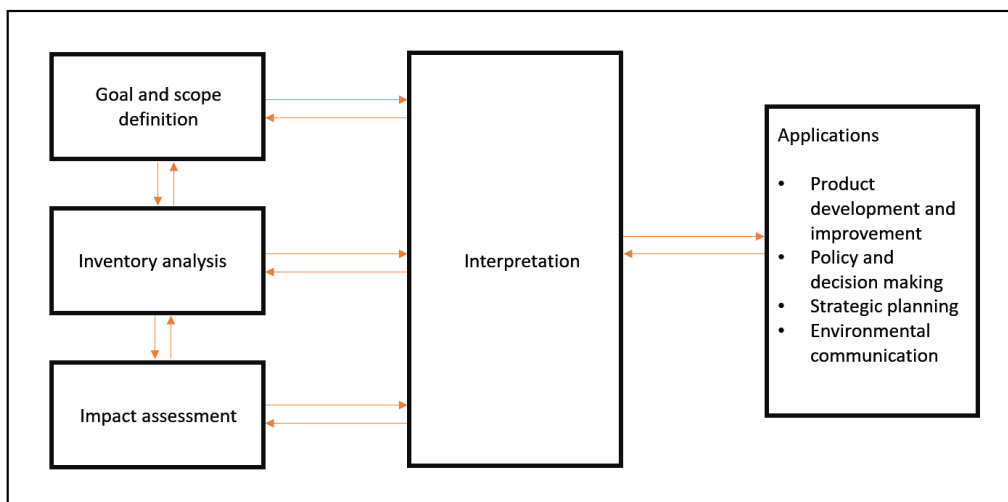


Figure 1. LCA framework with main phases (ISO 14040, 2006; ISO 14044, 2006)(ISO 14040, 2006; ISO 14044, 2006)(ISO 14040, 2006; ISO 14044, 2006).

The functional unit, an essential aspect of the goal and scope definition, quantifies the identified functions of the product and serves as a reference for relating all inputs and outputs (ISO 14040, 2006). It must align with the goals, be measurable, and be clearly defined. The system boundary outlines the unit processes included in the LCA and must be coherent with the objectives. The scope definition involves decisions regarding the depth of study for the included unit processes. While the assessment typically covers the entire life cycle from raw material acquisition to end-of-life and recycling (known as

"cradle-to-grave" and "cradle-to-cradle" approaches), certain stages or processes can be excluded if they have minimal impact on the overall LCA results. However, such exclusions must be clearly indicated, and their implications explained. A process flow diagram demonstrating the beginning and end points of the unit processes and how they are interconnected is used to depict the system boundaries (ISO 14044, 2006). The components of the system boundary are determined by the specification of the goal and scope, intended use, data limitations, and cut-off criteria (ISO 14040, 2006). (ISO 14040, 2006).

During the inventory analysis step, both primary and secondary data are gathered. Calculating and modeling the prospective environmental performance based on the inventory data are parts of the impact assessment and interpretation phases. A sensitivity analysis is also carried out to assess the reliability of the results. Details are provided later in this section. The inventory analysis, or life cycle inventory (LCI) analysis, entails collecting data and calculating inputs and outputs for each unit process within the system boundary. This phase is iterative and data-intensive, involving primary or secondary data collection from various sources such as production sites and the literature. The data is classified into energy inputs, raw materials, ancillary inputs, products, co-products, waste, and emissions. The collected data should be detailed, referenced, and any deviations from data quality requirements should be stated (ISO 14044, 2006). As the LCI phase progresses, limitations and new data requirements may arise, leading to adjustments in data collection procedures (ISO 14040, 2006).

Using the results from the LCI phase, the life cycle impact assessment (LCIA) phase assesses any potential environmental effects related to the functional units. It involves connecting LCI data to categories of environmental impact and delivers details for the interpretation stage. In order to conduct an accurate assessment in accordance with the specified aims and scope, LCIA takes into account the system boundary, LCI data quality, availability of results, and their suitability (ISO 14044, 2006). In this phase, LCI results are classified into impact categories and category indicator results are characterized (ISO 14044, 2006).

The interpretation phase involves discussing the findings from the LCI and LCIA. It should align with the defined goals and scope, identify and evaluate significant issues and their sensitivity to the overall LCA results, and indicate potential environmental impacts (ISO 14044, 2006). A contribution analysis is commonly used to determine the main contributors to LCIA results, identifying a product system's most significant processes and elementary flows. It can also help identify the need for additional data collection and guide product improvement. A sensitivity analysis is performed to assess the influence of choices made during the LCA, such as methodological choices and assumptions, on the accuracy of the overall results. It helps identify uncertainties and analyze their impact on the reliability of the LCA results (EC-JRC, 2010).

During the life cycle impact assessment (LCIA) phase of an LCA, the environmental performance is evaluated by characterizing elementary flows and their causally related impacts on AoPs. LCIA indicators can be classified into two levels: midpoint and endpoint. Midpoint indicators focus on specific environmental issues, while endpoint indicators describe the final impacts on AoPs (EC-JRC, 2010).

## 3.3  Modelling nitrogen recovery by scrubber

Using scrubbing with acidic media to remove and recover ammonia gas from drying fumes was modelled in Publication I. This modeling was utilized further in Publications II-IV, where stripping and scrubbing were utilized. Publication I was the basis of stripping and scrubbing calculations and modeling in Publication II-IV.

Scrubbing with acidic media was employed to control and neutralize ammonia emissions by producing ammonium sulfate fertilizer. The proposed model utilized a counter-current packed bed scrubber to recover ammonia from the drying fumes. The scrubber was designed using the ASPEN Plus simulator, a flowsheet simulation software for modeling chemical processes.

As an overview of reaction kinetics, which is also discussed in detail in section 3.3.5, the ammonia gas and water went through a dissociation reaction, resulting in the formation of ammonium ions. Similarly, sulfuric acid dissociates into bisulfate and hydronium ions. Ammonium and sulfate ions combine to form ammonium sulfate salt. Equilibrium constants for the reactions were calculated using a rate equation. The electrolyte non-random two liquid (ENRTL) method was used for a thermophysical property analysis to determine the thermodynamic properties of the liquid phase. The proposed model and scrubber design were validated using data from previous studies on ammonia scrubbing. The model was validated by comparing the predicted results with the experimental data from these studies.

The scale-up of the scrubber includes calculating scrubber dimensions, such as height and diameter. The height of the scrubber and packing height were calculated based on equations considering the height equivalent to the theoretical plate (HETP). A cost estimation was performed using cost curves and equations to estimate equipment, installation, and operational costs.

In general, the materials and methods described focused on the design, modeling, and cost estimation of a scrubber for ammonia removal from drying fumes produced by the thermal drying of sewage sludge. The methods employed included process simulation, thermodynamic analysis, and validation using previous studies.

### 3.3.1    Parameters of modeling

For the preliminary design of the scrubber, Khakharia et al. (2014) and Melse and Ogink (2005) were chosen as references. Acid packed bed scrubbers were developed in the research by Melse and Ogink (2005) to remove $NH_3$ from pig and poultry operations. Several scrubbers with various flow rates and ammonia concentrations were used, and the results were given. It was deduced from the study that $NH_3$ elimination ranged from 40% to 100%, with an average value of 96%. Khakharia et al. (2014) have also treated ammonia emissions from a post-combustion $CO_2$ capture plant using a scrubber. In order to test the scrubber under extreme circumstances, the ammonia inlet concentration was increased to 150 mg/$Nm^3$ from 15-20 mg/$Nm^3$ and the ammonia outlet concentration was dropped to 5 mg/$Nm^3$. These studies were the foundation for choosing the process parameters and conditions in Table 2.

Table 2. Process parameters of modeling for Publication I

| Parameter | Values |
|---|---|
| Flow rate of gas | 1,000 $m^3$/hr |
| Concentration of ammonia | 75 and 100 ppm |
| $H_2SO_4$ pH | 1-5 |
| Liquid to gas ratio (L/G) | 4.3 |
| Liquid flow rate | 70 L/min |
| Temperature | 100 °C |
| Pressure | 1 atm |
| Superficial velocity | 1.4 m/s |

### 3.3.2    Equipment sizing

The height of the column, packing height, and diameter of the scrubber were calculated as follows:

The height of the scrubber was calculated from Eq. 1 (Bhoi et al., 2015)

$$H_{column} = 1.4H_{pack} + 1.02D + 2.81 \qquad (1)$$

where

$H_{column}$ is the height of the scrubber in m

$H_{pack}$ is the height of the packing in m

D is the diameter of the scrubber in m

The height of the packing is calculated from Eq. 2 (Bhoi et al., 2015):

$$H_{pack} = N \times HETP \qquad (2)$$

where

$H_{pack}$ is the height of the packed bed in m

N is the number of equilibrium stages

HETP is the height equivalent to the theoretical plate.

and the diameter was calculated from:

$$D = \frac{\sqrt{4A}}{\pi} \qquad (3)$$

where

A is the area of the scrubber calculated from Eq. 4 in m$^2$, and the value of $\pi$ was taken as 3.14.

The area is calculated from Eq. 4:

$$A = \frac{Q}{u_s \times 3600} \qquad (4)$$

where

Q is the flow rate of gas in m$^3$/hr

$u_s$ is the superficial velocity and was taken as 1.4 m/s (Melse and Ogink, 2005)

### 3.3.3 Cost Estimation

The cost estimation of equipment is a vital part of the techno-economic analysis. In ammonia recovery, the scrubber cost is the critical equipment cost. The equipment cost can be calculated using three different methods, with varying accuracy. The price quoted by vendors is an accurate method but requires extensive details, specifications, and substantial engineering work. The second method is to find an equipment cost from databases with the same specification and processing; it can be challenging to find an exact match in databases with the same conditions. Due to the unavailability of exact equipment costs from vendors or databases, an alternative to these two methods is calculating from cost curves and equations for a preliminary estimation. This method was applied to estimate the cost in this study.

Several correlations and graphs are given in the literature based on different methods of calculation (Towler and Sinnott, 2012; and Peter and Timmerhaus, 2003). The method by Towler and Sinnott (2012) of cost calculation has been used for the equipment cost, and different percentages of variations have been taken from (Peter and Timmerhaus, 2003).

Eq. 5 was used to calculate the mass of the scrubber and the cost of equipment purchased:

$$C_e = a + bS^n \qquad\qquad (5)$$

where $C_e$ is Cost of the equipment purchased

a, b is the cost constants and values, taken as 11,600 and 34, respectively (Towler and Sinnott, 2012)

S is size parameter (Shell mass)

n is exponent for equipment, value 0.85 (Towler and Sinnott, 2012)

The shell mass of the scrubber, S, is the size parameter used to calculate purchase cost. It is calculated from Eq. 6:

$$S = \pi D_c L_c t_w \rho \qquad\qquad (6)$$

, where

$D_c$ is the vessel diameter, 2 m

$L_c$ is the vessel length, 11.5 m

$t_w$ is the wall thickness, 0.007 m

$\rho$ is the metal density, 7650 kg/m$^3$

The operational cost includes the cost of sulfuric acid, 0.4 €/kg (Lietjärvi, 2020) and water price 1 €/m$^3$, which is estimated on the basis that the scrubber is in the same facility and no additional transmission cost is needed (Saud, 2017). The annual cost was calculated first, including, and then excluding the cost of water, to analyze the difference in the cost of ammonia recovery per ton of sludge treated. Miscellaneous costs, such as installation, instrumentation, piping, electrics, and costs related to engineering, construction, and services, were also calculated from factors given by (Peter and Timmerhaus, 2003).

### 3.3.4    **ASPEN Plus modeling and simulation**

The scrubber was modelled in ASPEN Plus with ammonia gas with air entering from the bottom of the scrubber as a stream titled GASIN and diluted sulfuric acid entering from the top of the scrubber as a stream titled LIQIN. The scrubber's LIQOUT stream, which is made up of water and ammonium sulfate, exits from the bottom while the scrubber's EXHAUST stream vents treated air from the top, as shown in Figure 2. The counter-current scrubbing was used to maximize mass transfer. The necessary equilibrium data were gathered from the literature and the ASPEN databank, and equilibrium reactions were considered.
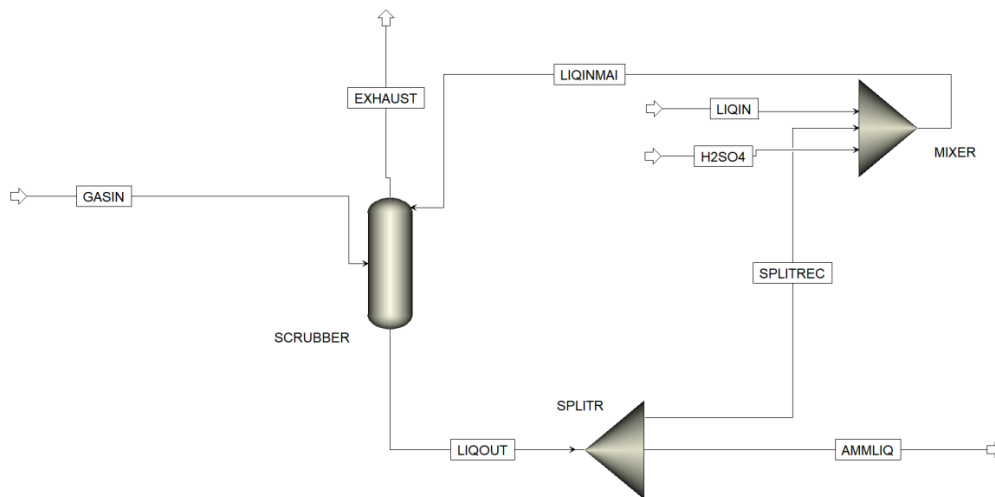
Figure 2. Process flow diagram of simulation of scrubber with sulfuric acid in Publication 1.

The ASPEN Plus simulator simulates the acid cleaning of ammonia gas. The ENRTL method was selected as a thermophysical property analysis to determine the thermodynamic properties of the liquid phase. The literature recommended the selection of this thermodynamic modeller system, including electrolytes (Bolzonella et al., 2018; Bravo et al., 2018; Tang et al., 2017). It was widely recommended that an RadFrac column be used when designing a scrubber. RadFrac is appropriate for simulating vapor-liquid stream-based distillation, stripping, and absorption processes and can simulate equilibrium, rate-based, and electrolytic chemical reactions.

For this model, a rate-based calculation was used. The Maxwell-Stefan model, which also computes binary coefficients to predict vapor and liquid mass transfer, is used in rate-based RadFrac modeling to compute multicomponent mass transfer. Absorption phenomena are explained based on the two-film theory, which describes how mass transfer resistance occurs in a thin layer next to or close to the bulk phase. The heat and mass balance related to discretized film equations to achieve a precise temperature and concentration profile in the column (Tang et al., 2017).

### 3.3.5 Reaction Kinetics

As shown in Table 3, a dissociation reaction produces ammonium $NH_4^+$ ions when ammonia and water come into contact. Similarly, sulfuric acid separates into hydronium ions ($H_3O^+$) and bisulfate ($HSO_4^-$) ions. Further dissociating bisulfate ions create

hydronium, $H_3O^+$, and sulfate ions $SO_4^{-2}$. Ammonium sulfate salt is created when the ions of ammonium ($NH_4^+$) and sulfate ($SO_4^{-2}$) react.

Table 3. Reaction kinetics of all involved reactions

| Reaction no. | Type of Reaction | Chemical Equation |
|---|---|---|
| 1 | Equilibrium | $H_2O + HSO_4^- \leftrightarrow H_3O^+ + SO_4^{-2}$ |
| 2 | Equilibrium | $H_2SO_4 + H_2O \leftrightarrow H_3O^+ + HSO_4^-$ |
| 3 | Equilibrium | $NH_3 + H_2O \leftrightarrow OH^- + NH_4^+$ |
| 4 | Equilibrium | $2H_2O \leftrightarrow OH^- + H_3O^+$ |
| 5 | Salt | $(NH_4)_2SO_4 \leftrightarrow NH_4^+ + SO_4^{-2}$ |

For the calculation of equilibrium constant K for reactions, the following rate equation was used for calculation:

$$lnK = A + \frac{B}{T} + C \ln T + DT$$

where T is the absolute temperature, A, B, C, and D are equilibrium parameters. Their values are available in the ASPEN databank, as shown in Table 4.

Table 4. Reaction parameters for the equilibrium constant K

| Parameter | A | B | C | D |
|---|---|---|---|---|
| Reaction 1 | -5.3932 | $1.733 \times 10^3$ | 0 | 0 |
| Reaction 2 | -3.8983 | $3.474 \times 10^3$ | 0 | 0 |
| Reaction 3 | -1.257 | -3335 | 1.4971 | -0.0371 |
| Reaction 4 | 132.9 | $-1.345 \times 10^4$ | -22.4773 | 0 |
| Reaction 5 | -216.6 | $4.262 \times 10^3$ | 37.518 | -0.0799 |

## 3.4   Integration of pyrolysis and combustion with scrubbing

### 3.4.1   Description of study

The focus of Publication II was on integrating pyrolysis or combustion with scrubbing to enhance nutrient and energy recovery from municipal sewage sludge. The results of Publication II were also the basis of the data for Publication III and Publication IV. The study was conducted based on data from a large-scale WWTP in Finland, where sewage sludge was collected and processed using different methods. Mass and energy balance

calculations were used to estimate the amount of nutrients and energy recovered from each process and the costs and revenues associated with each option.

The study focuses on two sludge treatment and recovery processes: pyrolysis and combustion, which are being piloted in Finland. A comparison was made of these processes in terms of their efficiency and nutrient recovery potential. Six different scenarios were analyzed, varying in the inclusion or exclusion of nitrogen recovery from thermal drying fumes and WWTP reject water. The scenarios investigated are discussed in detail in section 3.5.2.

Although the pyrolysis and combustion pilot plants and WWTPs were substantially different in size, the same sludge input flow was used for both processes to ensure comparability. The technical data from the pilot plants, specifically the pyrolysis and combustion scenarios, were utilized. In the pyrolysis scenario, sewage digestate and wood waste were pyrolyzed to produce biochar and pyrolysis gas. The reasons for the additional use of wood in the pyrolysis scenario are to make pyrolysis process energy self-sufficient and increase the biochar's carbon content. In the combustion scenario, the sewage sludge was combusted, and the excess energy was used for district heating. The nutrient-rich ash produced from combustion was used as forest fertilizer.

The recovery of nutrients, particularly nitrogen, was addressed in both scenarios. Nitrogen was recovered from the reject water and thermal drying fumes by employing a stripping and scrubbing column. The stripped gas was then directed to scrubber to capture ammonia using sulfuric acid and produce ammonium sulfate.

### 3.4.2 Scenarios

The technical information from the two pilot plants Viikinmäki WWTP in Helsinki, run by Helsinki Region Environmental Services (HSY) and Endev Oy, were used.

The following six scenarios were considered:

**PII-S1.0:** Pyrolysis without N recovery

**PII-S1.1:** Pyrolysis with N recovery from thermal drying fumes but not from WWTP reject water.

**PII-S1.2:** Pyrolysis with N recovery from both thermal drying fumes and WWTP reject water.

**PII-S2.0:** Combustion without recovering N.

**PII-S2.1:** Combustion with N recovery from thermal drying fumes but not from WWTP reject water.

**PII-S2.2:** Combustion with N recovery from both thermal drying fumes and WWTP reject water.

The base scenarios without nitrogen recovery are PII-S1.0 and PII-S2.0. The ash product was used as a forest fertilizer in the combustion base scenario, while the produced heat was used as district heat. The sewage digestate was used in the pyrolysis base scenario to produce biochar, pyrolysis oil and gas were burned to generate the heat energy needed for the thermal drying and pyrolysis process. To ensure energy sufficiency, wood waste was also used in the pyrolysis process; to keep the input energy the same in the scenarios, though there was no need for the extra energy from the technical point of view.

### 3.4.3 Wastewater Treatment Plant, Sludge Digestion, and Dewatering

The Viikinmäki WWTP facility received wastewater with a flow rate of $92 \times 10^6$ m³/a and total solids (TS) of 29,000 t/a. In terms of population equivalent, the plant potential was 3 m³/day/person. The nitrogen and phosphorus mass quantities were 4,900 t/a and 600 t/a, respectively. The rest of the properties of dewatered sludge and wood waste which used in calculations are presented in Table 5. After pre-treatment, screening, and aeration, the unprocessed sludge undergoes anaerobic digestion, which produces biogas and digestate. The biogas generates heat and electricity, while the remaining digestate is dewatered. The difference between the digestate and dewatered sludge masses determined the quantity of reject water.

Table 5. Properties of sewage sludge digestate and wood waste from Viikinmäki WWTP (HSY, 2018)

| Parameter | Dewatered Sludge | Wood Waste | Unit |
|---|---|---|---|
| TS | 29 | 58 | % of wet mass |
| Ash | 43 | 10 | % of TS |
| N | 0.04 | 0.02 | kg/kg TS |
| P | 0.03 | 0.002 | kg/kg TS |
| $LHV_{dry}$ | 13 | 18 | MJ/kg TS |
| $LHV_{ar}$ | 1.8 | 9.6 | MJ/kg |

The WWTP's electricity consumption was 0.42 kWh/m³ and the heat consumption was 0.38 kWh/m³. The digestate had a 95% moisture content after anaerobic digestion and sequential moisture removal procedures were essential for thermally treating the sludge. The pyrolysis of wet digestate produces steam in the reactor, resulting in either an increase in the product's moisture content or an increase in the production of non-condensable gases. In addition, the combustion of the high moisture digestate would not reach the temperatures mandated by waste combustion laws. To reduce the moisture content to 65% and 5%, respectively, sewage sludge was subjected to mechanical dewatering and thermal

dehydration (Kätkä, 2013). Dewatering was performed before thermal drying to reduce energy consumption (Schnell et al., 2020). Dewatering the digestate reduces the volume of the sludge by removing water, thereby facilitating transport and increasing the lower heating value as received ($LHV_{ar}$) (Wu et al., 2021).

The reject water as well as the thermal drying vapor, which was directed to the condenser and produces nitrogen-containing condensate, both contained recoverable nutrients, primarily nitrogen. Mass balance calculations were performed for both dewatering and thermal drying, with nitrogen being the primary focus in all phases. The nitrogen content of the reject water was approximately 16% of the total nitrogen in the wastewater, which falls within the range of 10–30% reported in previous investigations (Guo et al., 2010). The desired level of moisture removal during thermal drying was 80% (Deviatkin et al., 2018). The results of Publication I and (Deviatkin et al., 2018) about the nitrogen recovery from the thermal drying of sewage sludge utilizing conventional scrubber technology were used. Table 6 presents the mass balance for the WWTP and the dewatering and thermal drying processes.

Table 6. Mass flows of WWTP considered in Publication II (HSY, 2018).

| WWTP | Value | Unit |
|---|---|---|
| Wastewater | 250,000 | $m^3/d$ |
| Nitrogen | 13 | t/d |
| Phosphorus | 1.6 | t/d |
| TS | 79 | t/d |
| **Clean water** | | |
| Mass | 250,000 | t/d |
| Nitrogen | 1.2 | t/d |
| Phosphorus | 0.041 | t/d |
| **Mass to digestion** | | |
| Mass | 2,600 | t/d |
| TS | 88 | t/d |
| Nitrogen | 3.6 | t/d |
| Phosphorus | 1.4 | t/d |
| **Dewatered digestate** | | |
| Mass | 180 | t/d |
| TS | 52 | t/d |
| Nitrogen | 1.5 | t/d |
| Phosphorus | 1.4 | t/d |
| **Reject water** | | |
| Mass | 2,400 | t/d |

| | | |
|---|---|---|
| TS | 1.1 | t/d |
| Nitrogen | 2.1 | t/d |
| Phosphorus | 0.033 | t/d |
| **Thermally dried digestate** | | |
| Mass | 58 | t/d |
| TS | 52 | t/d |
| Moisture content | 10 | % |
| Nitrogen in drying fumes | 0.74 | t/d |
| **Condensate** | | |
| Mass | 120 | t/d |
| Nitrogen | 0.47 | t/d |

### 3.4.4  **Pyrolysis Scenario**

Figure 3 depicts a pyrolysis scenario in which wood waste is combined with digestate to produce biochar and pyrolysis gas. Wood waste was added to compensate for the lower heating value of the digestate and to introduce more carbon. 70% of the pyrolyzed mass was digestate, and the remaining 30% was wood waste. It was considered that the pyrolysis process is self-sufficient in terms of heat energy through the combustion of the pyrolysis gas and recovery of the heat for thermal drying and pyrolysis. The thermal drying process requires 0.45 kWh/kg of electricity (Đurđević et al., 2019), while the pyrolysis process requires an additional 0.1 kWh/kg (HSY, 2019). Table 7 lists the parameters                        of                        the                        pyrolysis                        process.

Table 7. Parameters for wood pyrolysis (HSY, 2018)

| Parameter | Value | Units |
|---|---|---|
| Wood waste | 0.42 | kg wood/kg of sludge |
| Electricity Demand | 0.1 | kWh/kg |
| Transfer Coefficient for Biochar | | |
| Ash | 100 | % |
| Volatile Solids (VS) | 11 | % |
| Water | 0 | % |

Figure 3. Block diagram of the pyrolysis process. Solid lines denote the current process and dotted lines show the possibilities to recover nitrogen from the reject water and thermal drying fumes.

### 3.4.5    Combustion Scenario

The sludge combustion data come from a novel sludge combustion facility located in Rovaniemi (Finland) and constructed by Endev Ltd., with a 10,000 t/a capacity. Following mechanical dewatering in the WWTP, the sludge enters a fluidized bed drying and combustion process as shown in Figure 4. First, the sludge is mixed with hot sand from a combustion reactor in a dryer to remove moisture, thereby increasing the sludge's total solid content to 95–98 wt.%. After the sludge has been dried, it is conveyed to a fluidized bed reactor for combustion. To ensure the annihilation of organic pollutants (pathogens, drug residues, microplastics, etc.), the temperature in the reactor is maintained at 850 °C. Ash is carried with flue gases and extracted at two points: the majority of the ash (>95 wt.%), the so-called product ash, is collected using a high-temperature cyclone after an air preheater, and the finer ash is removed as by-product ash

using a bag filter. The combustion-produced heat is used for thermal drying and district heating, while the hygienic and nutrient-rich product ash can be used as fertilizer. The fluidized bed dryer's water vapor is directed to a condenser, and the condensate is sent to the WWTP for effluent treatment. Up to 3,000 mg/L of recoverable nitrogen is present in this condensate, according to an analysis carried out at the pilot plant of Endev Oy. In terms of electricity the combustion plant consumes 0.147 kWh/t of dewatered sludge, and the plant's heat efficiency is 82%, as shown in Table 8.

Table 8. Parameters for combustion scenario (Endev, 2020).

| Parameters | Value | Units |
| --- | --- | --- |
| Electricity demand | 0.147 | kWh/kg sludge |
| Heat efficiency | 82 | % |
| Nitrogen to condensate | 30 | % |
| Ash to product ash | 95 | % |
| By-product ash | 5 | % |
| NaOH demand | 0.0048 | kg/kg sludge |

The addition of a biomass boiler made the mass and energy balance comparable to the pyrolysis scenario. Using Equation (7), the lower heating value, $LHV_{ar}$ of thermally dried digestate and the wood residue can be calculated as

$$LHV_{ar} = LHV_{dry} \times (1 - w) - l_{25}w$$

where the lower heating value for total solids ($LHV_{dry,D}$) for thermally dried digestate was 13 MJ/kg, and for wood ($LHV_{dry,W}$) was 18 MJ/kg. In addition, the moisture content, w, for thermally dried digestate was 5%, for wood waste it was 41%, and the heat of vaporization of water, $l_{25}$, was 2,443 MJ/kg. Therefore, the $LHV_{ar,D}$ of thermally dried digestate was 12 MJ/kg, while the $LHV_{ar,W}$ of wood was 9.6 MJ/kg.
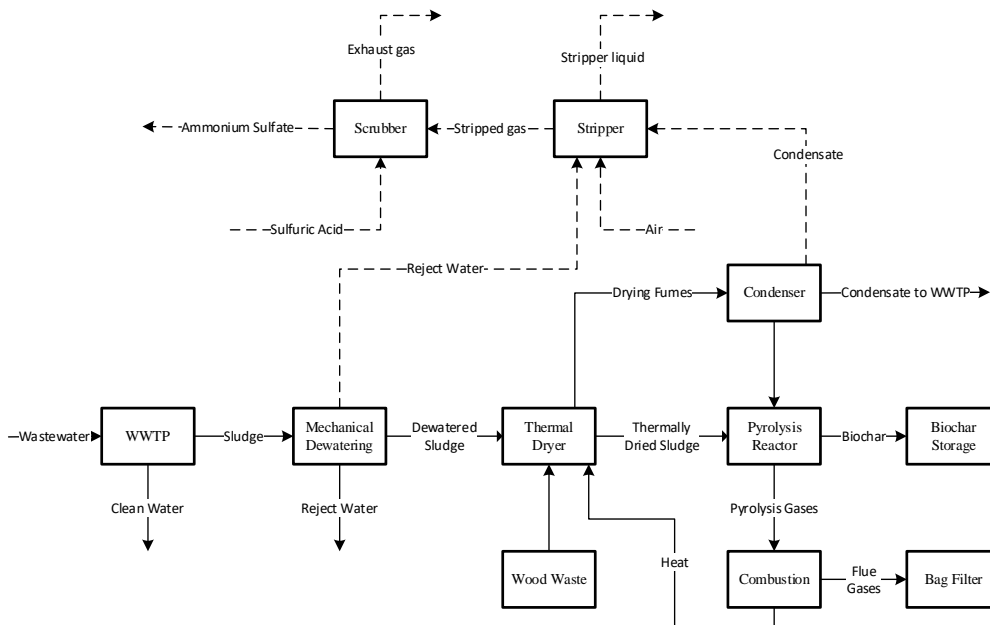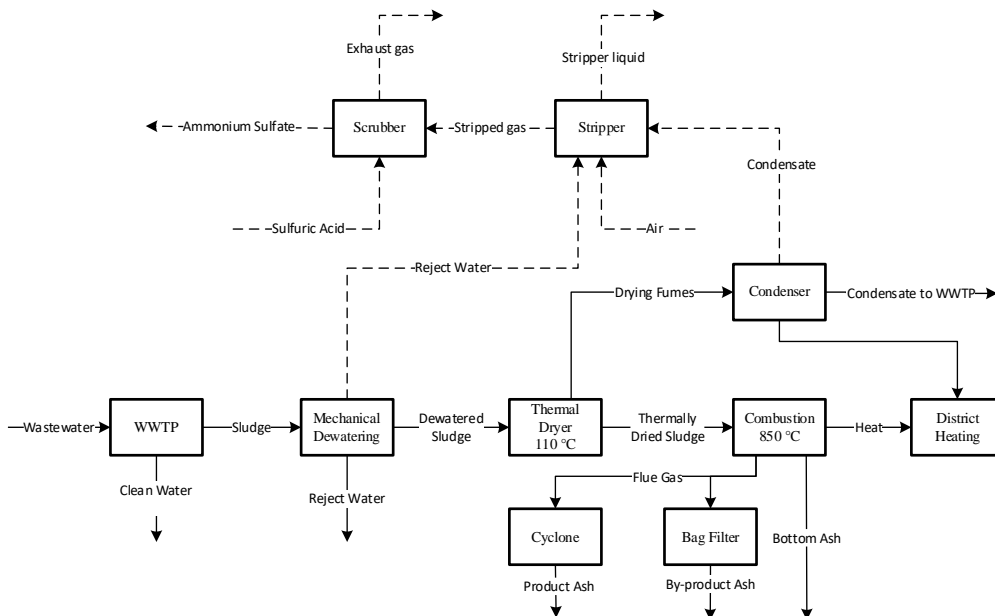
Figure 4. Block diagram of the combustion process. Solid lines denote the current process and dotted lines show the possibilities to recover nitrogen from the reject water and thermal drying fumes.

### 3.4.6 Nutrient recovery

In order to recover 780 t/a of nitrogen from the reject water in both the pyrolysis and combustion scenarios, the addition of a stripper and scrubber to the systems was implemented. Moreover, according to Horttanainen et al. (2017), it is assumed that 12% of the total nitrogen in the dewatered sludge is released in the thermal drying vapour as $NH_3$. Due to the high moisture content of the drying vapour, the fertilizer would be diluted. To prevent this, a condenser is added to the process prior to the stripper–scrubber combination so that the drying fumes first pass through the condenser; the condensate is then diverted to the stripper–scrubber combination along with the reject water. The parameters used for stripping and scrubbing in Publication II, III and IV, are presented in Table 4 in section 3.3.5 in more detail.

### 3.4.7 Revenue and profit estimation

The cost estimation takes into account the prices of sewage sludge-derived biochar and district heat in Finland. The investment costs for the pyrolysis scenario are calculated

based on sewage sludge-derived biochar values, while district heat prices are used to calculate the revenue.

The general cost estimation was based on the biochar derived from the sewage sludge and the district heat cost in Finland. The value of 400 €/t (VVY, 2019) was used in the calculation for the cost of biochar. Due to differences in regional transmission and supply and demand, district heat prices fluctuate significantly. Therefore, the average of all values was considered when calculating district heating revenue. All rates for district heating exclude value-added taxes (VAT).

## 3.5   Nitrogen recovery from sewage sludge and side streams

Publication III and Publication IV followed the four LCA steps: goal and scope definition, LCI, LCIA, and interpretation of results. The selected LCA method for impact assessment was ReCiPe 2016 v1.1 (midpoint hierarchical timeframe) with long-term impacts. The selected mid-point impact categories for Publication III included climate change due to its global importance, focusing on: particulate emissions and acidification, which are essential when energy production is concerned; eutrophication (as nitrogen and phosphorus flows are significant in biological treatment); mineral scarcity (to consider the depletion of these reserves); and terrestrial and human toxicity (to estimate the potential impact of heavy metals). However, in Publication IV, the global warming potential, terrestrial acidification, marine water eutrophication, and freshwater eutrophication were selected as impact categories based on a literature review (Matuštík et al., 2020). The EASETECH program was used in Publication III, whereas GaBi 10.5.1.124 was used in Publication IV.

### 3.5.1   Goal and scope

The environmental performance of different treatment technologies for dewatered sewage sludge digestate from a municipal WWTP (Publication III) and nitrogen recovery from reject water of sewage sludge dewatering and condensate from thermal drying of sewage sludge (Publication IV) were demonstrated using LCA methodology based on ISO standards 14040 and 14044.

One metric ton (t) of digestate from dewatered sewage sludge was used as the functional unit. The focus was on the long-term effects of a decision about the current sewage sludge digestate treatment; hence consequential modeling was employed. Publication IV aimed to assess the environmental performance of nitrogen recovery from liquid waste streams of sewage sludge treatment in a municipal WWTP. The functional unit was 870 kt/a reject water and 45 kt/a condensate. Publication III and Publication IV on the Viikinmaki WWTP of the Helsinki Region Environmental Services Authority (HSY) serve as the basis for the functional unit.

Both Publication III and Publication IV consider Finland's geographical scope and provide specific data and parameters for each treatment scenario, including energy and material demands, nitrogen recovery efficiencies, and transport distances. The collected data for the life cycle inventory came mainly from existing plants or pilot-scale operations, and the reference year for the study was 2020.

The life cycle impact assessment was conducted to evaluate the selected impact categories. A contribution analysis was performed to identify the main contributing processes, and a sensitivity analysis was used to identify the most sensitive parameters. A high and low-performance sensitivity analysis determined the range of the net results. Overall, the materials and methods section detailed the research design, data sources, and analytical techniques used to assess the environmental performance of nitrogen recovery from sewage sludge treatment in different scenarios.

### 3.5.2 System Boundary

Publication III evaluated three scenarios for treating dewatered sewage sludge: windrow composting, fluidized bed combustion, and pyrolysis. Each scenario involves different processes and technologies for sludge treatment and utilization. The focus was on the long-term effects of a decision about the current sewage sludge digestate treatment; hence consequential modeling was employed. On the other hand, Publication IV analyzed three scenarios: conventional WWTP, stripping and scrubbing, and adsorption on biochar. In Publication III, the LCI provides detailed information on the composition of dewatered sewage sludge digestate, wood waste used in pyrolysis, and other materials and processes involved in each scenario. In Publication IV, the LCI provided information about the properties of reject water of sewage sludge dewatering and condensate from sewage sludge's thermal drying, WWTP parameters, adsorption and stripping and scrubbing process.

Figure 5 and Figure 6 illustrate the system boundaries for Publication III and Publication IV, respectively. The following three scenarios were taken into consideration for Publication III. The wood waste used for pyrolysis in scenario 3 was directed to a boiler for district heat production in scenarios 1 and 2.

1. Composting in windrows, followed by manufacturing of compost soil and compost use on arable land (PIII-S1).
2. Combustion, followed by using the heat as district heating and the ash as a fertilizer for forests (PIII-S2). Condensate from the thermal drying fumes was directed to:
a) WWTP (PIII-2.1)
b) Nitrogen recovery (PIII-2.2)
3. Pyrolysis with wood and the use of biochar from pyrolysis on arable land (PIII-S3).

On the other hand, in Publication IV, the following scenarios were considered:

1.  Scenario PIV-S1 (WWTP) includes the conventional treatment of reject water and condensate in a municipal WWTP; consequently, nitrogen is not recovered but is primarily removed and released into the atmosphere as $N_2$ via nitrification/denitrification.
2.  In Scenario PIV-S2 (stripping and scrubbing), nitrogen is recovered through a combination of air stripping and scrubbing with acidic media.
3.  Scenario PIV-S3 (adsorption) investigates nitrogen recovery from reject water and condensate via ammonia adsorption on biochar derived from sewage sludge and wood pyrolysis instead of air stripping. Biochar infused with ammonia is applied to land for soil improvement and carbon sequestration, replacing nitrogen fertilizers from fossil fuels.
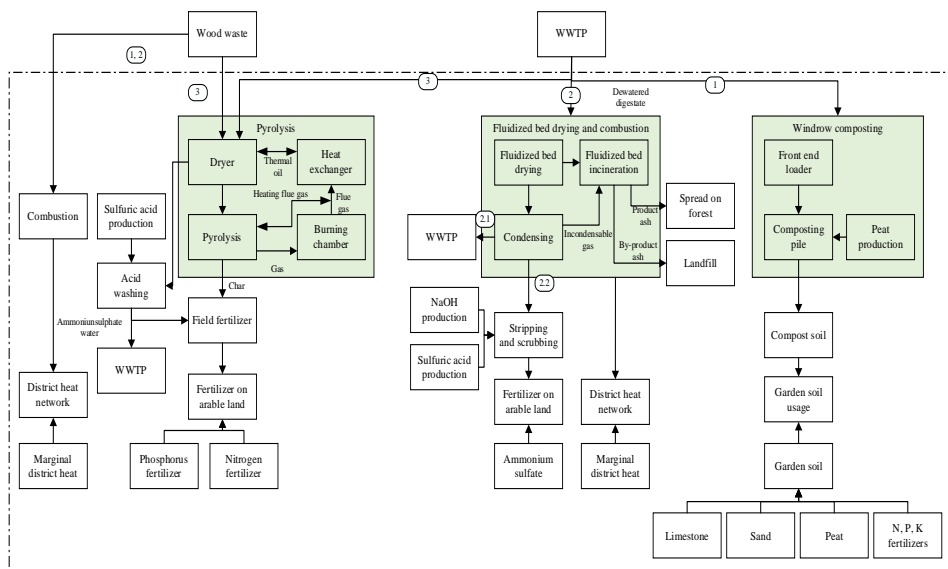
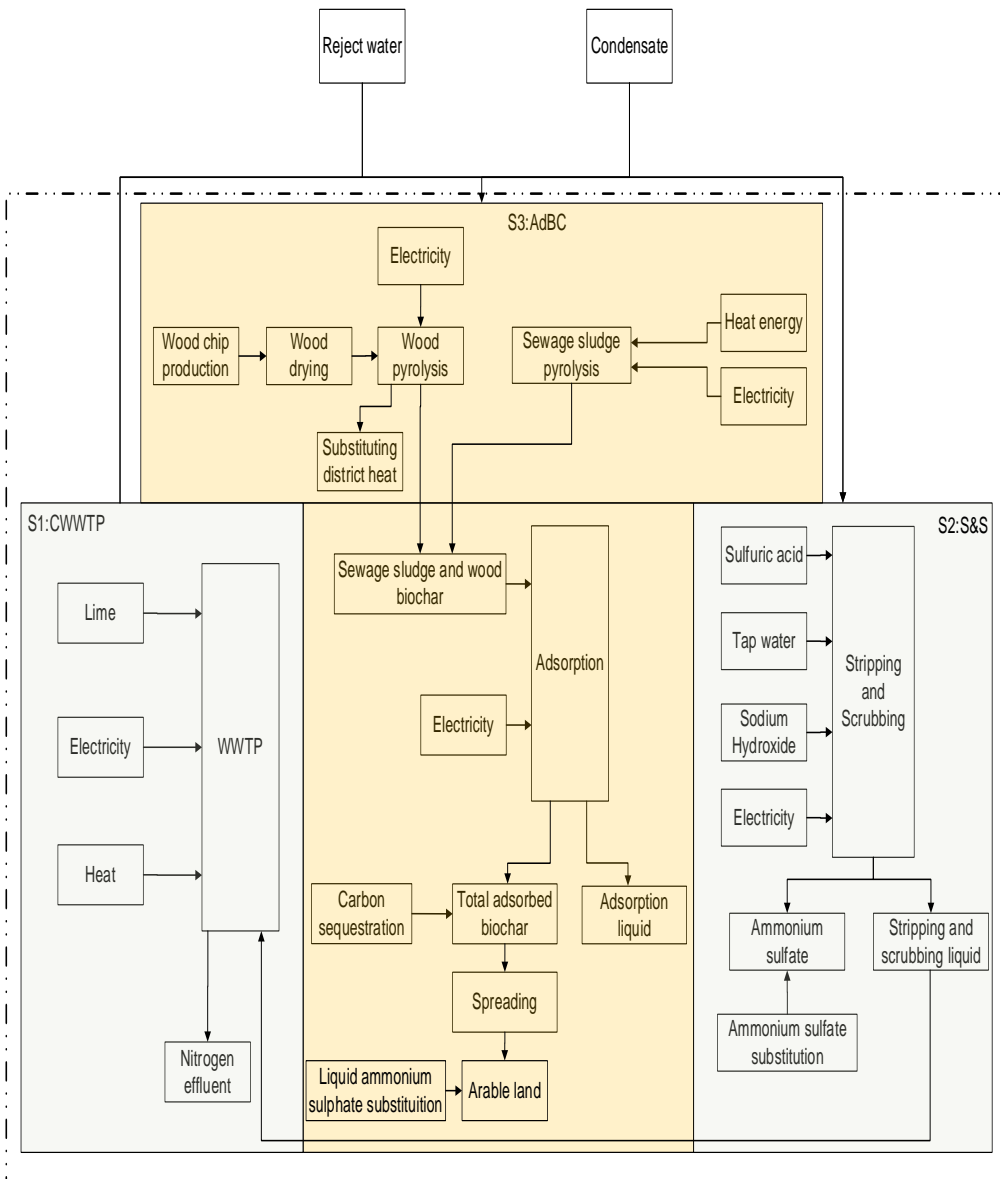

Figure 5. System boundary for Publication III.

Figure 6. System boundary for Publication IV.

### 3.5.3 Life Cycle Inventory

### 3.5.3.1    Utilizing dewatered sewage sludge digestate

### 3.5.3.2    Composting

Composting sewage sludge digestate in a windrow is the standard operating procedure (PIII-S1). The dewatered sewage sludge digestate windrow composting method used in the Metsäpirtti composting area managed by HSY was the basis for the PIII-S1 data (Mölsä, 2020). Peat was used as a support material in the composting process mechanical turning windrows using a front-end loader powered by light fuel oil (LFO) aerates the composting pile (Gareis, 2020). Peat was beneficial in the composting due to its small particle size, eliminating waste screening and lowering high pH levels in the sludge compost. It is also essential for producing soil products.

The sludge is stored in a silo to maintain a steady fuel input. In the dryer, hot sand is mixed with the sludge, evaporating the water and resulting in a dry mix. The dryer operates at 110 °C, utilizing heat from the transferred sand and flue gases passing through a heat exchanger. Water vapor is partially recirculated for fluidization, while the rest goes to a scrubber-type condenser. Odorous gases are separated from the condensed water and injected back into the reactor, minimizing odour issues. The dry sludge and sand mix is then combusted in the reactor at 850 °C, ensuring complete destruction of organic compounds. Ash is collected through a hot cyclone and bag filter. A wet scrubber removes sulfur from the flue gas, releasing purified gas through a stack. Excess heat can be extracted for other purposes, and the nutrient-rich product ash can be used as fertilizer. Drying fumes are sent to a condenser, with the condensate containing recoverable nitrogen that can be converted into ammonium fertilizers using air stripping and scrubbing techniques. The efficiency of nitrogen recovery depends on the pH and temperature.

### 3.5.3.3    Pyrolysis of sewage sludge

In Publication III, dewatered digestate was sent to a pyrolysis process. The information was primarily gathered from laboratory testing done to create the pyrolysis pilot plant for HSY (Huber, 2020; Kainulainen, 2020).

To make the pyrolysis process energy self-sufficient and to raise the biochar's carbon content, wood waste was combined with sewage sludge. The feedstock first goes through a thermal dryer, where most moisture is removed, preparing it for processing in the pyrolysis reactor. Wet digestate will produce steam if it is pyrolyzed. Due to the condensation of moisture, the final product will become more diluted, and there will be a rise in the number of non-condensable gases (Syed-Hassan et al., 2017). The pyrolysis reactor is intended to function between 450 and 650 °C. The pyrolysis gas that leaves the reactor and may contain tar is directed toward a combustion chamber. The thermal drier

and pyrolysis reactor receive the heat that was recovered from the gas. At the point where exhaust gases escape, bag filters are used to filter the exhaust gas and collect impurities (HSY, 2021).

Thermal drying fumes are directed to a scrubber to recover nitrogen and remove odours. In order to absorb ammonia and create ammonium sulfate liquid, liquid acid is either sprayed or circulated through a packed column while nitrogen-containing air is injected from the bottom of the scrubber. The liquid-to-gas ratio, pH, temperature, and ammonium ion concentration all significantly impact the final product's quality in scrubbing with acidic media. To maximize the nitrogen recovery from the reaction between ammonia gas and sulfuric acid, the equilibrium must change in favour of ammonium (Publication I). A pH range between 1 and 5 is crucial in achieving this equilibrium. Similarly, a good liquid-to-gas ratio (L/G) improves the ammonia recovery effectiveness. For countercurrent packed-bed scrubbers, the liquid flow rate must be such that it completely moistens the packing while avoiding flooding in the column. It typically exceeds the minimum flow rate by 11 to 13 times (Schnell et al., 2020). $2.4 \times 10^4$ is the recommended L/G ratio for spray scrubbers.

### 3.5.3.4 Reject water and condensate

In the baseline configuration of Publication IV, scenario PIV-S1 (CWWTP), reject water was recirculated back into the wastewater treatment process for nitrogen removal via nitrification/denitrification. After dewatering, the sewage sludge digestate undergoes thermal drying to reduce its water content. Up to 12% of the total nitrogen ($N_{tot}$) in the digestate is released in the drying vapour (Horttanainen et al., 2017), and another ammonium-rich liquid stream, condensate, is formed after condensation. Significant amounts of nitrogen can be recovered and recycled by combining these streams. Table 9 lists the characteristics of the reject water and the condensate.

Table 9. Properties of reject water and condensate

| **Reject Water** | | | |
|---|---|---|---|
| **Parameter** | **Value** | **Units** | **References** |
| $N_{tot}$ | 1 | kg/t | (HSY, 2018) |
| $NH_4$ | 0.8 | kg/t | |
| $NH_4$-N | 0.62 | kg/t | |
| **Condensate** | | | |
| **Parameter** | **Value** | **Units** | **References** |
| $N_{tot}$ | 0.09 | kg/t | (Deviatkin et al., 2018) |
| $NH_4$-N | 0.09 | kg/t | |

### 3.5.3.5 Wastewater treatment plant (WWTP)

The parameters considered in WWTP for Publication IV are listed in Table 10.

Table 10. WWTP properties for Publication IV.

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Electricity consumption | 1.52 | MJ/t of water | (HSY, 2018) |
| Heat consumption | 1.33 | MJ/t of water | |
| Lime consumption | 0.030 | kg/t of water | |
| N removal efficiency | 85 | % | |

Biogas was used to generate the necessary heat for the WWTP as well as 64% of the electricity that the WWTP needs. The remaining electricity is supposed to come from either the Finnish grid mix or clean energy sources like wind power. Finland produces a significant amount of renewable electricity (47%) and electricity with low emissions (34.7%), while less than 20% of the nation's total energy is derived from fossil fuels. Most of the imported electricity (18%) comes from Sweden, which produces less pollution than Finland's entire energy production mix. Only 8% of the imported power is from Russia, but due to Russian electricity's high emissions intensity, it accounts for 25% of all emissions (Rossi et al., 2018). It was decided to use wind power in Finland as an example of renewable energy in the sensitivity analysis because it has a lower emission factor than other sources (Zhaurova et al., 2023).

### 3.5.3.6 Stripping and Scrubbing

In the stripping procedure, air was used to separate gaseous $NH_3$ from the liquid in the column. The efficiency of the stripping column was assumed as 95% (Vaneeckhaute et al., 2014) and the same for the scrubbing column (Melse and Ogink, 2005). The stripping gas was then directed to a scrubber using sulfuric acid to capture ammonia and produce ammonium sulfate (Publication II). The scrubber's exhaust gas was directed to combustion to eliminate any odorous compounds. The scrubbers introduce nitrogen-containing air from the bottom and spray or flow liquid acid through a packed column to absorb ammonia and make ammonium sulfate liquid. The scrubbing performance depends on the liquid-to-gas ratio, pH, temperature, and ammonium ion concentration. Publication I determined that the equilibrium must shift toward ammonium to maximize the nitrogen recovery from the ammonia gas and sulfuric acid. This equilibrium requires a pH range of 1–5. An appropriate liquid-to-gas ratio (L/G) also improves the ammonia recovery. The L/G ratio for countercurrent packed-bed scrubbers depends on the liquid flow rate to completely make the packing wet but without flooding the column (Schnelle and Brown, 2016). Table 11 shows the spray scrubber electricity demands and recovery

efficiency for an L/G ratio of $2.4 \times 10^{-4}$, which was used in Publication III. Table 11 also summarizes the inventory information for the stripping and scrubbing processes for Publication III and Publication IV.

Table 11. Parameters for stripping and scrubbing for Publication III and Publication IV.

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Electricity use | 0.028 | MJ/kg | (Vaneeckhaute et al., 2014) |
| Heat use | 0.188 | MJ/kg | (Ervasti et al., 2018; Vaneeckhaute et al., 2014) |
| $H_2SO_4$ use | 3.5 | kg $H_2SO_4$/kg $NH_4$-N | (Batstone et al., 2015) |
| NaOH use | 3.3 | kg NaOH/kg $NH_4$-N | (Endev, 2020) |
| Water use | 2.1 | kg water/kg $NH_4$-N | Calculated |
| Stripper–Scrubber efficiency | 95 | % | (Hadlocon et al., 2015; Melse and Ogink, 2005) |
| **Transport** | | | |
| Biochar | 43 | km | (Mölsä, 2020) |
| Fertilizer | 43 | km | (Statistics Finland, 2017b) |
| $H_2SO_4$ | 201 | km | (Statistics Finland, 2017b) |

### 3.5.3.7 Adsorption on biochar

The produced sewage sludge biochar was calculated in Publication II, and this data was also used in Publication IV. Electricity and heat demand for sewage sludge biochar production was based on the data from HSY, which was further used to calculate the electricity and heat required per kg of biochar. Similarly, the electricity and heat demand for wood biochar production was based on the values from (Leppäkoski et al., 2021).

In Publication IV, wood and sewage sludge-based biochar was used since the amount of biochar produced alone was insufficient for the adsorbing ammonia. Since the amount of biochar produced by sewage sludge alone was insufficient, wood needed to be pyrolyzed in order to meet the adsorbent requirement. The wood must be chipped and dried before pyrolysis, and the pyrolysis reactor needs energy. Electricity and heat (steam) from biomass and natural gas are needed to dry the wood. In the production of wood biochar, extra heat from non-condensable pyrolysis gas was used for district heating and is currently replacing the typical Finnish district heating mix (Leppäkoski et al., 2021). The

condensate and reject water streams were fed into a biochar-filled adsorption column to perform ammonia adsorption. Additional information was gathered from Leppäkoski et al. (2021). The parameters for calculating the pyrolysis process are listed in Table 12.

Table 12. Parameters of the scenario S3 (AdBC) of Publication IV.

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Sewage sludge biochar | | | |
| Mass of sewage sludge | 65,000 | t | Publication I |
| Mass of SS biochar | 12,000 | t | Publication I |
| Nitrogen adsorption capacity | 0.004 | kg N-NH$_4$+/kg biochar | (Tang et al., 2019; Yin et al., 2019; Zhang et al., 2020) |
| Electricity demand, SS biochar production | 0.827 | MJ/kg biochar | (HSY, 2018) |
| Heat demand, SS biochar production | 0.003 | MJ/kg biochar | (HSY, 2018) |
| Electricity demand, wood biochar production | 0.750 | MJ/kg biochar | (HSY, 2018) |
| Heat demand, wood drying | 0.003 | MJ/kg biochar | (HSY, 2018) |
| SO$_2$ removal | 0.021 | kg CO$_2$ eq./kg biochar | Publication III |
| Carbon share in biochar | 34 | % | (Leppäkoski et al., 2021) |
| Biochar nitrogen usability | 64 | % | (Puga et al., 2020) |
| Carbon footprint biogenic | 0.45 | kg CO$_2$ eq. /kg CO$_2$ | (Leppäkoski et al., 2021) |
| Wood biochar | | | |
| Nitrogen adsorption capacity | 0.005 | kg N-NH$_4$+/kg biochar | (Hailegnaw et al., 2019; Paramashivam et al., 2016; Yang et al., 2017) |
| Mass of wood biochar | 97,000 | t | Calculated |
| Yield of wood biochar | 0.34 | kg biochar/kg dry wood | (Abo-State et al., 2014) |
| Moisture (dry wood) | 10 | % | (Leppäkoski et al., 2021) |
| Moisture (wet wood) | 28 | % | (Leppäkoski et al., 2021) |
| Mass of wood | 280,000 | t | Calculated |
| Wood drying electricity demand | 0.252 | MJ/kg removed water | (Leppäkoski et al., 2021) |
| Wood drying heat demand | 4.504 | MJ/kg removed water | (Leppäkoski et al., 2021) |
| Wood processing emissions | 0.018 | kg CO$_2$/kg wood | (Havukainen et al., 2018) |
| Excess heat production | 4.9 | MJ/kg dry wood | (Leppäkoski et al., 2021) |
| Carbon content of wood biochar | 34 | % | (Leppäkoski et al., 2021) |
| C share remaining in soil | 68 | % | (Leppäkoski et al., 2021) |

| | | | |
|---|---|---|---|
| CF (carbon footprint) biochar land application | 0.45 | kg $CO_2$ eq./kg $CO_2$ | (Cherubini et al., 2016) |
| CF wood pyrolysis gas combustion | 0.45 | kg $CO_2$ eq./kg $CO_2$ | (Cherubini et al., 2016) |

# 4 Results and discussion

## 4.1 Recovery of nitrogen from drying fumes

The results of the Aspen plus modeling of scrubbing ammonia from sewage sludge drying fumes for Publication I are summarized in Figure 7. The results are compared with the reference models by Khakharia et al., (2014) and Melse and Ogink (2005) for validation. In Figure 7a, the designed model performed similarly to the referenced model. In the current investigation, the ammonia gas inflow concentration varied between 152–155 mg/m$^3$ (218–223 ppm), while the outflow gas concentration was calculated as 2 mg/m$^3$ (3 ppm). By keeping the liquid flow rate at 70 L/min, temperature 100°C and gas flow rate at 1,000 m$^3$/h constant, the pH showed no effect on the efficiency of the scrubber at pH 1-3, whereas by increasing the pH to 4 and 5, the efficiency decreased to 95% from 99% for both selected concentrations of 75 and 100 ppm as shown in Figure 7b and Figure 7c. Similarly, the L/G ratio showed no significant effect on the efficiency of the scrubber at a flow rate which varied between 50 and 130 L/min, while keeping pH at 3, gas flow rate at 1 000 m$^3$/h, and the temperature at 100 °C as shown in Figure 7d. The temperature did not affect the efficiency of the scrubber. The presence of $NH_4^+$ and $SO_4^{2-}$ ions were also analyzed with the acid flow rates. Even though the concentration of ammonia in the drying fumes was low, still 0.06 kg/h of $NH_4^+$ and 0.43 kg/h $SO_4^{2-}$ was found, as Figure 8 shows. The high mass flow rate of $SO_4^{2-}$ also highlights that $NH_4^+$ ions can be fully captured to produce ammonium sulfate.
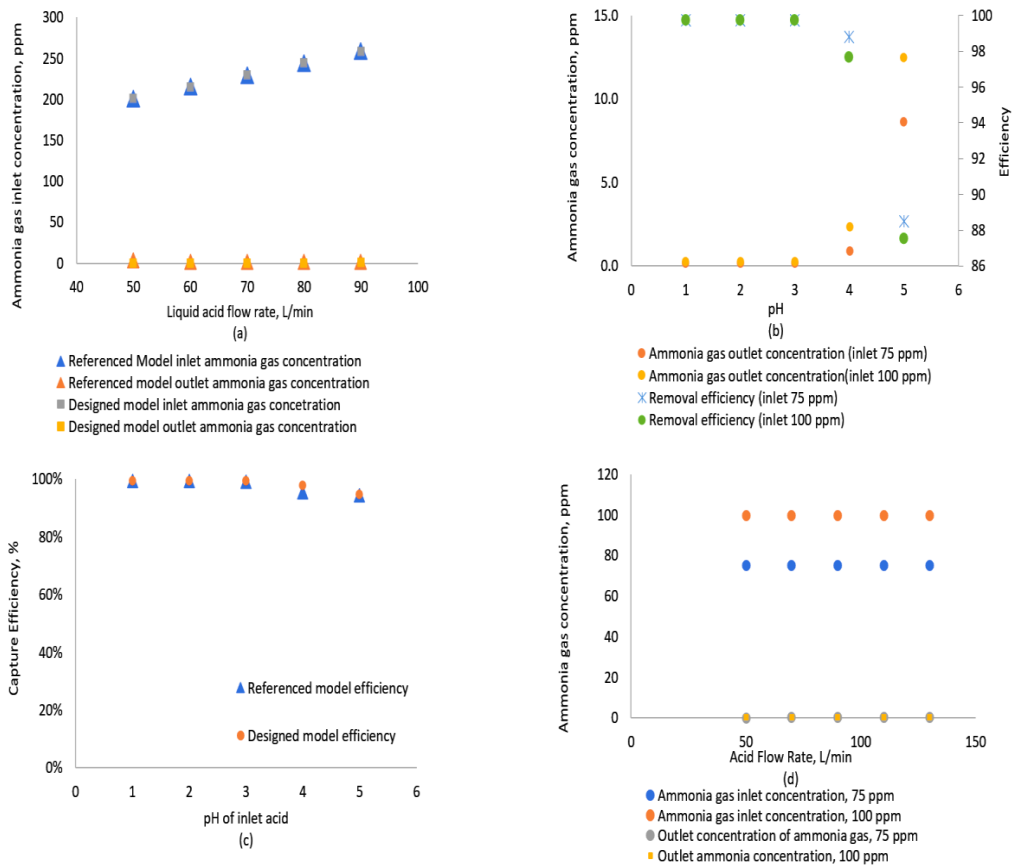
Figure 7. Results of the validation of the designed model with the referenced model with (a) the liquid flow rate and change in ammonia gas concentration, (b) the effect of the pH on the ammonia removal efficiency, (c) the pH of the inlet acid and the ammonia capture efficiency, (d) changes in the ammonia gas concentration with changes in the acid flow rate.
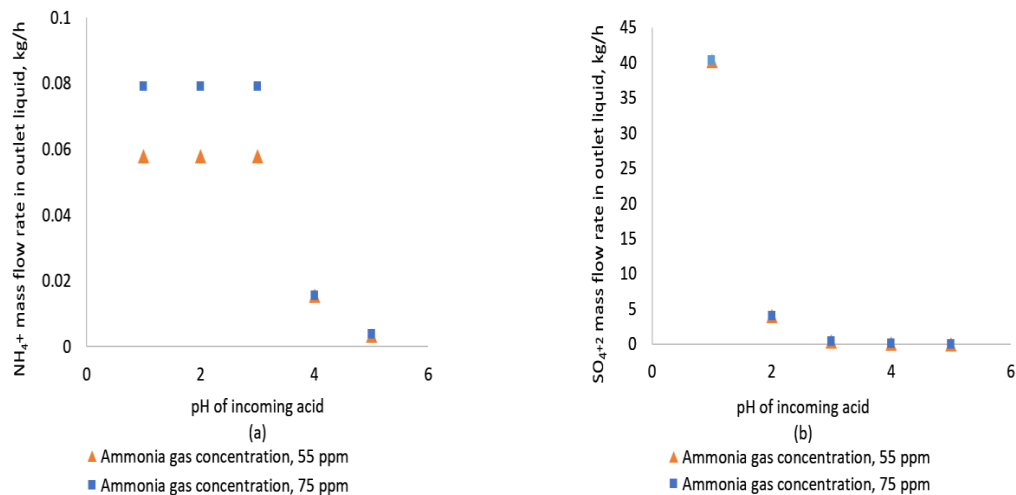
Figure 8. The mass flow rate at different pH levels of (a) $NH_4^+$, and (b) $SO_4^{-2}$ ions in liquid ammonium sulfate outlet stream.

### 4.1.1 Comparison of revenues and costs

The Aspen modeling results were utilized to scale up the scrubbing device for the WWTP at Lappeenranta in Finland. The Toikansuo wastewater treatment plant in Lappeenranta treats 16,000 $m^3$/day for 72,000 people, and the scrubbers were scaled up based on their sewage sludge production rate. 7,700 t/a of sludge had 20% solids, 5.3% nitrogen, and 12% ammonia (Horttanainen et al., 2017). One scrubber can handle 8,610 kg N/a at 75 ppm with a flow rate of 24,000 $m^3$/h. The sizing of the scrubber is presented in Table 13. Table 14 then presents the scrubber material and investment costs while Table 15 presents the estimations on revenues.

Table 13. Equipment sizing of designed scrubber

| Specifications | Values |
|---|---|
| Diameter of scrubber | 2.4 m |
| Height of scrubber | 12 m |
| Packing height | 6 m |
| Required number of scrubbers | 1 |

Table 14. Estimation of scrubber material and packing material with the installation cost

| Equipment | Material | Cost (k€) |
|---|---|---|
| Scrubber | Stainless steel | 78 |
| Packing (Sulzer Mellapakplus 252Y) | SS 304L | 21 |
| Erection and commissioning cost | | 165 |
| Start-up cost | | 26 |
| **Total** | | **290** |

Building a scrubber that can treat 24,000 $m^3$/h gas and has a liquid flow rate of 575 L/min has a total investment cost of 290 k€. Sulfuric acid and water account for 14 and 99 k€/a of the operational expense, respectively. The scrubber may be operated by the sludge treatment plant staff; therefore, it does not increase the personnel expenditure.

Table 15 shows the cost of the treated sludge and potential fertilizer income from recovered nitrogen. The mineral fertilizers production cost is 1.6 €/kg-N (Havukainen et al., 2018) and with the used ammonia removal efficiency of 99%, a total of 8,610 kg N-tot/a nitrogen is recovered. The ammonium sulfate production is 9 t/a and the ammonia recovery cost is 18 €/ton. At 10% interest assuming a 10-year scrubber lifespan, the total annual cost is 155 k€. The low concentration of the ammonia gas in the drying fumes is the main reason for the high cost and low production of ammonium sulfate.

Table 15. Estimation of revenues, operational cost, and annual cost.

| Recovery | Value |
|---|---|
| Income from recovered nitrogen | 14 k€/a |
| Total annual cost | 155 k€/a |
| Additional annual cost for sludge treatment | 20 €/t of sludge |
| Cost of ammonia recovery | 18 €/t of ammonium sulfate |
| Cost of ammonia recovery | 18 €/kg of nitrogen |

The relation of water costs to the L/G ratio and recycling rate of the liquid is presented in Figure 9. First, the L/G ratio was altered from 1.5 to 4.3, and the recycling rate remained at 50%. Then, the L/G ratio was kept constant at 1.5, and the recycling rate varied from 50–70 %. A flooding limit of 80% caused simulation inaccuracies over the 70% recycling rate. The values marked with the ellipse show that 70% recycling and 1.5 L/G ratio yield excellent outcomes with decreased water costs. In this case, the total operational cost, including water and sulfuric acid, is 113 k€/a.
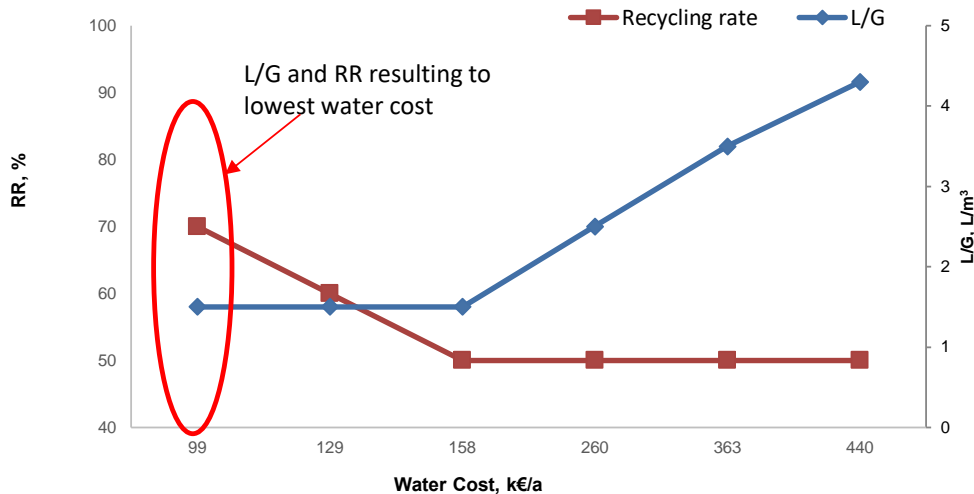
Figure 9. The relation of the water costs to the recycling rate (RR) and the liquid-to-gas ratio (L/G).

## 4.2 Recovery of nitrogen from sewage sludge digestate

The nitrogen recovery from sewage sludge digestate with the integration of pyrolysis and combustion was studied in Publication II. The results of Publication I, such as the L/G and pH are used in Publication II for comparison and to obtain a better economic analysis. Figures 10–15 show the outcomes of the pyrolysis and combustion scenarios with and without nitrogen recovery from the reject water and drying fumes. The base scenarios PII-S1.0 (pyrolysis without N recovery) and PII-S2.0 (combustion without N recovery) are shown in Figures 11 and 12, respectively. It is obvious that nitrogen may be collected from the drying fumes and reject water in both cases.

Figure 10 shows that in the pyrolysis process, 12,000 t/a of biochar is produced, containing approximately 150 t/a of nitrogen. On the other hand, in the case of installing a scrubber, 150 t/a of AS fertilizer can be generated in the scrubber as part of the pyrolysis process. The biochar also contains 500 t/a of phosphorus, making it commercially valuable as a soil amendment and fertilizer due to its nutrient content. The gases produced during pyrolysis are used for heating in the main plant equipment, such as the pyrolysis reactor and dryer. On the other hand, in the combustion scenario PII-S2.0 (Figure 11), sewage sludge combustion generates 120,000 MWh/a of heat, which is suitable for district heating. However, 400 t/a of nitrogen in the thermally dried sludge is combusted and cannot be utilized. Phosphorus, on the other hand, ends up in the ash from sewage sludge combustion, with nearly 95% found in the product ash and the remaining 5% in the non-utilizable by-product ash (Endev, 2020).
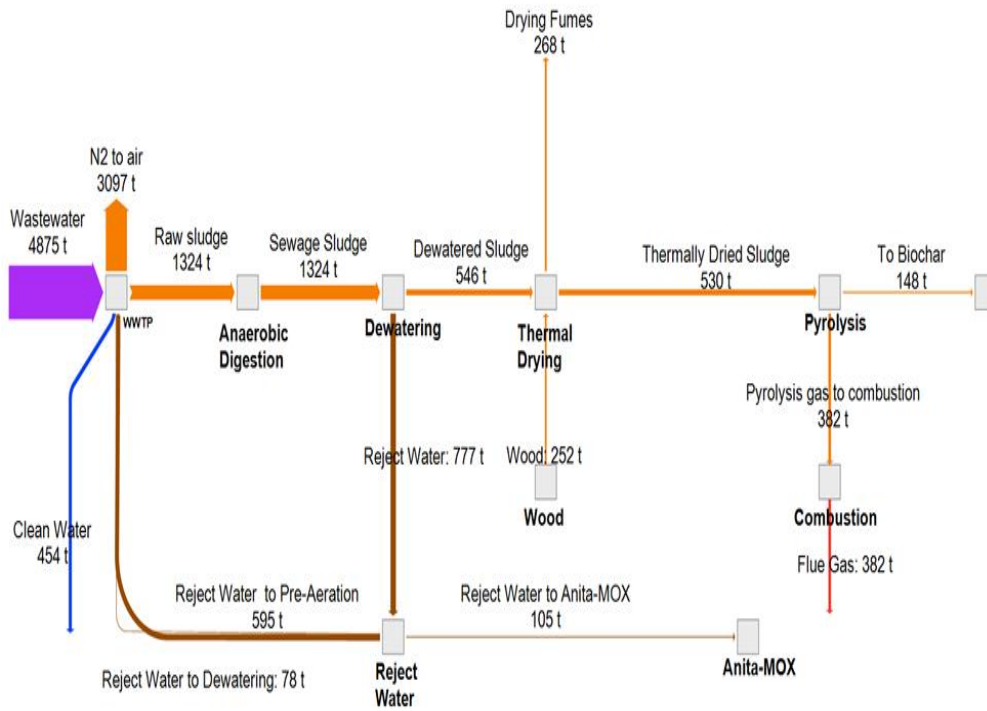
Figure 10. the nitrogen balance for PII-S1.0 (pyrolysis base scenario, no N recovery).
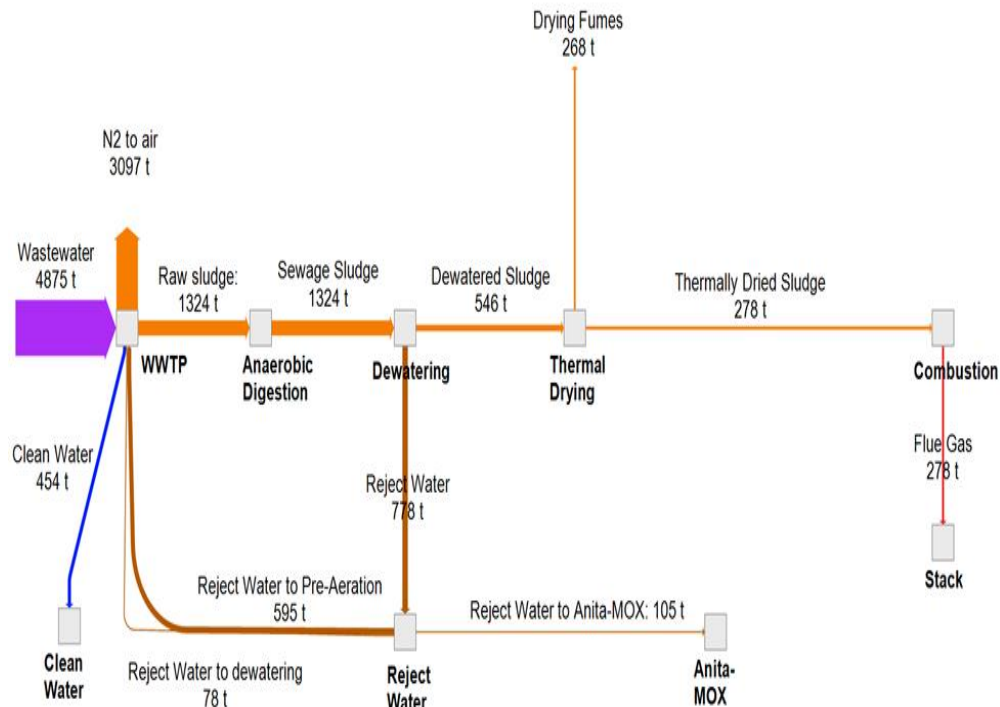
Figure 11. The nitrogen balance for PII-S2.0 (combustion base scenario, no N recovery).

Figures 12 and 13 depict the pyrolysis scenario PII-S1.1 and combustion scenario PII-S2.1, where nitrogen is recovered from drying fumes but not from reject water. The mass balance calculations reveal that in the WWTP process, approximately 73% of the total nitrogen is lost, with 3,100 t/a released as $N_2$ into the atmosphere and 450 t/a as nitrate and ammonium ions into clean water. About 11% (550 t/a) of the nitrogen remains in the dewatered digestate, while the remaining 16% (780 t/a) is mainly present in the reject water and can be recovered. Additionally, 3% of the nitrogen can be recovered from drying fumes containing 160 t/a of nitrogen suitable for ammonium sulfate fertilizer production. The addition of a condenser in the system served two purposes: removing moisture from non-condensable odorous gases entering the combustion process and recovering nitrogen from condensed water. Assuming a scrubber efficiency of 95%, approximately 160 t/a of nitrogen can be captured for use as fertilizer. The odorous gases are directed to combustion.
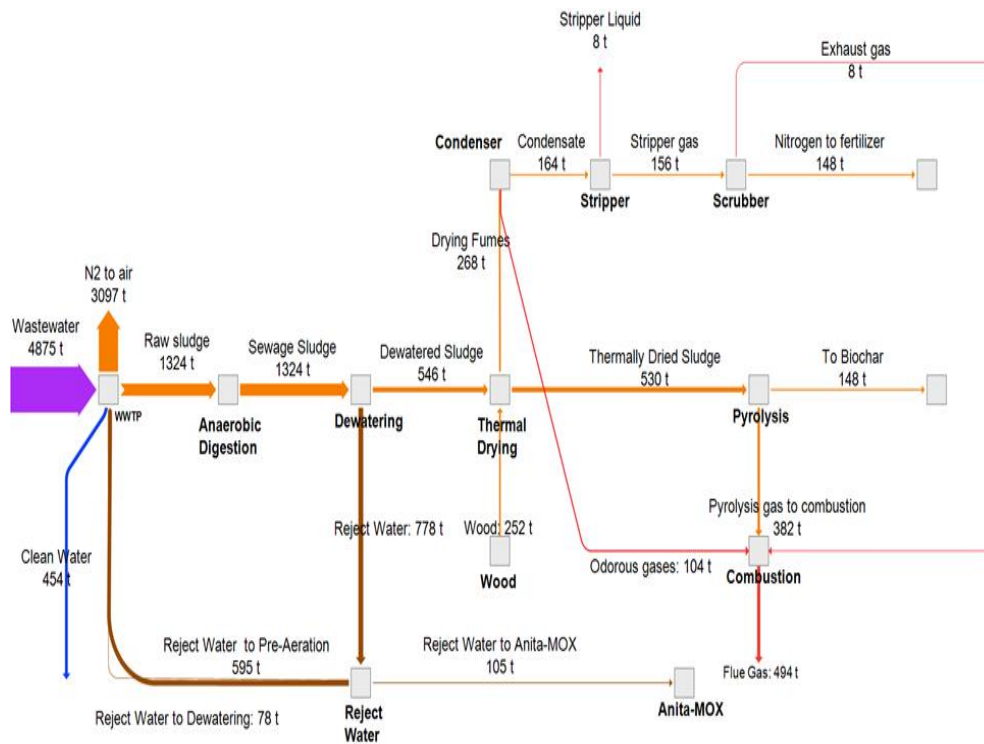
Figure 12. The nitrogen balance for PII-S1.1 (pyrolysis with N recovery from drying fumes).
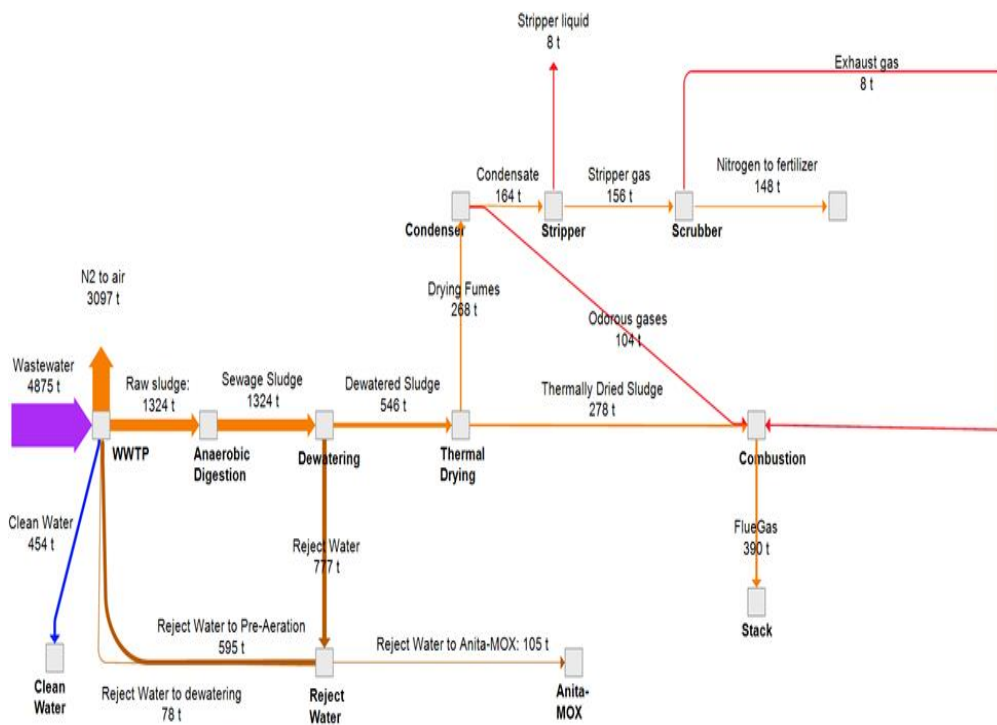
Figure 13.The nitrogen balance for PII-S2.1 (combustion with N recovery from drying fumes).

### 4.2.1 Recovery of nitrogen from drying fumes and reject water

Combining the recovery from the thermal drying fumes with recovery from the digestate dewatering reject water, which includes most of the recoverable nitrogen, can boost the overall practicality of nitrogen recovery. Figures 14 and 15 indicate that 780 t/a of nitrogen in the WWTP reject water is accessible for recovery and further use as ammonium fertilizer. This significantly increases the 268 t/a of nitrogen recoverable from the drying fumes.
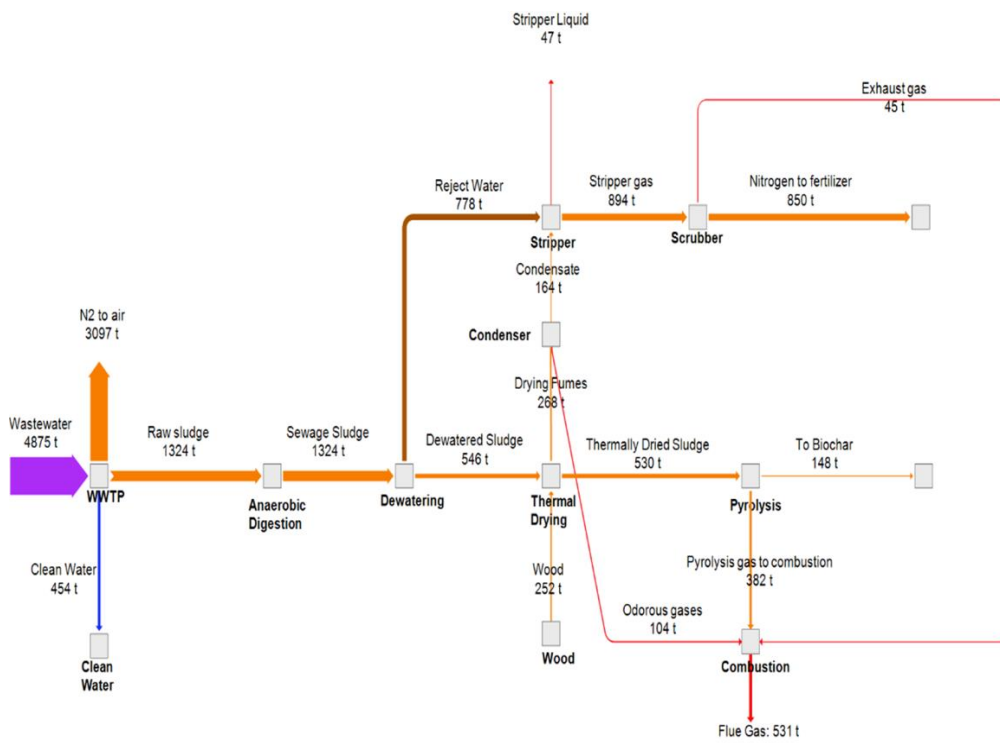
Figure 14. The nitrogen balance for PII-S1.2 (pyrolysis with N recovery from drying fumes and reject water).
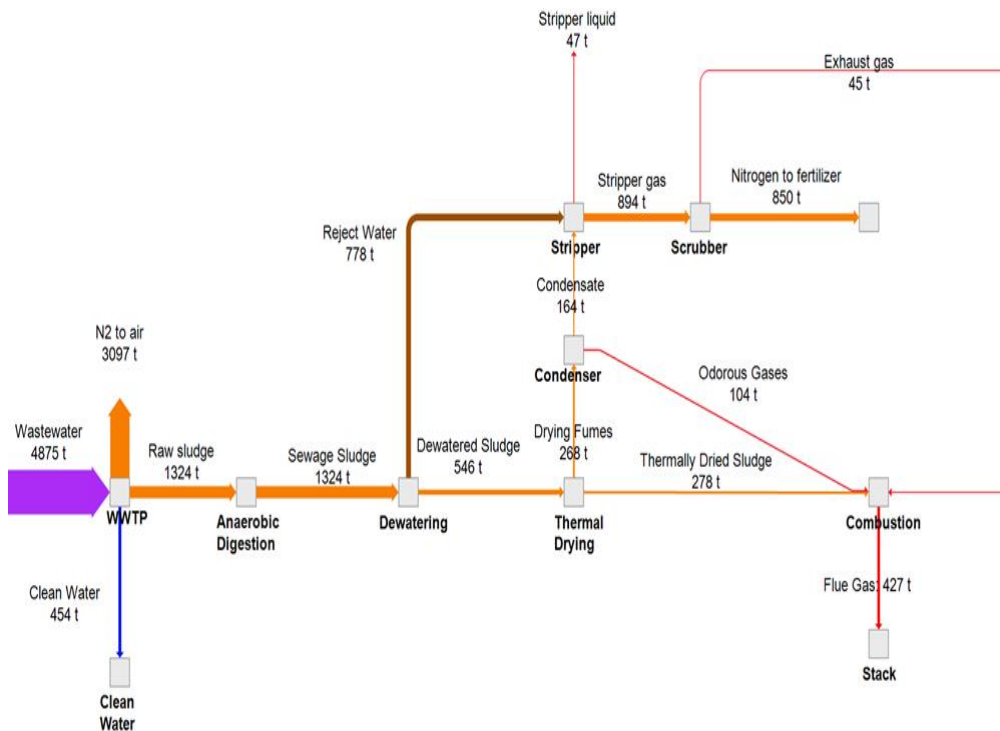
Figure 15. The nitrogen balance for PII-S2.2 (combustion with N recovery from drying (combustion with N recovery from drying fumes and reject water).

### 4.2.2 Comparison of mass and energy balances

The only variation between the mass balances of the pyrolysis and combustion scenarios presented in Table 16 is the mass of the end products. At the same time, combustion generates 9,700 t of ash as a by-product along with district heat, including 1,600 t of wood ash; pyrolysis process produces 12,000 t of biochar without producing ash. More than 95% of the sewage sludge combustion ash was separated by a cyclone at a high temperature to lower the content of heavy metals in the ash. However, only 10–25% of the phosphorus is available in $P_2O_5$ compared to the 5–40% in phosphate ores; the resulting ash still contains phosphorus and can be used in forestry or agriculture. The ash can also be used as a raw material for Portland cement, as an additive in cement production, and to create lightweight bricks and tiles (Donatello and Cheeseman, 2013). In the meantime, biochar can be utilized as a soil amendment (Brassard et al., 2019) and as an adsorbent to extract heavy metals and contaminants from aqueous solutions (Zielińska and Oleszczuk, 2015). The application of sewage sludge derived biochar in soil is still a problem due to its heavy metals and other hazardous substances, such as

dissolved organic carbon, which may wind up in the soil and drinking water after leaching or rainfall (Gopinath et al., 2021).

Table 16. Comparison of mass balance of pyrolysis and combustion.

| Mass Balance | Pyrolysis | Combustion | Units |
|---|---|---|---|
| Inputs | | | |
| Wastewater | 92,000,000 | 92,000,000 | t/a |
| Wood waste | 27,000 | 27,000 | t/a |
| $H_2SO_4$ | 3,700 | 3,700 | t/a |
| NaOH | 3,100 | 3,100 | t/a |
| Intermediate products | | | |
| Sewage sludge to digestion | 950,000 | 950,000 | t/a |
| Biogas | 13,000 | 13,000 | t/a |
| Reject water from mechanical dewatering | 867,000 | 867,000 | t/a |
| Dewatered sludge | 65,000 | 65,000 | t/a |
| Thermally dried sludge | 27,000 | 27,000 | t/a |
| Outputs | | | |
| Clean water | 91,500,000 | 91,500,000 | t/a |
| Ash | 0 | 9,700 | t/a |
| Biochar | 12,000 | 0 | t/a |

The energy balance of the pyrolysis and combustion processes differs as presented in Table 17. Compared to pyrolysis, which yields biochar while using the energy of the pyrolysis gas to heat the pyrolysis process and dry the sludge, combustion primarily produces district heat. The greater mass flow rate of the water, which must be heated to 60 °C for stripping, is the cause of the increased energy consumption in the scenario where ammonia is also recovered from the reject water. As a result, the stripper-scrubber's electricity usage is three times higher than in the scenario where only nitrogen is recovered from the condensate. Without the reject water recovery, in the case of recovering nitrogen from the condensate alone, there is no need for heating since the condensate entering the stripper has a temperature of 60 °C (Endev, 2020); therefore, additional heating is not necessary. However, with reject water recovery, it is expected that the reject water is 28°C before heating since the digested mass is typically 28–32°C after dewatering; as a result, the liquid needs to be heated to 60 °C first to achieve the best stripping performance (Arnold et al., 2000).

Table 17. A comparison of the energy balance for pyrolysis and combustion with and without reject water recovery.

| Energy Balance | Pyrolysis (MWh/a) | Combustion (MWh/a) |
|---|---|---|
| N recovery from condensate only | | |
| Consumption | | |
| Reactor electricity consumption | −6,500 | −9,600 |
| Stripper and scrubber electricity consumption | −680 | −680 |
| Production | | |

| | | |
|---|---|---|
| District heat from sewage sludge | 0 | 56,000 |
| District heat from wood | 0 | 62,000 |
| Net balance of heat | 0 | 120,000 |
| Net balance of electricity | −7,200 | −10,000 |
| NH₃ recovery from condensate and | | |
| Consumption | | |
| Reactor electricity consumption | −6,500 | −9,600 |
| Stripper and scrubber electricity consumption | −3,900 | −3,900 |
| Stripper heat consumption | −34,000 | −34,000 |
| Production | | |
| District heat from sewage sludge | 0 | 56,000 |
| District heat from wood | 0 | 62,000 |
| Net balance of heat | −34,000 | 84,000 |
| Net balance of electricity | −10,000 | −13,000 |

When comparing the final outputs, it can be seen that both methods additionally generate 3,500 t of ammonium sulfate, $(NH_4)_2SO_4$, from the drying fumes and reject water in addition to the respective amounts of biochar and district heat as shown in Table 18. In addition, both scrubbing and stripping use chemicals. Depending on the process circumstances, the stripper uses NaOH to increase the liquid pH to 8–11, and $H_2SO_4$ serves as a scrubbing agent to remove ammonium ions, as the results of Publication I show. Depending on its concentration, $(NH_4)_2SO_4$ is utilized in conjunction with other fertilizers in agriculture. Additional post-treatment, such as crystallization or evaporation, may be necessary to produce commercial-grade fertilizer, which raises the cost of the procedure, as shown in the results of Publication I.

Table 18. Production of ammonium sulfate $(NH_4)_2SO_4$ from drying fumes and reject water.

| $(NH_4)_2SO_4$ **Production** | **Pyrolysis** | **Combustion** | **Units** |
|---|---|---|---|
| $(NH_4)_2SO_4$ /kg NH₃ | 4 | 4 | $kg_{ammsulfate}$/kg $_{NH3}$ |
| $(NH_4)_2SO_4$ from total off-gases | 600 | 600 | $t_{ammsulfate}$/a |
| $(NH_4)_2SO_4$ (from reject water) | 2,900 | 2,900 | $t_{ammsulfate}$/a |
| Total $(NH_4)_2SO_4$ | 3,500 | 3,500 | $t_{ammsulfate}$/a |

### 4.2.3    Comparison of revenues and costs

The cost and revenue estimates used both the combustion and pyrolysis option for dewatered digestion with and without NH₃ recovery from reject water recovery. The main outputs of the two possible sludge treatment processes, combustion and pyrolysis, are heat and biochar, respectively, both of which have a monetary value. Biochar can be valuable commercially, but its market price varies widely depending on various factors, including the source of the feedstock, the conditions of the process, the quantity of equipment employed, and running expenses (Ahmed et al., 2016). Table 19 presents the

costs and the revenue generation in the pyrolysis scenarios PII-S1.1 and PII-S1.2 with or without reject water recovery. The other factors contributing to its ambiguous market value are the lack of technologies for commercial-scale biochar production and a well-established market for various biochar of varying grades. Consumers cannot use biochar in significant quantities due to its expensive cost, but if it is produced in larger quantities commercially, this cost will drop.

The anticipated investment costs for the various pilot plants pyrolysis and combustion processes are presented in Tables 19 and 20. The revenues and significant chemical costs are comparable to these expenses. Compared to pyrolysis, which has annual investment expenditures between 2 and 3 M €, combustion costs between 2–4 M €. The overall revenue for combustion and pyrolysis would be 3–5 M €/a and 3–3.5 M €/a, respectively, when all the by-product revenues are added together, and the chemical expenditures for the reject water recovery scenario are subtracted. These income figures are significantly higher than the annual investment cost.

Since both technologies are in the pilot stage and precise information on the investment and operation expenses at full scale are not yet available, the economic estimates of the scenarios can only be made with a rough degree of accuracy. The Rovaniemi plant's overall investment cost was around 4.3 M €; using a 5% interest rate and a 20-year depreciation period, the investment cost was determined to be around 30 €/t of sludge cleaned. The combined investment expenses for the 55,000 t/a fluidized bed combustion plant in Geneva and the 90,000 t/a plant in Zurich, Switzerland, are between 30 and 50 M €. In Finland, a large fluidized bed plant with a 70,000 t/a capacity and 25% TS costs roughly 25 M €. Similar to this, an investment cost of 13–17 M €, or 35–45 €/t of sludge treated, would be incurred by a pyrolysis plant with a capacity of 30,000 t/a, assuming a 5% interest rate and a 20-year depreciation period (VVY, 2019).

Table 19. Revenue and cost estimation of fertilizer and used chemicals for PII-S1.1 and PII-S1.2.

| Pyrolysis | | | | Reference |
|---|---|---|---|---|
| **Costs** | **Mass, t/a** | **Cost, €/t** | **Total, M €/a** | |
| Chemical used | | | | |
| $H_2SO_4$ | 3,700 | 160 | −0.59 | (Alibaba, 2021a) |
| NaOH | 3,100 | 380 | −1.2 | (Alibaba, 2021b) |
| Annual investment | | | | |
| Sewage sludge | 65,000 | 35–45 | −2.3–(−2.9) | (VVY, 2019) |
| **Revenues** | **Mass, t/a** | **Price (€/t)** | **Total, (M €/a)** | |
| Ammonium sulfate-N | 4,000 | 660 | 2.6 | (Havukainen et al., 2018) |
| Biochar | 12,000 | 400 | 4.8 | (VVY, 2019) |
| **Total** | | | 2.8–3.4 | |

The commercial value of district heat provided through combustion is also contrasted with current district heating pricing, excluding VAT, for scenarios PII-S2.1 and PII-S2.2, as shown in Table 20. The primary difference between the two scenarios is the greater heat consumption during reject water recovery, which must be deducted from the overall district heat production. In addition, the cost of sewage sludge disposal in landfills in Europe ranges from 60 to 200 €/t of TS, and while landfilling is not allowed in Finland, it suggests that district heating has the potential to be profitable also elsewhere in Europe. Even though pyrolysis and combustion have significant treatment costs, they offer an environmentally sound method of utilizing sewage sludge (European Commission, 2002).

Table 20. Revenue and cost estimation of fertilizer and used chemicals for PII-S2.1 and PII-S2.2.

| Combustion | | | | Reference |
|---|---|---|---|---|
| **Costs** | **Mass, t/a** | **Cost, €/t** | **Total, M €/a** | |
| Chemical used | | | | |
| $H_2SO_4$ | 3,700 | 160 | −0.59 | (Alibaba, 2021a) |
| NaOH | 3,100 | 380 | −1.2 | (Alibaba, 2021b) |
| Annual investment | | | | |
| Sewage sludge | 65,000 | 30–60 | −2 - (−3.9) | (VVY, 2019) |
| **Revenues** | **Mass, t/a** | **Price (€/t)** | **Total, (M €/a)** | |
| Ammonium sulfate | 4,000 | 660 | 2.6 | (Havukainen et al., 2018) |
| Phosphorus | 480 | 1,600 | 0.76 | (Havukainen et al., 2018) |
| | **MWh/a** | **Price (€/MWh)** | **Total, (M €/a)** | |
| District heat | 120,000 | 66 | 7.9 | (Hakala, 2016; Helen Ltd, 2021; HSY, 2021; Motiva Ltd, 2021) |
| **Total** | | | 3.3–5.2 | |

AS is created in greater quantities when nitrogen is also recovered from rejected water. Table 19 and Table 20 show the revenue in terms of phosphorus and nitrogen (in the form of AS) for both pyrolysis and combustion. While phosphorus can produce 0.8 M €/a in revenue from combustion, ammonium sulfate can create 2.6 M €/a by combining nitrogen recovery and combustion. The ash produced in addition to district heating is the cause of the combustion's high revenue.

On the other hand, the recovery of reject water also increases the amount of chemicals needed for scrubbing and stripping. The operational cost is increased by 1.2 M €/a and 0.6 M €/a by $H_2SO_4$ and NaOH, respectively.

### 4.2.4    **LCA of dewatered sludge digestate**

The environmental performance of dewatered sewage sludge digestate utilization with composting, pyrolysis, or combustion, including nitrogen recovery, was studied in Publication III. Figure 16 shows the net LCIA results, the contribution of the processes to the direct impacts and avoided impacts, and the range of net result with error bars (high and low performance) and selected impact categories, whereas Figure 17 shows the relative contribution of processes to produced and avoided emissions.

In the results, PIII-S2.2 (combustion with nitrogen recovery) had the lowest net impact on climate change, PIII-S1 (composting) on fine particulate matter, marine eutrophication, mineral resource scarcity, and human toxicity (cancer), and PIII-S2.1 (combustion without nitrogen recovery) on terrestrial acidification, freshwater eutrophication, and terrestrial ecotoxicity. PIII-S3 (pyrolysis with nitrogen recovery) has the widest range of impact results across the impact categories. The pyrolysis data, which is based on laboratory tests, is more uncertain than the data for PIII-S1 (composting) and PIII-S2, collected from existing facilities, albeit the data for PIII-S2 is from a pilot plant. Despite the range of net results PIII-S3 does not outperform the other scenarios in most impact categories. PIII-S3 outperforms PIII-S2.1 and PIII-S2.2 in terms of human toxicity (cancer) and PIII-S1 does so for climate change.

The high and low-performance values of the parameters for flue gas emission, such as the flue gas emissions concerning particle formation and terrestrial acidification and the NaOH demand for cleaning flue gas in other impact categories, contribute to a wide range of net results for PIII-S3. The wide range of net results is also due to the electricity consumption in pyrolysis, scrubbing, and biochar and ammonium sulfate liquid production, which affect transport emissions. $N_2O$ emissions contribute 22% and 16% of the total emissions to climate change in combustion scenarios PIII-S2.1 and PIII-S2.2. In PIII-S2.2, nitrogen recovery from the condensate increases the net impacts for most impact categories compared to PIII-S2.1, which is due to the usage of $H_2SO_4$ and NaOH. In the case of the climate change impact category, recovering nitrogen from condensate increases emissions by roughly 50%, whereas the avoided impacts due to AS usage reduces emissions, resulting in a lower net impact for PIII-S2.2 than PIII-S2.1. NaOH generation increases emissions by 80% and $H_2SO_4$ production by 10%. The production of NaOH and $H_2SO_4$ causes freshwater eutrophication, mineral resource scarcity, and toxicity effect categories (80–94%). In marine eutrophication, PIII-S2.1 directs nitrogen-containing condensate to the WWTP, resulting in 95% greater emissions than PIII-S2.2 and a lower net impact for PIII-S2.1. Adsorption from condensate or stripping gas would reduce chemical use and nitrogen recovery emissions.
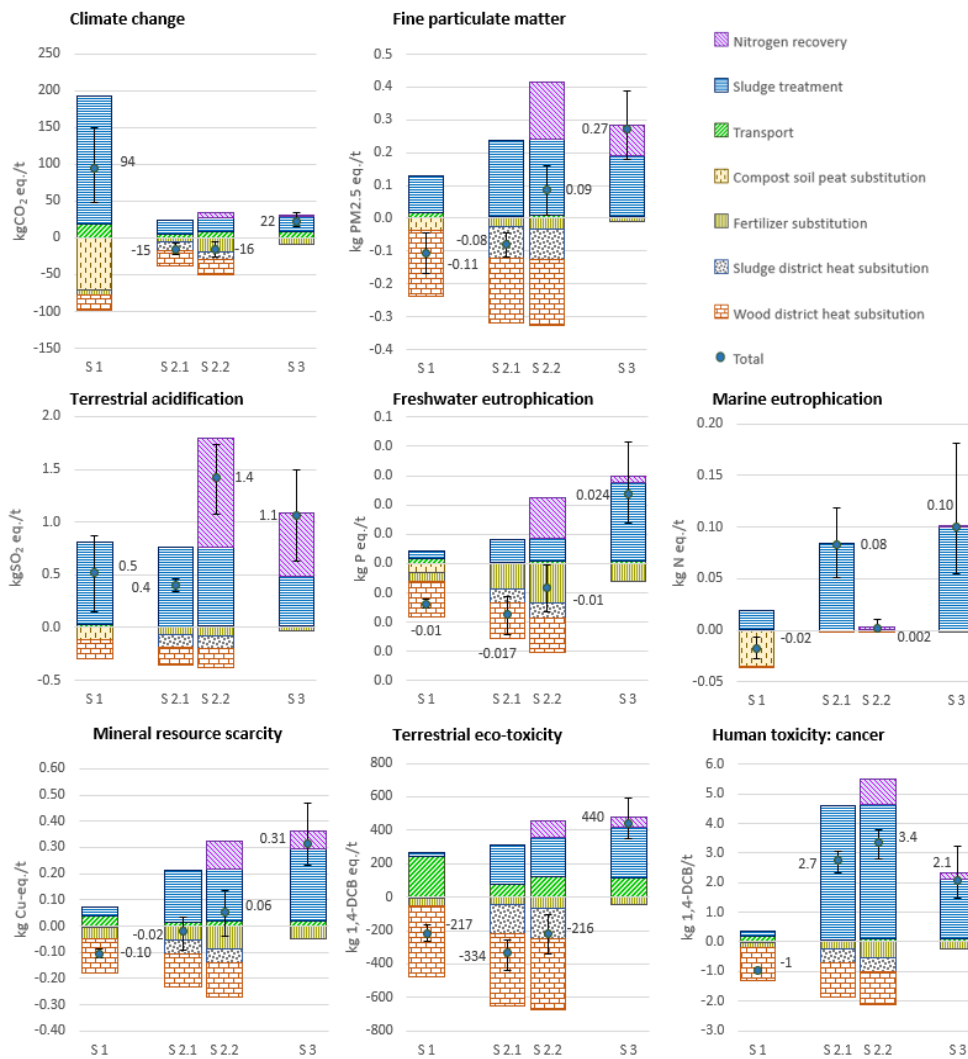
Figure 16. LCIA results for the contribution assessment for scenarios PIII-S1(Composting), PIII-S2.1(Combustion without nitrogen recovery) and PIII-S2.2 (Combustion with nitrogen recovery), PIII-S3(Pyrolysis) for the selected impact categories.

Figure 18 shows the contributions of each process to the produced and avoided emissions. Sludge treatment causes the majority of the emissions in almost all impact categories in each scenario, except for the toxicity impacts in PIII-S1, where transport mainly causes emissions (more than 90%), and in PIII-S2.2, where nitrogen recovery has the highest impacts on acidification and eutrophication (60–85%). For the climate change impact category, using compost in garden soil production contributes to 70% of the avoided emissions. In PIII-S2.1 and PIII-S2.2, replacing, marginal district heat (biomass-based

district heat) with district heat produced from incinerating the digestate accounts for 20–30% of the avoided emissions across the impact categories. In scenarios PIII-S1, PIII-S2.1, and PIII-S2.2, replacing marginal district heat with district heat from wood waste reduces emissions by 40–50% across impact categories. Due to wood waste being used in pyrolysis, for PIII-S3 the only substituted product is fertilizer. Out of the avoided impacts 60% are related to the substitution of mineral phosphorus and nitrogen by the phosphorus and nitrogen contained in biochar and the remaining 40% is attributable to ammonium sulfate. Among the effects that biochar prevents, 60% can be attributed to reduced use of phosphorus fertilizer, and 40% can be attributed to reduced use of nitrogen fertilizer.



Figure 17. LCIA result of the relative contribution of direct and substituted emissions.

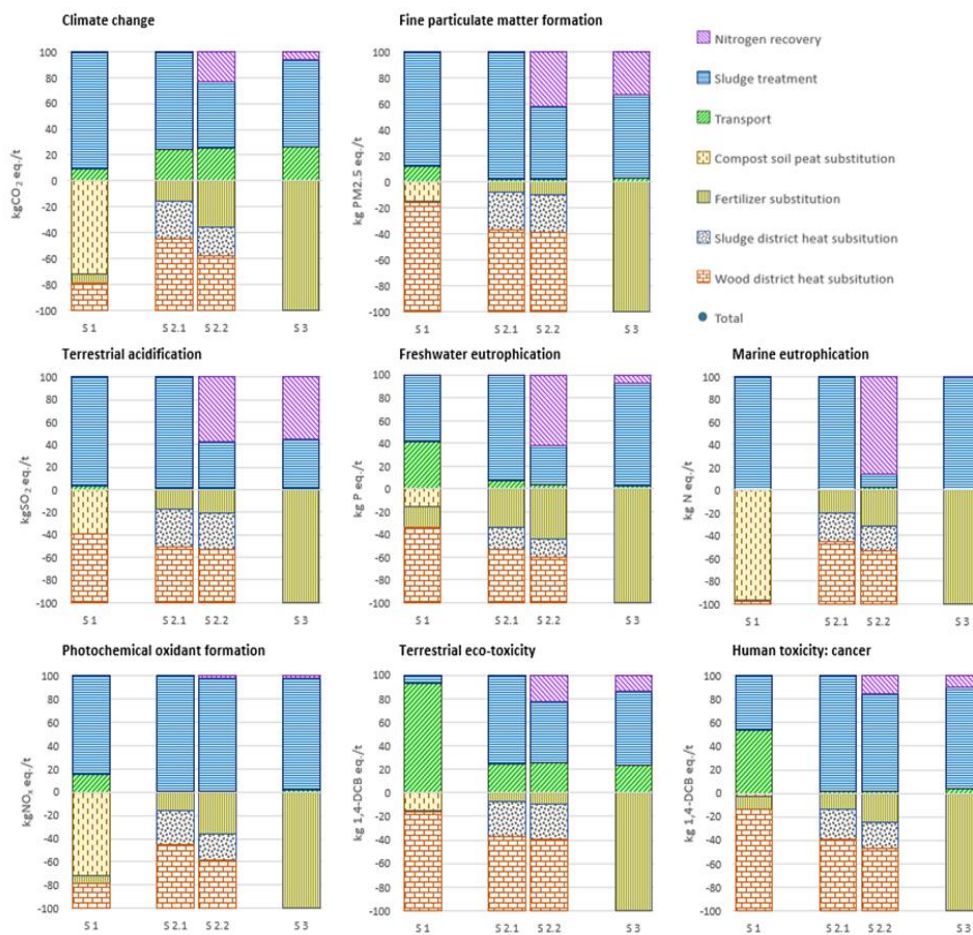In PIII-S1, composting air emissions (30–50% for climate change, particulate emissions, and terrestrial acidification), peat production emissions (60–100% for eutrophication), and sand production (40–80% for toxicity and mineral resource scarcity) dominate the sludge treatment emissions. The sludge treatment emissions in PIII-S2.1 and PIII-S2.2 stem from NaOH production (40% for climate change), combustion emissions (80–90% for particle emissions, acidification, and human toxicity), and electricity demand (90–100% for eutrophication, mineral scarcity, and terrestrial ecotoxicity). In PIII-S3, the main emissions from sludge treatment are electricity usage in the pyrolysis process (50–90% for climate change, particulate emissions, acidification, and eutrophication), combustion of the pyrolysis gas (70% for mineral resource scarcity and eco-toxicity) and directing residual ammonium sulfate to the WWTP (95% for marine eutrophication).

The LCIA results are based on the LCI data, and the selection of parameter values and modeling choices impact the results, with some choices and parameters being more sensitive to the results than others. In the composting scenario, PIII-S1, the substitution of compost soil and garden soil was assumed to be based on volume, which means that their densities affect the results. Furthermore, literature data was used to quantify the climate change impacts of the composting emissions and soil displacement, and therefore accurate field-based data could alter the net results. In the case of the combustion scenarios (S2.1 and PIII-S2.2) the heat efficiency, electricity consumption, and phosphorus in the product ash were based on the measurements from the facility, and their uncertainty is low. When nitrogen recovery is added to the combustion process in PIII-S2.2, the condensate nitrogen content, NaOH demand, and recovery efficiency of the scrubber are highly sensitive parameters, especially for the climate change impact category. In PIII-S3, the sensitive parameters concern the wood demand and pyrolysis gas combustion characteristics such as the NaOH demand, NOx emissions, and sulfur transfer coefficient. Pyrolysis reactor operating conditions affect the pyrolysis gas combustion parameters, while drying and pyrolysis heat requirements define the wood demand.

## 4.3 Recovery from reject water of dewatering and condensation from drying fumes

The LCIA results of the nitrogen recovery from reject water and condensate resulting from thermal drying fumes for selected impact categories are presented in Figure 18. In addition, Figure 19 depicts the shares of contributing processes to the produced and avoided emissions.

In Publication IV, it is not easy to establish clear-cut conclusions on the superiority in terms of environmental performance. Scenario PIV-S3 (adsorption) performs better in three of the five impact categories, including climate change with biogenic carbon, freshwater eutrophication, and marine water eutrophication. PIV-S1 (WWTP) exhibits the lowest net impact for the remaining two categories: climate change without biogenic carbon and acidification. Except for having the greatest net impact in two categories—

climate change with biogenic carbon and freshwater eutrophication—PIV-S2 (stripping and scrubbing) is positioned in the middle.

In this study, it is important to consider the biogenic carbon impact because biochar can be used for carbon sequestration in PIV-S3 (adsorption). On the other hand, using biochar for carbon sequestration allows for the avoidance of 139 kt $CO_2$ eq./FU, and an additional 6 kt $CO_2$ eq./FU, when substituting district heat and fossil-based nitrogen fertilizers. The carbon dioxide emissions from the biochar-based carbon capture in PIV-S3 (adsorption) were net negative. During pyrolysis in PIV-S3 (adsorption), a 90%/10% mixture of wood and sewage sludge biochar were produced. One metric ton of this mixture removes 1.41 metric tons of $CO_2$ eq. from the atmosphere when applied to soil for carbon sequestration. Depending on the source of the biochar, this amount typically ranges from 0.8 to 2.9 t $CO_2$ eq./t, according to Jeswani et al. (2022). According to Cao and Pawłowski (2013), 0.8 t $CO_2$ eq./t of carbon can be sequestered by biochar made from sewage sludge, while 2-2.6 t $CO_2$ eq./t of carbon can be sequestered by biochar made from forest waste as per Azzi et al. 2019. PIV-S2 (stripping and scrubbing) generated 3.5 t $CO_2$ eq./FU overall but prevented 1.5 t $CO_2$ eq./FU. Significant emissions come from the combined stripping and scrubbing process, which uses chemicals (NaOH, 0.9 kt $CO_2$ eq./FU, and sulfuric acid, 0.5 kt $CO_2$ eq./FU) and electricity (1 kt $CO_2$ eq./FU). Only direct emissions (0.2 kt $CO_2$ eq./FU) are produced in the baseline scenario PIV-S1 (WWTP) in this category.

The direct emissions from PIV-S3 (adsorption) in the scenario for climate change without biogenic carbon were 12 kt $CO_2$ eq./FU, while a total of 5 kt $CO_2$ eq./FU were avoided. Climate change without biogenic carbon dioxide does not include the sequestration of biogenic carbon in biochar, which means that the net impact is considerably higher than in the case of including biogenic carbon dioxide. The net climatic impacts in PIV-S2 (stripping and scrubbing) and S1 (CWWTP) are essentially the same.

PIV-S1 (WWTP) performed better concerning freshwater eutrophication than PIV-S2 (stripping and scrubbing), which was mostly because of the consumption of NaOH and sulfuric acid during stripping and scrubbing and, to a lesser extent, because of the fuel consumption during fertilizer spreading. The chemical consumption can also be optimized.
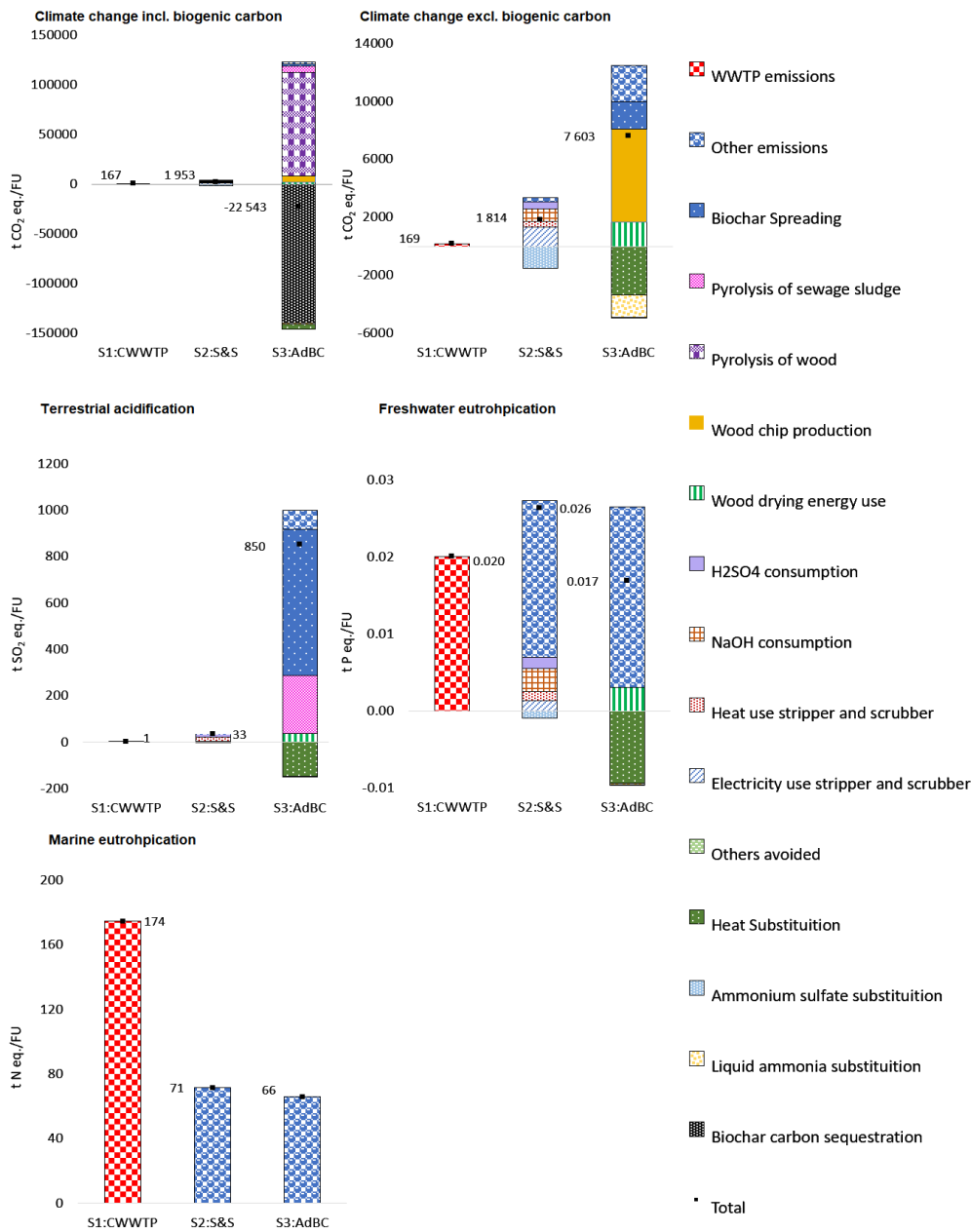
Figure 18. LCIA results for the selected impact categories in PIV-S1 (WWTP), PIV-S2 (stripping and scrubbing), and PIV-S3 (adsorption).

A contribution analysis was used to evaluate the environmental impact of each process further, as illustrated in Figure 18. PIV-S1 (WWTP) only has the WWTP process

producing emissions. In the case of climate change including biogenic carbon, the generation of wood biochar and the spreading of biochar account for up to 68% and 21%, respectively, of the total emissions in PIV-S3 (adsorption). Significant emissions are produced due to the production of wood biochar and its high demand for electricity. However, excess heat from wood pyrolysis can be used for district heat generation. In addition, biochar-based carbon sequestration accounts for 96% of the avoided emissions.

In the case of PIV-S2 (stripping and scrubbing), the electricity consumption during stripping and scrubbing is the process which most contributes to the climate change impact (both including and excluding biogenic carbon). The consumption of NaOH and $H_2SO_4$ accounts for 15% and 25%, respectively, of the total emissions concerning climate change, including biogenic carbon. The leading causes of high electricity usage observed here are the large input flows of condensate and reject water. Nitrogen recovery, which enables the replacement of fertilizers based on fossil fuels, is the cause of all the avoided emissions. Vaneeckhaute et al. (2014) showed values as low as 0.01 MJ/kg $NH_4$-N, while Publication IV used an electricity usage of 0.2 MJ/kg $NH_4$-N.

Regarding terrestrial acidification, PIV-S2 (stripping and scrubbing) has an environmental impact mostly due to using heat, energy, and sulfuric acid during stripping and scrubbing. In PIV-S3 (adsorption), biochar spreading has the most influence, although there is also a sizable impact from the pyrolysis of sewage sludge. The switch from wood pyrolysis to district heating also results in 150 t $CO_2$ eq./FU of avoided emissions.

In the case of eutrophication impact categories, other emissions are the most important. In PIV-S2 (stripping and scrubbing), "other emissions", which include, for example, the transportation of chemicals, the application of nitrogen fertilizer (ammonium sulfate) on fields, the use of water for acid dilution, and the recycling of residual stripping liquid, have the greatest influence on freshwater and marine eutrophication. In PIV-S3 (adsorption), "other emissions" also refer to transporting and applying sewage sludge and wood biochar.
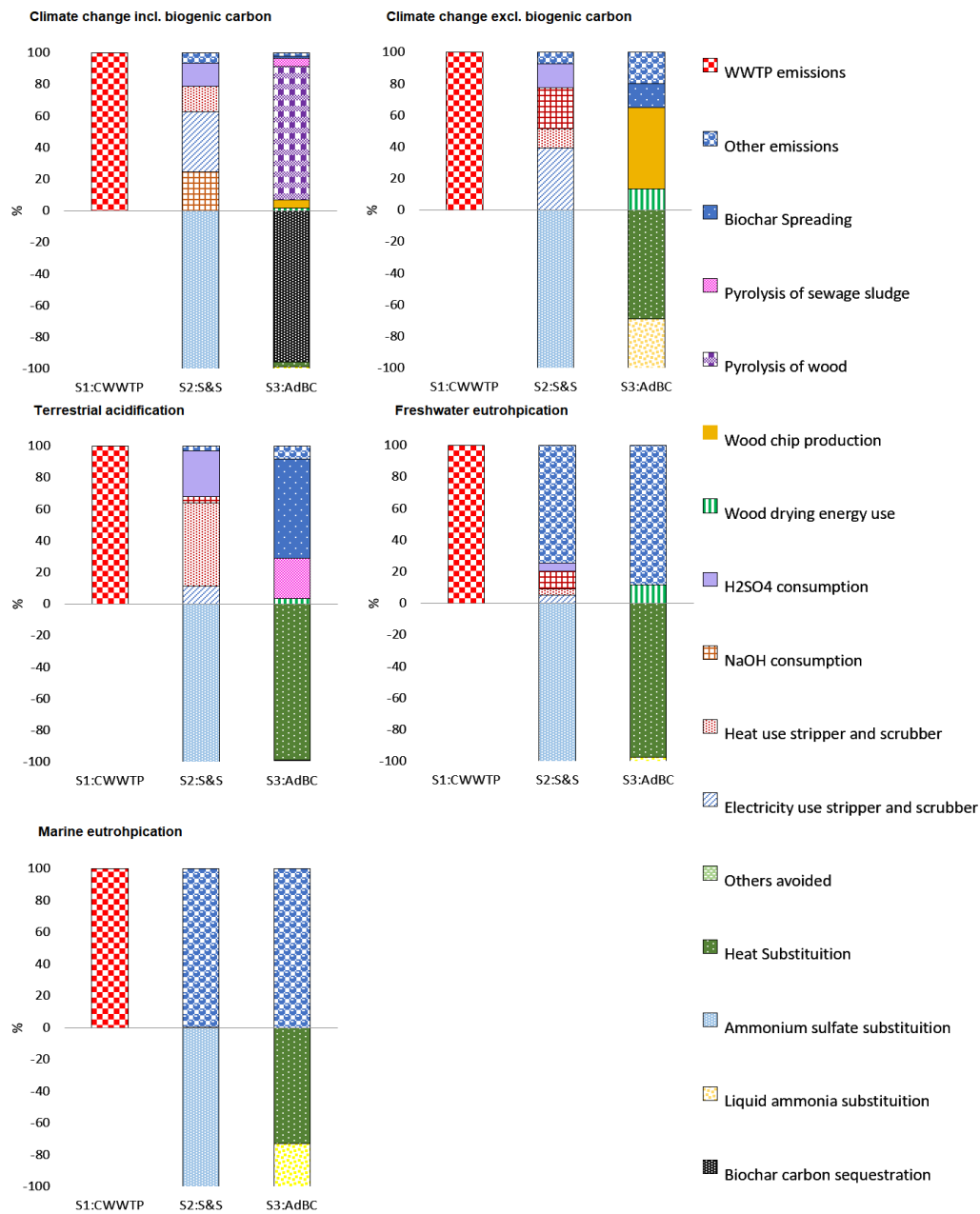
Figure 19. LCIA contribution assessment of scenarios PIV-S1 (WWTP), PIV-S2 (stripping and scrubbing) and PIV-S3 (adsorption) for the selected impact categories.

In general, efficiency, operational circumstances, and equipment design all affect electricity demand, as shown in the results of Publication II. Additionally, the source of electricity generation significantly impacts the environmental performance; when using renewable electricity (wind power), the net emissions decreased by 95% in Publication IV.

The requirements for the end product influence the acid choice during the scrubbing procedure. Here, ammonium sulfate was made using sulfuric acid ($H_2SO_4$). Ammonium sulfate has a wide range of possible uses in addition to being used as a nitrogen fertilizer in agriculture. For instance, it functions as a chemical in flame retardants and a wood preservative (Speight, 2017). The primary use of the ammonium sulfate recovered from waste streams, however, is the replacement of synthetic ammonium sulfate produced by the ammonia from the energy-intensive Haber-Bosch process as a source of important macronutrients N and S. Ammonium sulphate accounts about 50% of the global sulphur fertilizer consumption. Ammonium-based fertilizers are known to volatilize $NH_3$ in soils with pH > 7. Hence the usage of ammonium sulfate is also decreasing to control $NH_3$ emissions and alternatives are being considered to replace sulfur fertilizers such as superphosphate, potassium sulphate, magnesium sulphate, gypsum, and polysulphate (Powlson and Dawson, 2022). To recover ammonium, organic acids like citric acid to produce ammonium citrate or acetic acid to produce ammonium acetate, may be used in place of sulfuric acid. However, ammonium citrate could be useful in phosphorus-saturated lands compared to ammonium acetate, which has limited usage in agriculture (Jamaludin et al., 2018).

The sensitivity of the net results to individual parameters was quantified utilizing the sensitivity ratios summarized in Table 21 for the studied impact categories. Nitrogen removal efficiency is the only sensitive parameter in PIV-S1 (WWTP) that significantly varies across all effect categories, indicating a solid risk to marine eutrophication. The influence on marine eutrophication can be reduced due to the efficient removal of nitrogen, and the net impact is highly sensitive to this parameter with an SR of -8.5. In the case of PIV-S1 (WWTP), other factors, such as electricity and heat demand, have a negligible impact on all impact categories.

Table 21.Sensitivity ratios (SRs) for parameters in each scenario against selected impact categories.

| S1 (CWWTP) | CC incl. biogenic | CC excl. biogenic | FWE | ME | TA |
|---|---|---|---|---|---|
| Parameter | | | | | |
| Electricity | 0.58 | 0.58 | 0.65 | 0.00 | 0.66 |
| Heat | 0.28 | 0.28 | 0.35 | 0.00 | 0.34 |
| Lime | 0.15 | 0.15 | 0.00 | 0.00 | 0.00 |
| N removal efficiency | 0.00 | 0.00 | 0.00 | -8.49 | 0.00 |
| Electricity biogas | 0.02 | 0.02 | 0.09 | 0.00 | 0.06 |

| | CC incl. biogenic | CC excl. biogenic | FWE | ME | TA |
|---|---|---|---|---|---|
| Heat biogas | -0.23 | -0.22 | 0.35 | 0.00 | 0.30 |
| **S2 (S&S)** | **CC incl. biogenic** | **CC excl. biogenic** | **FWE** | **ME** | **TA** |
| Parameter | | | | | |
| Electricity use | 0.71 | 0.76 | 0.05 | 0.00 | 0.12 |
| Heat use | 0.29 | 0.21 | 0.04 | 0.00 | 0.54 |
| $H_2SO_4$ use | -0.26 | -0.31 | 0.02 | 0.00 | 0.21 |
| NaOH | 0.32 | 0.32 | 0.08 | 0.00 | 0.03 |
| Water use | 0.00 | -0.01 | 0.00 | 0.00 | 0.00 |
| Stripper-Scrubber efficiency | -0.16 | -0.27 | -0.01 | -1.44 | -0.01 |
| Distance of fertilizer spreading | 0.00 | -0.01 | 0.00 | 0.00 | 0.00 |
| **S3 (AdBC)** | **CC incl. biogenic** | **CC excl. biogenic** | **FWE** | **ME** | **TA** |
| Parameter | | | | | |
| Nitrogen adsorption capacity, SS biochar | 0.12 | -0.11 | 0.03 | 0.0001 | -0.07 |
| Electricity demand, SS biochar production | 0.02 | 0.07 | 0.03 | 0.0001 | 0.01 |
| Heat demand, SS biochar production | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Electricity demand, wood biochar production | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Heat demand, wood drying | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| $SO_2$ removal | 0.00 | 0.00 | 0.00 | 0.0000 | 0.29 |
| Carbon share in biochar | -0.31 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Biochar nitrogen usability | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Nitrogen adsorption capacity, wood biochar | 0.52 | -0.50 | 0.13 | 0.0002 | -0.31 |
| Electricity demand wood biochar production | 0.04 | 0.12 | 0.06 | 0.0002 | 0.00 |
| Heat demand biochar | 0.05 | 0.10 | 0.13 | 0.0002 | 0.04 |
| Wood processing emissions | 0.28 | 0.84 | 0.00 | 0.0000 | 0.00 |
| Yield of wood biochar | -6.83 | -0.43 | 0.26 | 0.0004 | 0.09 |
| Excess heat production | -0.19 | -0.40 | -0.51 | -0.0009 | 0.15 |
| Substituted district heat emissions | 0.00 | 0.00 | 0.00 | 0.0000 | 0.01 |
| C share remaining in soil | -11.37 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Carbon content of wood biochar | 9.57 | 0.00 | 0.00 | 0.0000 | 0.00 |

| CF (Carbon footprint) | 5.77 | -0.09 | 0.02 | 0.0000 | -0.06 |
| Steam from biomass and natural gas | 4.06 | 12.62 | -0.01 | -0.0002 | -0.04 |

$|SR|>1$ Particularly important,   $|SR|=0.8-1$ Important parameter,
$|SR|=0.2-0.8$ Slightly important,   $|SR|<0.2$ Minor importance.
**CC incl. biogenic** is Climate change including biogenic carbon
**CC excl. biogenic** is Climate change excluding biogenic carbon
**FEW** is Fresh water eutrophication
**ME** is Marine water eutrophication
**TE** is Terrestrial acidification

The results in PIV-S2 (stripping and scrubbing) are most sensitive to two parameters, namely electricity use and stripper-scrubber efficiency, as shown by the highest and lowest SR values. Raising the demand for electricity increases the impact on the global warming potential, but other impact categories are unaffected. The efficiency of the stripping and scrubbing process, on the other hand, has a negative SR concerning the marine eutrophication impact, so a declining efficiency considerably worsens the eutrophication impact (for example, a 10% reduction increases the impact by 14%).

The yield of wood biochar, the amount of carbon remaining in the soil, the total carbon content of wood, the CF (carbon footprint), and steam produced from biomass and natural gas had the most substantial influence on the potential for global warming among all the variables in PIV-S3 (adsorption). When the biochar yield is raised, the impact on the climate change categories is reduced (for example, a 10% increase will reduce the impact by 68%). The biochar yield with SR < -6 implies a wide range of results. Although the range of biochar yield is typically broad, it can be managed by changing the pyrolysis conditions. Therefore, the environment should be adequately controlled to give a high amount of biochar.

Additionally, both categories of climate change in PIV-S3 (adsorption) are greatly impacted by the amount of steam produced by biomass and natural gas combustion. As a result, this parameter is regarded as particularly sensitive because switching just a tiny fraction of biomass to natural gas as the source of steam generation would create a noticeable change in the outcomes. On the other hand, the need for external energy from biomass or natural gas increases with the yield of biochar from pyrolysis. The biochar yield and steam from biomass are contradictory. The higher the share of biomass converted to biochar, the less it can be used for steam production. The biochar yield is, however, more sensitive to climate change including biogenic $CO_2$.

On the other hand, an increase in the biochar yield over a certain limit may reduce the share of C remaining in the soil. As a result, it is important to aim for the highest pyrolysis yield possible. The most sensitive variable for climate change, including the biogenic $CO_2$ impact category, is the share of carbon in the soil. The emissions will rise by 110% if this

variable falls by 10%. The parameter is still very speculative due to the relatively little long-term experimental evidence on carbon persistence in various soil conditions.

By 2050, the quantity of carbon absorbed by biochar is predicted to rise to 0.3-2 Gt $CO_2$ annually worldwide (Fawzy et al., 2021). On the other hand, several variables, including temperature, residence time, pressure, and feedstock composition affect the production and characteristics of pyrolysis products. As a result, the findings for PIV-S3 (adsorption) are highly sensitive, and more information is required to accurately estimate the ammonia adsorption and the biochar's capacity to contribute to global warming (Fawzy et al., 2021). However, new research indicates that biochar may be able to facilitate carbon capture while potentially neutralizing GHG emissions, despite varied production processes and environmental factors (Matuštík et al., 2022).

## 4.4  Discussion

N recovery is in high demand to make wastewater treatment more efficient, reduce the usage of natural raw materials and reduce the energy demand to produce N fertilizers. Moreover, N recovery is also aimed at reducing the environmental impacts of production of N fertilizers from Haber-Bosch process.

In light of the aims and objectives of this dissertation, the recovery of N is inevitable to reduce the load of natural materials and to produce alternative fertilizers, energy and ultimately reduces the environmental impact by avoiding carbon emissions.

In this work, the starting point was to determine the potential for nitrogen recovery from the exhaust fumes of the thermal drying of sewage sludge. The selection of scrubbing with acidic media as the means to recover nitrogen from the exhaust fumes is based on the maturity of the technology. The results of the first publication have shown that a low concentration of nitrogen can be recovered. However, on the other hand, diluted ammonium sulfate, the high capital cost, and chemical usage with a focus on producing commercial grade AS fertilizer, make the economy of N recovery from drying fumes very challenging.

Moreover, the post-treatment of diluted ammonium sulfate from the drying fumes would also elevate the production costs. It was also found in the results of Publication I that the L/G ratio and recycling rate of the liquid in the scrubber performed a critical role in estimating costs. It was found that the L/G ratio of 1.5 and recycling rate of 70% produced optimal results at a lower cost, as well as providing the maximum N recovery, and avoiding flooding the scrubber.

The result of Publication I led to the second research question related to integrating pyrolysis and combustion with scrubbing to maximize nitrogen recovery. The purpose of condensing the drying fumes is to recover energy from the drying fumes and reduce the

water content. N could then be potentially recovered from the condensate. In the case of reject water from dewatering digestate the presence of $NH_3$ in the reject water makes it possible to combine it with the drying fumes condensate to maximize the N recovery and improve AS production. The integration of scrubbing with pyrolysis and combustion not only increased the nitrogen recovery but also led to the production of additional value-added products. In addition to AS from scrubbing with acidic media, biochar is produced from pyrolysis and district heat can be produced from combustion. The difficulty recovering nitrogen with a low concentration of ammonia in the thermal drying fumes of sewage sludge was solved by condensing and combining this stream with reject water to maximize the nitrogen recovery in the form of ammonium sulfate. In the pyrolysis process, the produced biochar has applications as a viable and cost-effective way to sequester carbon (Sun et al., 2022), as an adsorbent to remove pollutants and heavy metals from aqueous solutions (Brassard et al., 2019), can act as a soil amendment (Zielińska and Oleszczuk, 2015), and as catalysts in advanced oxidation processes and energy storage devices (Mian et al., 2022). Pyrolysis at high temperatures reduces the bioavailability of heavy metals and converts them into stable forms (P. Zhang et al., 2020). The concentration of heavy metals such as zinc (Zn), copper (Cu), chromium (Cr) and lead (Pb) present in biochars increases with increasing in temperature, but due to the conversion into the oxidized forms of these heavy metals due to high temperatures, their bioavailability is reduced (Gopinath et al., 2021). The heavy metal effects in biochar need further study (Li et al., 2021). At 500 °C, microplastics are eliminated, reducing environmental danger (Ni et al., 2020). Additionally, sewage sludge combustion produces heat energy that can be utilized for district heating, and ash containing phosphorous which can be utilized in forestry or agriculture if the heavy metal concentrations are low enough according to legislation limits.

In the cost and revenue calculations in Publication II, the main products are biochar and heat, which have monetary value. However, the price of biochar can be affected significantly by the willingness to pay for it and that is related to the quality and suitability of the product for use and its competitiveness against alternative products. Other factors such as the feedstock source, process parameters, equipment used, and operating costs can also vary biochar prices. The commercial production of biochar is limited, resulting in uncertain market demand and high costs. Nevertheless, biochar made from sewage sludge digestate could potentially have added value if utilized in pyrogenic carbon capture and storage (PyCCS), where the sequestered carbon dioxide can be sold on the market.

In the estimation of the environmental impacts of selected technology in Publication III, the results showed that the climate impact of pyrolysis of sewage sludge is 79 kg $CO_2$ eq./$t_{TS}$ (range 55- 124 kg $CO_2$ eq./$t_{TS}$), which is higher in comparison to the previous research work by Cao and Pawłowski (2013) (12-108 kg $CO_2$ eq./$t_{TS}$). The combustion scenarios have a climate impact of 29 kg $CO_2$ eq./$t_{TS}$ (69 to 6 kg $CO_2$ eq./$t_{TS}$), which is at the lower end of the ranges discovered by Zhang et al. (2019) and Yoshida et al. (2013), i.e., 32-103 and 36-3183 kg $CO_2$ eq./$t_{TS}$, respectively. Regarding the effects of climate

change, the $N_2O$ emissions from sewage sludge mono-incineration are significant (Svoboda et al., 2006).

The adsorption efficiency of biochar for ammonia is lower than other adsorbents. It seems that it is not sufficient in practice considering the amounts of biochar needed for adsorption of N from liquids (or gas). This leads to the need for better adsorbents if it is desired to avoid low concentration scrubber products (Song et al., 2023). The combination of anaerobic digestion and pyrolysis has been deemed a promising technology that could provide net energy and reduce climate change. Additionally, in Publication III, it was discovered that sewage sludge digestate combustion with phosphorus recovery from ashes had a lower climate impact than traditional sludge management and phosphate rock mining techniques. These results demonstrate how various technologies may be used to lessen the environmental impact of sewage sludge treatment (Singh et al., 2022).

In further investigation to utilize biochar as an adsorbent to recover ammonia from the reject water from dewatering processes and condensate from thermal drying fumes, biochar as an adsorbent was proven to sequester carbon in Publication IV. Additionally, according to Matuštík et al. (2022), biochar is able to facilitate carbon capture while potentially neutralizing GHG emissions, despite varied production processes and environmental factors. The environmental effects in the stripping and scrubbing are caused mainly by the usage of chemicals ($H_2SO_4$ and NaOH) and electricity consumption during the stripping and scrubbing process. Vaneeckhaute et al. (2014) used a value as low as 0.01 MJ/kg $NH_4$-N, while the current investigation used an electricity usage of 0.2 MJ/kg $NH_4$-N. Efficiency, operational circumstances, and equipment design generally affect electricity demand (Publication 1). Additionally, the method used to generate electricity significantly impacts how environmentally friendly it is. When using renewable electricity (wind power), net emissions were reduced by 95% in PIV-S2 (stripping and scrubbing).

Chemical consumption can also be minimized. The requirements for the finished product influence the acid choice during the scrubbing procedure. Here, ammonium sulfate was made using $H_2SO_4$. Ammonium sulfate has a wide range of possible uses in addition to being used as a nitrogen fertilizer in agriculture. For instance, it functions as a chemical in flame retardants and as a wood preservative (Speight, 2017). The primary use of the ammonium sulfate recovered from waste streams, however, is the replacement of synthetic ammonium sulfate made by the energy-intensive Haber-Bosch process as a source of important macronutrients N and S. To recover ammonium, organic acids like citric acid or acetic acid may be used in place of sulfuric acid (Jamaludin et al., 2018).

The European Union (EU) is actively working towards implementing a circular economy model that integrates various sectors for more effective raw materials and waste management. The goals of the circular economy policy framework, including resource recovery, are closely linked to wastewater treatment processes and sewage sludge

management. Stakeholder involvement, information dissemination, and the transition of water management utilities into resource recovery advocates are essential for overcoming these obstacles and promoting sustainable practices in waste management (Preisner et al., 2022).

Declaring one technology as the ideal solution to recover nitrogen from sewage sludge and side streams of WWTPs is impossible. The impact of different sludge management strategies on the environment cannot be universally reduced under all conditions, as stated by Lombardi et al. (2017). According to Campbell (2000), the critical factor in selecting the appropriate sludge management route is ensuring it aligns with the specific local conditions. However, comparing the environmental impact of different strategies remains a complex task that is difficult to generalize. Furthermore, using sulfuric acid recycled from oil refineries is another way to lower environmental effects (Asof et al., 2017; Merkel et al., 2021). The end-product from using nitric acid ($HNO_3$) would be ammonium nitrate, the second-most used nitrogen fertilizer after urea. Additionally, it can be used as a nitrogen oxide adsorbent or as a component in pesticides (Zuo et al., 2022).

Future research could focus on investigating the toxic effects of various pollutants and pathogens generated during sludge treatment processes, particularly emerging pollutants like perfluorinated chemicals, phthalates, and phenolics (Teoh and Li, 2020). The decomposition of organic components in sludge may also be accelerated by investigating pretreatment treatments like thermolysis and ultrasonic treatment. (Nakakubo et al., 2012). Furthermore, the treatment of landfill leachate and sludge waste liquid should be considered in sludge management strategies (Guo et al., 2015). This will encourage utilizing sewage sludge in the future and would also be considered a valuable product.

In all the above-mentioned discussion, there is a clear indication the N recovery is needed not only to handle growing sewage sludge volumes but also to produce alternative fertilizer, adsorbents, to recover energy and to sequester carbon. The selection of technology to recover N from sewage sludge depends on the required outcomes. In this dissertation, pyrolysis, combustion, and stripping followed by scrubbing are suggested. To deal with technical difficulties, more research work is needed to generate accurate data to apply on a commercial scale. Another important factor, which hinders forward development, is the feasibility of these technologies. Even though these mentioned technologies are well established, dealing with sewage sludge requires more details such as the moisture content, geographical location, number of total solids etc. The products of N recovery from sewage sludge have a promising outlook but they need post treatment. In the stripping and scrubbing of thermal drying fumes, the resultant ammonium sulfate is very diluted and post treatment such as evaporation and crystallization may be required to enhance the quality of product. Similarly, in the pyrolysis of sewage sludge, the selection of an adequate temperature to maximize the production of biochar and to reduce the bioavailability of heavy metals, would increase the demand for biochar. The adsorption capacity of sewage sludge biochar could be increased by adding biochar from other feedstock to maximize the nitrogen recovery. In general, it is also needed to focus on the commercialization of these N recovery approaches from sewage sludge, and on the

economic concerns related to capital and operational costs and characterizations to estimate the requirements for modification and treatments to enhance the sustainability and quality of subsequent products.

# 5 Conclusions

The primary aim of this dissertation is to investigate and evaluate the recovery of nitrogen from sewage sludge treatment processes, as well as the recovery from thermal drying processes and reject water from mechanical dewatering, as well as the environmental and economic implications of these recovery methods. In the first part of the study, an acid scrubber was used to recover ammonia from the drying fumes produced during the thermal drying of sludge. ASPEN Plus simulations were conducted for two ammonia concentrations: 75 ppm and 100 ppm. It was observed that low pH values (between 1–3) were suitable for efficient ammonia recovery, achieving an impressive efficiency of 99%. However, with higher pH values (4–5), the efficiency decreased to 88%. A liquid to gas (L/G) ratio of 1.5 with a recycling rate of 70% was found to be feasible for this process. The total capital investment for the scrubber in a sludge treatment plant with a capacity of 7,700 ton/a was calculated to be 290 k€, with an operational cost of 113 k€/a, including the cost of sulfuric acid and water. The cost of ammonia removal was estimated at 20 €/t of sludge, and the production cost of ammonium sulfate was 18 €/t of sludge. The conclusion is that in current conditions the cost of producing ammonium sulfate fertilizer was high, making it impractical for commercial use. It answers the RQ1 that scrubbing with acidic media can recover $NH_3$ from the exhaust fumes of thermal drying even at the lowest concentration.

In order to maximize the recovery of nutrients and energy from municipal sewage sludge, the second portion of the study emphasized on the integration of pyrolysis and combustion processes with gas scrubbing technology at a large-scale WWTP. Of the entire nitrogen in the wastewater entering the WWTP (4 900 t/a), over 73% (3,600 t/a) was lost to the air and pure water. The remainder was split up among the various sludge treatment phases. Nitrogen recovery was feasible from two stages: the mechanical dewatering of sewage sludge digestate (16%, 780 t_$N_{tot}$/a) and from the thermal drying of dewatered sewage sludge digestate (3%, 270 t_$N_{tot}$ /a). The combustion scenario resulted in 120 GWh/a of district heat and 9,700 t/a of ash with 500 t/a phosphorus, while the pyrolysis scenario produced 12,000 t/a of biochar with 500 t/a phosphorus. The addition of a stripper and a scrubber for nitrogen recovery increased the total electricity consumption in both scenarios. The annual investment costs for combustion and pyrolysis were estimated to be 2–4 M €/a and 2–3 M €/a, respectively, with projected product revenues of 3–5 M €/a and 3–3.5 M €/a. These results answer the RQ2 that integration of pyrolysis and combustion of sewage sludge with scrubbing using acidic media such as $H_2SO_4$ can recover $NH_3$ from reject water from dewatering of sewage sludge and exhaust fumes of thermal drying. In pyrolysis, addition to nitrogen recovery, biochar is also produced which can be used for soil amendment and as adsorbent to recover nitrogen. Similary, in combustion, in addition to nitrogen recovery, heat energy is also produced which can be used for district heating. The results highlights that integration of above mentioned technology is optimal in terms of nitrogen recovery and generation of value added products such as biochar and heat energy.

The third part of this study focused on the environmental impact assessment of N recovery from dewatered sewage sludge digestate in Finland. The study found that combustion or composting of dewatered sewage sludge outperformed pyrolysis in terms of environmental impact. Combustion reduces sludge volume and produces ash that can be used as a nutrient-rich fertilizer. The primary consequences were caused by sludge treatment operations, while transport emissions had minimal significance (except for toxicity categories). In composting, air emissions and peat use had significant impacts across categories, but also led to emission reductions by replacing peat-containing gardening soil. In the combustion scenario, N recovery from condensate emissions outweighed the fertilizer production benefits, indicating improved efficiency and reduced process chemistry requirements. Wood waste can be used for pyrolysis without extra fuel, however it cannot be used for district heating. Due to uncertainty in full-scale process performance, heavy metal fate, and carbon, pyrolysis was the least chosen choice in multiple impact categories. Important technology parameters in pyrolysis include the ratio of wood waste to sludge, which produces enough pyrolysis gas for drying, combustion, flue gas purification, and electricity consumption.

Combustion or composting dewatered sewage sludge was more environmentally friendly than pyrolysis. Composting is viable if the remaining compost can be used. Ash from combustion is a nutrient-rich fertilizer and reduces the sludge volume. Sludge treatment operations have the main impact, while transport emissions had little effect in the toxicity categories. Air emissions and peat consumption affected composting across categories, although substituting peat-containing gardening soil reduced emissions. N recovery from condensate emissions surpassed the fertilizer production benefits in the combustion scenario, improving efficiency and reducing the process chemistry. Wood waste can be used for pyrolysis without extra fuel; however, it cannot be used for district heating. Due to uncertainty concerning the full-scale process performance, heavy metal fate, and carbon, pyrolysis was the least chosen choice in multiple impact categories. However, the wood waste-to-sludge ratio creates adequate pyrolysis gas for drying, combustion, flue gas purification, and electricity consumption. More process data is needed to analyse the environmental impact of pyrolysis. The LCA did not examine crucial environmental impacts such as medication residue dispersion, bacterial resistance, and microplastic dissemination. Composting may not be feasible if potential consequences are to be avoided.

The final part of the study assessed the environmental performance of nitrogen recovery for fertilizer purposes from sewage sludge treatment. Three different scenarios were investigated: one without nitrogen recovery and two with nitrogen recovery using air stripping/scrubbing and pyrolysis-derived biochar adsorbent. The results showed that targeting both reject water and thermal drying fumes for nitrogen recovery improved the total recovery rate. The scenarios performed differently in different environmental impact categories. PIV-S3 (adsorption) performed better in three of the five impact categories considered, including climate change with biogenic carbon, freshwater eutrophication, and marine water eutrophication. Concerning climate change including biogenic carbon, carbon capture as a biochar was the most important factor in all impact categories. The

third and fourth part of this study answers RQ3 that comparison of environmental impacts of nitrogen recovery from sewage sludge in Finland, finding combustion and composting more eco-friendly than pyrolysis. Combustion produced nutrient-rich ash and reduced sludge volume, while composting had air emission and peat use impacts but also led to emission reductions. Nitrogen recovery scenarios differed in environmental impact categories, with adsorption outperforming in climate change including biogenic which also includes carbon capture as biochar, freshwater eutrophication and marine water eutrophication.

In conclusion, the study provided valuable insights into the feasibility, environmental impacts, and economic implications of recovering ammonia and nitrogen from sewage sludge treatment processes. While certain recovery methods showed promise in terms of efficiency and resource utilization, further research is needed to optimize the processes and reduce investment costs. The study emphasizes the potential benefits of integrated resource recovery in wastewater treatment systems for addressing global environmental concerns and promoting sustainable practices.

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# Publication I

Saud, A., Havukainen J., Mänttäri M., and Horttanainen M.
**Evaluation and techno-economic analysis of packed bed scrubber for ammonia recovery from drying fumes produced during the thermal drying of sewage sludge**

# Evaluation and techno-economic analysis of packed bed scrubber for ammonia recovery from drying fumes produced during the thermal drying of sewage sludge

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**Abstract.** This study investigates the recovery of ammonia from drying fumes during thermal drying of sewage sludge with packed bed acid scrubbers to recover ammonia and to produce ammonium sulfate. The process is modelled for two concentrations, 75 and 100 ppm, and 1000m$^3$/h inlet flowrate of drying fumes containing air and ammonia gas. It results in finding optimal parameters for scale-up of drying fumes during thermal drying of 7700t/a sewage sludge of Lappeenranta city. It is found that a single scrubber, with a 24000 m$^3$/h of inlet gas and an ammonia concentration of 75 ppm, liquid to gas ratio of 1.5, temperature and pH of liquid acid as 100 ℃ and 3 respectively, gives the efficiency of more than 99%, and reduces ammonia concentration in the outlet stream to 0.2 ppm. The capital cost is 290 k€, operating cost is 113k€/a, removal cost with and without revenue of ammonium sulfate is 20 €/t and 18 €/t of sludge. The packed bed acid scrubber would be suitable to remove ammonia in the drying fumes to recover ammonia from the drying fumes, but the initial economic analysis highlights that the production of commercial grade ammonium sulfate fertilizer would be an expensive option.

## 1. Introduction

The population of the world is increasing and is predicted to reach 8-10 billion in 2050. This increase in population results in a high demand for food production [1]. Given this, the demand for fertilizers is also increasing by 4% annually to support food needs for an additional 2.3 billion people in 2050. Thus, the production of sustainable fertilizers is the need of the hour [2].

Nitrogen and phosphorus are the two unique elements of agriculture. Annually, 120 million tons of atmospheric nitrogen is converted into reactive nitrogen for the production of fertilizers by the Haber Bosch process, but the high temperature and pressure requirements for reactions consume a large amount of energy: it has been estimated that 1kg of liquid ammonia requires 42MJ of energy, and emits 1.9 kg of $CO_2$ [3]. Sludge is, however, a global growing waste problem and, at the same time, a potential source of recoverable nutrients. Therefore, in many countries, it is incinerated [4], which is an effective disposal method but easily destroys the nutrients. Sewage sludge contains nutrients such as phosphorus and nitrogen along with harmful substances. Due to this fact, sewage sludge utilization poses the risk of soil contamination directly or after treatment [5,6]. During the thermal drying of sewage sludge, a large amount of ammonia combusted to $N_2$ and NOx which

can be recovered. Previous research [7], proposed ammonia recovery from the drying fumes resulting from the thermal drying of mechanically dewatered sewage sludge. The ammonia exiting as fumes during thermal drying can be absorbed with sulfuric acid or nitric acid to produce ammonium sulfate and ammonium nitrate, which are commercially used fertilizers.

Wet acid scrubbers have shown remarkably efficient ammonia recovery, between 91 and 99%, from exhaust air in animal facilities [8]. The application of ammonia scrubbing has been studied in different animal facilities, including poultry and swine farms, to neutralize gas emissions. Scrubbers have been installed in animal facilities in which the ammonia in exhaust air reacts with dilute sulfuric acid to produce ammonium sulfate [9] . The design of scrubbers have been widely studied and optimized in many studies, details of which are given in [8,10,11]. On the other hand, the absorption of ammonia to produce a commercial product such as ammonium sulfate fertilizer from the drying fumes produced during the thermal drying of sewage sludge has been little studied.

This study is part of the ongoing effort to find ways to recover nitrogen from mechanically dewatered sewage sludge during its thermal drying phase. The aim of this study is evaluating the possibilities and feasibility of acid scrubbing for the recovery of ammonia from sludge drying fumes for fertilizing purposes by changing

different parameters such as pH, liquid flow rate and temperature and by conducting initial economic analysis.

## 2. Methods

### 2.1 Process Description

A process flow diagram of scrubbing system with inlet and outlet streams is shown in Fig. 1. The air containing ammonia enters from the bottom of the scrubber at atmospheric pressure as stream GASIN, and sulfuric acid enters as stream H2SO4 and water enters as stream LIQIN from the top of scrubber. From the top of the scrubber, the EXHAUST stream vents out air to ambient environment after treatment, and the LIQOUT stream, comprised of water and ammonium sulfate, leaves the scrubber from the bottom. The LIQOUT stream further goes into splitter where 50% of liquid goes in RECYCLE stream to circulate again with the diluted acid and remaining goes to PRODUCT stream. A MIXER combines streams before entering scrubber to stream LIQMAI and liquid ammonium sulfate comes out in stream AMMLIQ. Equilibrium reactions are considered, and the required equilibrium data is taken from the literature and the ASPEN databank. Acid scrubbing of ammonia gas is modeled in the ASPEN Plus simulator. ASPEN Plus also supports estimations of process behavior by applying engineering knowledge. The Radfrac column with rate-based calculation method is selected as a suitable option to design scrubber for vapor liquid streams, equilibrium and rate based reactions. [11]. The electrolyte NRTL (ENRTL) method is selected for thermophysical property analysis to determine the thermodynamic properties of the liquid phase, including electrolytes [12,16].
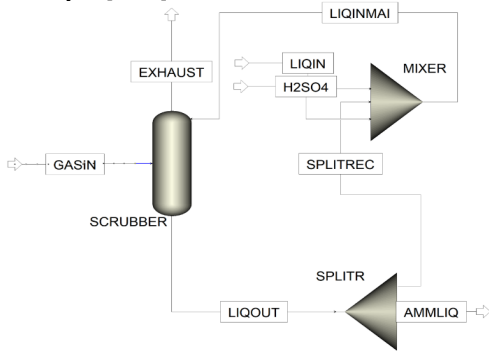


**Fig. 1.** A simplified representation of a scrubber in ASPEN Plus V9.

### 2.2 Reactions

In modeling of scrubber, the reactions shown in Table 1 are considered. For the calculation of equilibrium constant K for reactions, the following rate equation was used for calculation [17]:

$$lnK = A + \frac{B}{T} + C\,ln\,T + DT \qquad (1)$$

**Table 1.** Equilibrium reactions included in the model.

| Reaction | Type of Reaction | Chemical Equation |
|---|---|---|
| R1 | Equilibrium | $H_2O + HSO_4^- \leftrightarrow H_3O^+ + SO_4^{-2}$ |
| R2 | Equilibrium | $H_2SO_4 + H_2O \leftrightarrow H_3O^+ + HSO_4^-$ |
| R3 | Equilibrium | $NH_3 + H_2O \leftrightarrow OH^- + NH_4^+$ |
| R4 | Equilibrium | $2H_2O \leftrightarrow OH^- + H_3O^+$ |
| R5 | Salt | $(NH_4)_2SO_4 \leftrightarrow NH_4^+ + SO_4^{-2}$ |

where, T is the absolute temperature and A, B, C, and D are equilibrium parameters. Their values are available in the ASPEN databank, as shown in Table 2.

**Table 2.** Reaction parameters for the equilibrium constant K.

|  | A | B | C | D |
|---|---|---|---|---|
| R1 | -5.393 | $1.73 \times 10^3$ | 0 | 0 |
| R2 | -3.898 | $3.47 \times 10^3$ | 0 | 0 |
| R3 | -1.257 | -3335 | 1.497 | -0.037 |
| R4 | 132.9 | $-1.34 \times 10^4$ | -22.477 | 0 |
| R5 | -216.6 | $4.26 \times 10^3$ | 37.518 | -0.0799 |

### 2.3 Data for modelling

Two studies were selected as a reference for the preliminary design of the scrubber Melse and Ognik et al. [8] and Khakharia et al. [16]: The first one was the work of Melse and Ognik et al. [8], in which acid packed bed scrubbers were developed for the removal of ammonia ($NH_3$) from pig and poultry facilities. It was concluded in the study that $NH_3$ removal was in the range of 40% to 100% with an average value of 96%  and the second study was work of Khakharia et al. [16] which describes an acid scrubber to treat ammonia emissions from a post-combustion $CO_2$ capture plant and in this study, the ammonia inlet concentration of 150 mg/m$^3$, and the ammonia outlet concentration decreased to 5 mg/m$^3$. These two studies are used to estimate required parameters as listed in Table 3, to evaluate the model and configuration, cost estimation, and scale-up of ammonia scrubber for the drying fumes produced by sewage sludge. The selected data for validation is mentioned in Table 3.

**Table 3.** Selected process parameters for modeling.

| Parameter | Values |
|---|---|
| Flow Rate of Gas | 1000 m$^3$/h |
| Inlet concentration of Ammonia | 75 and 100 ppm |
| H$_2$SO$_4$ pH | 1,2,3,4,5 |
| Liquid to Gas Ratio (L/m$^3$) | 4.3 |
| Liquid Flow Rate | 4025 kg/h |
| Inlet Temperature | 100 °C |
| Pressure | 1 atm |
| Superficial Velocity | 1.4 m/s |

The proposed scrubber is validated with the data from Khakharia [16] and Melse [8]. In first validation, liquid flow rate and inlet concentration of ammonia is varied to analyze the outlet concentration of ammonia. pH, gas

flow rate and the temperature is kept constant while in second validation, pH is varied to analyze the ammonia removal efficiency of scrubber. Liquid flow rate, gas flow rate and temperature are kept constant. The selected packing material is Mellapackplus 252Y packing and liquid to gas (L/G, L/m$^3$) ratio is 4.3 [8,16].

## 2.4 Scale-up and Cost Estimation

Scale up is needed to investigate the initial feasibility of acid scrubbing for ammonia removal. The scale up includes the calculation of scrubber dimensions. The height of the column, packing height, and diameter of the scrubber were calculated as follows:

The height of the scrubber and packing is calculated with Eq. 2 and Eq. 3 [10]:

$$H_{column} = 1.4H_{pack} + 1..02D + 2.81 \qquad (2)$$

where, H$_{column}$ is the height of the scrubber, H$_{pack}$ is the height of the packing, and D is the diameter of the scrubber.

$$H_{pack} = N \times HETP \qquad (3)$$

where, N is the number of equilibrium stages, and HETP is the height equivalent to theoretical plate.
The superficial velocity, u$_s$ is estimated as 1.4 m/s [8], and diameter, D$_c$ was calculated from:

$$D_c = \frac{\sqrt{4A}}{\pi} \qquad (4)$$

where, A is the area of the scrubber calculated from Eq. 5, and the value of $\pi$ was taken as 3.14.
The area is calculated from Eq. 5:

$$A = \frac{Q}{u_s \times 3600} \qquad (5)$$

where, Q is the flow rate of gas in m$^3$/h.

## 2.5 Cost Estimation

In cost estimation, the method cost curves and equations for preliminary estimation is considered due to unavailability of empirical data. The method of cost calculation for equipment cost, and different percentages of variations have been taken from literature [18,19]. Eq. 6 is used to calculate the mass of the scrubber and cost of equipment purchased:

$$C_e = a + bS^n \qquad (6)$$

where, C$_e$ = Cost of equipment purchased, €
a, b = cost constants and values, taken as 11600 and 34 respectively [18]
S = size parameter (Shell mass, kg)
n = exponent for equipment, value 0.85 [18]
The shell mass of the scrubber, S, is the size parameter to calculate purchase cost. It is calculated from Eq. 7:

$$S = \pi D_c H_c t_w \rho \qquad (7)$$

where, D$_c$ = vessel diameter, 2.4 m
H$_c$ = vessel height, 12 m
t$_w$ = wall thickness, 0.009 m
$\rho$ = metal density, 8000 kg/m$^3$
The values of wall thickness, t$_w$ and metal density, $\rho$ are taken from literature [18].

Other investment includes costs for installation, instrumentation, piping, electrics, and costs related to engineering, construction, and services, are also calculated from factors given in literature [34] and multiplying with cost of purchased equipment, C$_e$. The operational cost includes the cost of sulfuric acid and water, these being 0.4 €/kg and 1 €/m$^3$ respectively.

## 3. Results and Discussion

### 3.1 Model Validation

Model validation is required to verify the designed model consistency and to analyze behavior of system. The process of model validation in this study is based on the data from literature.

In Fig.2(a), the designed model behaved in the similar way as in the referenced model. In the work of Khakharia [16] and in this current study, the inlet concentration of ammonia was varied from 152-155 mg/m$^3$ (218-223 ppm) and outlet concentration was measured below 2 mg/m$^3$ (3 ppm).
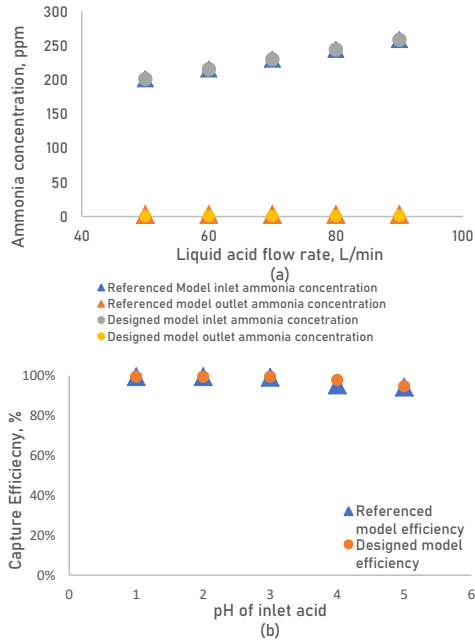


**Fig. 2. a)** Validation off referenced and modeled scrubber with changing liquid flow rate and ammonia concentration **b)** effect of changing pH on efficiency of scrubbers.

In the second case of validation, the efficiency of both referenced and modeled scrubber were above 99%. In Fig.2(b), it can be seen clearly seen that with the lower pH, the efficiency of both scrubbers is above than 99% and as pH increases, the efficiency started to decrease due to lower concentration of acid.

### 3.2 Effect of pH

pH is very substantial in the absorption of ammonia, and according to literature should be kept in the range of 1-5 for acid scrubbers [8,16,20]. The lower water flow rate assists in ammonia capture. Hence, the pH of acid is varied from 1-5 to analyze changes in the capture of ammonia and the efficiency of the scrubber. The high solubility of ammonia in water and establishing reaction equilibrium between gas and liquid phase in absorption led to the use of sulfuric acid to keep the ammonia in an ionized form [16].

The water flow rate, temperature, and gas flow rate were kept constant at 70 L/min, 100 °C, and 1000 m$^3$/h, and only the pH of acid varied from 1 to 5 to estimate the effect of pH. Two inlet concentrations of $NH_3$ in the inlet of scrubber, 75 ppm and 100 ppm, were studied. Fig. 3 shows that the pH had no effect on efficiency of scrubber from pH 1-3, but that efficiency started reducing at pH 4 and pH 5. The ammonia gas outlet concentration was declined to 0.2 ppm and 0.3 ppm at pH 1 to 3 for both inlet concentration of 75 and 100 ppm respectively.

On the other hand, at pH 4 and 5, the ammonia concentration reduced to 0.9 ppm and 8.6 ppm from 75 ppm respectively, while on 100 ppm it was reduced to 2.3 and 12.5 ppm respectively as shown in Fig. 3. The observed efficiencies are higher than 99% but for pH 5, this reduced to 88%. The same trend was observed in the study of Khakharia et al.[16] and Melse and Ognik [8] have also suggested a pH range from 1-4.
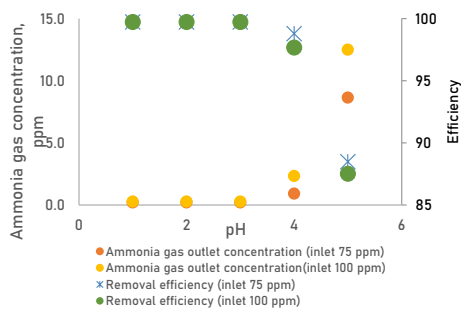


**Fig. 3.** Outlet gas ammonia concentration and removal efficiency with varying pH of inlet sulfuric acid.

### 3.3 Effect of Flowrate

The appropriate L/G ratio is always required for the efficient flow of liquid through selected packing. The L/G ratio promotes mass transfer between gas and liquid phases [8,21].

L/G ratio is the reason that the flowrate of liquid is varied between 70 and 150 L/min (L/G ratio of 4.3-8) to evaluate the effect of flowrate on ammonia removal and scrubber efficiencies. The gas flow rate is fixed at 1000m$^3$/h, pH of acid at 3, temperature at 100 °C, and only the liquid flow rate was varied in the simulation.
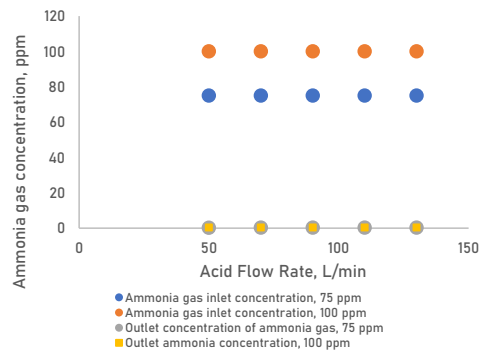


**Fig. 4.** Effect of inlet acid flow rate on the outlet concentration of ammonia for both 75 and 100 ppm ammonia concentration

In Fig. 4, the variation in flow rate has had little effect on the removal of ammonia because, at the lowest flow rate, 70 L/min, the efficiency of ammonia removal is greater than 99%. It is also noticeable that if the concentration of ammonia is increased, the liquid flow is sufficient to remove the additional ammonia to the desired level.

### 3.4 Presence of ammonium sulfate

Ammonium sulfate is present in a very diluted form in the liquid outstream of the scrubber. The presence of ammonium ions ($NH_4^+$) and sulfate ions ($SO_4^{-2}$) in the liquid outlet stream confirms the existence of ammonium sulfate. The production (or concentration) of ammonium ions varies with the concentration of inlet ammonia gas, and the concentration of sulfate ions varies with the pH of the liquid. Higher the inlet concentration of ammonia gas, the more ammonium ions will be produced. In Figs. 5(a) and 5(b), it shows that the selection of pH 3 is very favorable for a higher amount of ammonium ions as the mass flow rate of $NH_4^+$ ions is about 0.06 kg/h, whereas $SO_4^{2-}$ ions is 0.43 kg/h respectively. In Fig. 5(b), the dissociation of acid decreases with increasing pH, resulting in a decreased amount of $SO_4^{2-}$. Similarly, $NH_4^+$ also shows a low dissociation behavior at pH 4 and 5.

### 3.5 Scale-Up and Cost Estimation

The scale-up of acid scrubber is done on the sewage sludge production rate of Toikansuo wastewater treatment plant, situated in the small Finnish town of Lappeenranta, treats wastewater with a capacity of 16000 m$^3$/day for 72,000 habitants. The total sludge 7700 t/a with 20% total solids and a total nitrogen content of 5.3% of TS, and, on average 12% of the total nitrogen ended up in drying fumes as ammonia [7]. It summarizes that

one scrubber with a flow rate of 24000 m$^3$/h and 8610 kg-Nitrogen/a of ammonia with concentration of 75 ppm swill be treated.
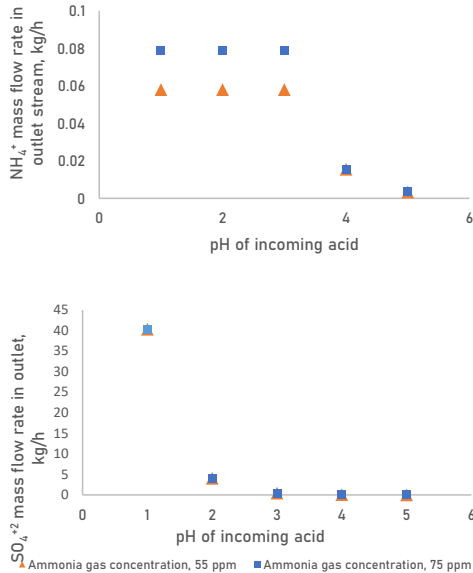




▲ Ammonia gas concentration, 55 ppm   ■ Ammonia gas concentration, 75 ppm

**Fig. 5.** Mass flow rate at different pH levels of **(a)** NH4+; and **(b)** SO4-2 ions in liquid ammonium sulfate outlet stream.

Table 4 summarizes the cost of a scrubber, packing, and selected material whereas Table 5 summarizes process specifications, the number of scrubbers required, and the dimensions of a scrubber.

**Table 4.** Dimensions of scrubber.

| Specifications | Values |
|---|---|
| Diameter of scrubber | 2.4 m |
| Height of scrubber | 12 m |
| Packing height | 6 m |
| Required number of scrubbers | 1 |

**Table 5.** Cost calculated for scrubber.

| Equipment | Material | Cost (k€) |
|---|---|---|
| Scrubber | Stainless steel | 78 |
| Packing (Sulzer Mellapakplus 252Y) | SS 304L | 21 |
| Erection and commissioning cost | | 165 |
| Start-up cost | | 26 |
| **Total** | | **290** |

The total capital cost for scrubber with the capacity of handling 24000 m$^3$/h of gas and 575 L/min of liquid is 290 k€. In operational cost, sulfuric acid and water are the main contributor with cost of 14 k€/a and 99 k€/a. We assume, that the scrubber can be operated by the personnel of the sludge treatment plant and so it does not increase the personnel costs.

In Fig.6, a graph between liquid to gas ratio (L/G) and recycling rate is made to predict the best economical cost of water. Firstly, liquid to gas ratio is changed from 1.5 to 4.3 and recycling rate kept constant at 50% and then L/G ratio kept constant at 1.5 and recycling rate was varied from 50-70 %.
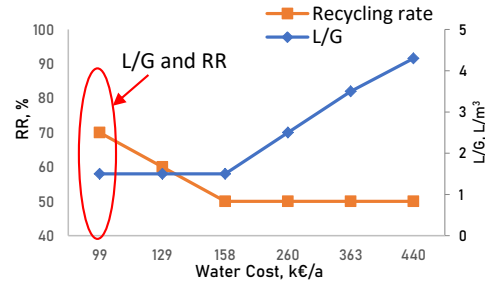


**Fig. 6.** Relation of water cost to recycling rate (RR) and liquid-to-gas ratio (L/G).

When recycling rate is varied above 70%, simulation started giving errors due to flooding limit of 80%. The encircled values show that the 70% recycling rate and 1.5 liquid to gas ratio gives optimal results with lower cost of water. It helps in calculation of operational cost of sulfuric acid and water which is 14 k€/a and 99 k€/a respectively with total of 113 k€/a.

Table 6 gives further details of the per-ton cost of treated sludge and possible income generated from recovered nitrogen by using it in fertilizer production. The value of mineral fertilizers in terms of nitrogen is 1.6 €/kg-Nitrogen [22]. The annual nitrogen production from recovered ammonia (based on 99% recovery) is calculated as 8610 kg-Nitrogen/a. The expected production of ammonium sulfate is 9 t/a and the cost of ammonia recovery per ton of sewage sludge treated would be 18 €/ton. The total annual cost is calculated as 155 k€ taking into account an interest rate of 10% and a 10-year lifetime for scrubbers.

**Table 6.** Annual cost and expected income generation

| Recovery | Cost |
|---|---|
| Income from recovered nitrogen | 14 k€/a |
| Total annual cost | 155 k€/a |
| Additional cost for sludge treatment | 20 €/t of sludge |
| Cost of ammonia recovery | 18 €/t of ammonium sulfate |
| Cost of ammonia recovery | 18 €/kg of nitrogen |

## 4 Conclusions

A preliminary study of acid scrubber to recover ammonia from the drying fumes produced during the thermal drying of sludge was conducted including modeling and cost estimation. The ASPEN Plus simulations were conducted for ammonia concentrations of 75 and 100

ppm. It was observed that low pH values between 1-3 are suitable for ammonia recovery, but with higher pH such as 4-5, efficiency would decrease from 99% to 88%. The L/G ratio was changed by changing the liquid acid flow rate and value of 1.5 with recycling rate of 70% was found to be a feasible L/G ratio.

The total capital investment of a scrubber for a sludge treatment plant with capacity of 7 700 ton/a, was calculated as 290 k€ with an operational cost of 113 k€/a, including the cost of sulfuric acid and water. The cost of ammonia removal is 20 €/t of sludge and the cost of ammonium sulfate produced is 18 €/t of sludge which is practical treatment cost, but product is very diluted as compared to commercial grade and cost is high for optional source of ammonia fertilizer production and treatment.

The result is disproving the previous proposals that this kind of scrubbing could be feasible not only for ammonia emission reduction but also for production of ammonia fertilizer. If the costs will be covered by sludge treatment costs and costs, it can be possible to utilize the end product in some cases. But the additional costs for refining the product to fertilizer seem to be too high to compete with other nitrogen fertilizers and more cost-effective methods are needed. Further research by the authors will focus on finding more suitable and feasible methods to recover ammonia from the fumes produced by the thermal drying of sewage sludge.

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# Publication II

Saud, A., Havukainen J., Peltola P., and Horttanainen M.
**Integrating pyrolysis or combustion with scrubbing to maximize
the nutrient and energy recovery from municipal sewage sludge**

*Article*

# Integrating Pyrolysis or Combustion with Scrubbing to Maximize the Nutrient and Energy Recovery from Municipal Sewage Sludge

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**Abstract:** Based on mass and energy balance calculations, this work investigates the possibility of recovering heat and nutrients (nitrogen and phosphorus) from municipal sewage sludge using pyrolysis or combustion in combination with a gas scrubbing technology. Considering a wastewater treatment plant (WWTP) with 65,000 t/a of mechanically dewatered digestate (29% total solids), 550 t/a nitrogen and 500 t/a phosphorus were recovered from the 4900 t/a total nitrogen and 600 t/a total phosphorus that entered the WWTP. Overall, 3600 t/a (73%) of total nitrogen was lost to the air (as $N_2$) and clean water, while 90 t/a (15%) of total phosphorus was lost to clean water released by the WWTP. Both in combustion and in pyrolysis, the nitrogen (3%) released within thermal drying fumes was recovered through condensate stripping and subsequent gas scrubbing, and together with the recovery of nitrogen from WWTP reject water, a total of 3500 t/a of ammonium sulfate fertilizer can be produced. Furthermore, 120 GWh/a of district heat and 9700 t/a of ash with 500 t/a phosphorus were obtained in the combustion scenario and 12,000 t/a of biochar with 500 t/a phosphorus was obtained in the pyrolysis scenario. The addition of a stripper and a scrubber for nitrogen recovery increases the total electricity consumption in both scenarios. According to an approximate cost estimation, combustion and pyrolysis require annual investment costs of 2–4 M EUR/a and 2–3 M EUR/a, respectively, while 3–5 M EUR/a and 3–3.5 M EUR/a will be generated as revenues from the products.

**Keywords:** combustion; energy recovery; nutrient recovery; pyrolysis; sewage sludge; thermal treatment; waste to energy

## 1. Introduction

The global population is estimated to increase from 7.7 billion to 9.7 billion by 2050 [1]. This expected rapid increase, with the concentration of people in cities, raises several problems in terms of waste, particularly sewage sludge, which is the semi-solid byproduct generated during municipal and industrial wastewater treatment [2]. Billions of tons of sewage sludge and wastewater are produced every year, with the final disposal depending on the country's policies and regulations [3]. The current disposal pathways of sewage sludge in the EU include combustion (27%), agricultural fertilizer (42%), landfill (14%), and other applications (17%) [4].

Sewage sludge contains nutrients such as phosphorous and nitrogen [5]. However, it also contains harmful substances, including heavy metals, aromatic hydrocarbons, pathogens, and, as was recently discovered, microplastics and medical residues. Due to this fact, sewage sludge utilization as it is, and even after biological treatment, risks soil contamination [6].

Nutrients, mainly nitrogen and phosphorus, are crucial for the growth of plants and are thus in high demand, specifically as fertilizers [7]. About 75% of the remaining phosphorus reserves are located in Morocco and, according to one estimate, these will

be depleted within 45 to 100 years [8–10]. Phosphate, as a critical raw material, has received increased interest in the recovery and treatment of waste, specifically wastewater, to regenerate phosphorus using current technologies.

Nitrogen is a vital nutrient for all living organisms. While nitrogen is readily available in the atmosphere in the form of nitrogen gas ($N_2$), most plants cannot consume it due to the nonreactivity of molecular nitrogen. Some plants and crops use biological nitrogen fixation to absorb $N_2$ in the form of ammonium or nitric oxide, but nitrogen fertilizers are needed to increase the crop yield [11].

The Haber–Bosch process was developed to transform stable $N_2$ into nitrogen fertilizers to boost crop production [12]; however, producing 1 ton of ammonia ($NH_3$) fertilizer consumes about 35–50 MJ/kgN of energy and 950 $m^3$ of natural gas, while emitting 1.6 tons of carbon dioxide ($CO_2$). However, only about 17% of the nitrogen produced in this way is ultimately consumed by humans through food sources, and the rest is lost to the environment [13]. This reactive nitrogen abundantly introduced to the ecosystem represents a significant source of pollution [14]. Although it has been possible to increase food production through the use of fertilizers, this has led to the disposal of nitrogen in the form of urea and $NH^{4+}$ via human defecation, ending up in sewage [15]. This, in addition to other nitrogen leakages from crop production systems, such as the leaching of nitrogen from fields, causes eutrophication in water bodies [16].

A wastewater treatment plant (WWTP) is an industrial facility that removes pollutants through a series of operations, including the mechanical, chemical, and biological treatment of incoming wastewater. Wastewater treatment is an energy intensive process in which about 90% of total energy demand is in the form of electric energy. Furthermore, 50–60% of operating costs are taken by sludge treatment whereas electric energy takes a 25–40% share of total operating costs [4]. Therefore, efficient energy and nutrient recovery from a WWTP can lead to a better efficiency and reduced costs [17].

Nitrogen is first removed from an aeration tank via nitrification and denitrification, which convert ammonium and nitrate, respectively, into $N_2$, releasing it into the atmosphere. The remaining nitrogen and phosphorus are bound in the sludge, which advances towards sludge treatment. Anaerobic digesters are one of the most common sludge treatment techniques, whereby the sludge is anaerobically digested to produce biogas, which is utilized in energy generation. The remnants of the process are mechanically dewatered, producing dried sludge and reject water [18].

The availability of organic matter in wastewater makes it an energy source from the thermodynamic perspective. After wastewater treatment, sewage sludge has an energy content of about 60% in the form of organic carbon, making it a potential energy source [19]. Moreover, sewage sludge is continuously generated during all stages in a WWTP. Sewage sludge contains organic pollutants, which can undergo thermochemical treatment while reducing the volume and producing energy. Combustion, pyrolysis, and gasification are three main thermochemical treatment methods, with the selection depending on the required temperature range and desired products. Figure 1 summarizes the thermal treatment options and their products.

In pyrolysis, the organic matter of sewage sludge is thermally degraded at high temperatures in the absence of oxygen. The products are bio-oil, biochar, and syngas. Biochar, which is also produced from biomass such as wood, is used mainly as a fuel, but it can also act as a low-cost adsorbent to remove pollutants in the form of pharmaceutical compounds, heavy metals, organic dyes, and phenolic compounds [20,21]. Ammonia inhibition and acid stress in microbes during anerobic digestion are reduced by increasing alkalinity and pH of solution which improves microbial colonization and results in increased yield of methane and digestate quality [22]. The yield of products depends on temperature, pressure, residence time, heating rate, sludge properties, and particle size. Pyrolysis is further classified in slow, fast, and flash pyrolysis. Slow pyrolysis uses temperatures ranging between 300 and 700 °C and a long residence time at a slow heating rate to maximize the yield of biochar [23]. In fast pyrolysis, the heating rate is 10–200 °C/s and the residence

time is between 0.5 and 10 s to increase the yield of bio-oil. In flash pyrolysis, the heating rate is between $10^3$ and $10^4$ °C/s and the residence time is less than 0.5 s, with the bio-oil yield slightly higher than in fast pyrolysis [24].
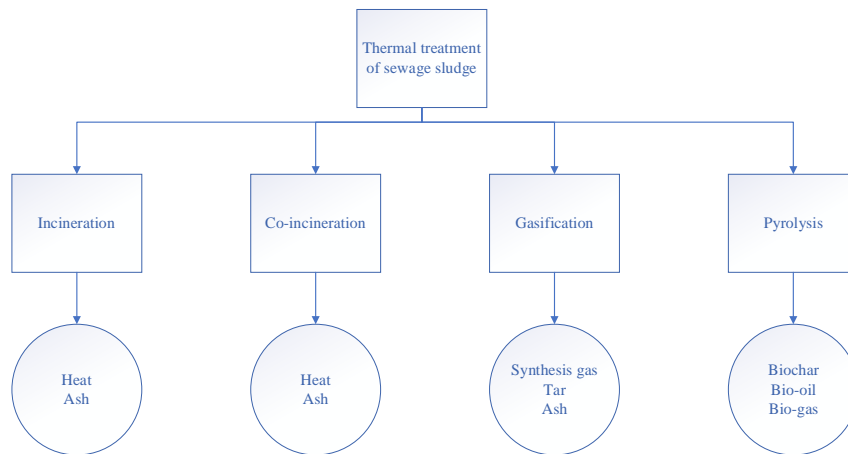


**Figure 1.** Sewage sludge thermal treatment classification.

In combustion, sewage sludge is thermally decomposed at elevated temperatures between 850 and 1300 °C [25], generating heat and electricity that can be utilized for several operations. Meanwhile, so-called bottom ash is extracted from the bottom of the furnace, and metals can be recovered, while the fly ash escaping with the flue gas can also be captured, e.g., by bag filters or wet scrubbers [25,26]. Gasification based on the partial oxidation of the organic solids produces synthesis gas that can be utilized for energy production; however, this process is not further described here as it is not considered as a treatment option.

Thermochemical treatment requires a lower moisture content, but the moisture content of sewage sludge is still 73–84% after mechanical dewatering [25,27]. A moisture content too high can decrease the combustion temperature, meaning that the EU Waste Incineration Directive's minimum required temperature of 850 °C is not met. The high amount of moisture further introduces complexities into the pyrolysis process, such as the formation of high-liquid products and an increase in non-condensable gases [28,29]. Thermal drying, usually utilizing heat from the combustion of the sludge or non-condensable (odorous) gases, offers a solution to reduce the sludge moisture content to 5–10%, reducing the complexities and increasing the product quality [25,30].

Previous research indicates that nitrogen recovery (instead of simple removal) is difficult to justify economically [31]. Hence, the efficiency should be improved, and possibilities should be found to at least decrease the costs of nitrogen capture. Single technical solutions, particularly for sludge or wastewater flow, are expensive compared to the yield and value of the utilizable product. Integrating nitrogen capture from more than one stream could improve the total recovery rate and make the investment less costly. For this reason, the current study focuses on the integration of pyrolysis or combustion with nitrogen recovery in the WWTP sludge treatment process to maximize the recovery of nutrients and energy for cases in which biological treatment is insufficient to remove harmful substances from the sludge. It also aims to evaluate the increase in the value and quality of products from sewage sludge by using mass and energy balance calculations and approximating the costs and revenues of each option.

## 2. Materials and Methods

Two different sludge treatment and recovery processes currently being piloted in Finland, namely pyrolysis and combustion, are selected as the scenarios for analysis in this study. The pyrolysis-based process produces biochar, destined for use as an additive in composting. The other process is based on thermal drying and subsequent combustion of sludge, producing district heat and nutrient-rich (mainly P and Ca) ash, which is used as forest fertilizer. Neither process currently incorporates nitrogen recovery in the pilot plants. In this study, comparability is achieved by using the same sludge input flow for both processes (although the pilot plants and WWTPs have very different sizes).

This study focuses on the following scenarios:

S1.0: Pyrolysis without N recovery.
S1.1: Pyrolysis with N recovery from thermal drying fumes but not from WWTP reject water.
S1.2: Pyrolysis with N recovery from both thermal drying fumes and WWTP reject water.
S2.0: Combustion without N recovery.
S2.1: Combustion with N recovery from thermal drying fumes but not from WWTP reject water.
S2.2: Combustion with N recovery from both thermal drying fumes and WWTP reject water.

Scenarios S1.0 and S2.0 are the base scenarios without the recovery of nitrogen. In the combustion base scenario, part of the fuel energy is recovered and utilized as district heat and the ash product is used as a forest fertilizer. The pyrolysis base scenario uses the sewage digestate in biochar production, and other pyrolysis products are combusted to produce the heat energy required for the thermal drying and pyrolysis process. The pyrolysis process additionally utilizes wood waste (to ensure energy self-sufficiency); in the combustion scenario, this waste is directed towards additional district heat production.

The technical data from the two pilot plants are used in this study. In the pyrolysis scenario, the sewage digestate obtained from the WWTP is pyrolyzed with wood (waste) to produce biochar. In the combustion scenario, the sewage sludge is combusted, and the excess energy is used for district heating. The comparison between the different technologies is enabled by applying the technical performance values for the common sludge mass flow, namely the sludge mass flow of the Viikinmäki WWTP in Helsinki, operated by Helsinki Region Environmental Services (HSY).

### 2.1. Wastewater Treatment Plant, Sludge Digestion, and Dewatering

In the WWTP plant, wastewater enters with a flow rate of $92 \times 10^6$ m$^3$/a with $29 \times 10^6$ kg/a total solids (TS). The plant potentiality in terms of population equivalent is 3 m$^3$/day/person. Nitrogen and phosphorus are the two primary nutrients, with mass flow rates of 4900 t/a and 600 t/a, respectively. After the wastewater treatment processes of pre-treatment, screening, and aeration, the raw sludge enters anaerobic digestion, resulting in biogas and digestate. The biogas is used for heat and electricity production, whereas the remaining digestate goes to dewatering. The amount of reject water is calculated based on the difference in the mass of the digestate and dewatered sludge.

The total energy consumption of the WWTP is 0.42 kWh/m$^3$. After anaerobic digestion, the sewage sludge has a 95% moisture content, and sequential moisture removal processes are key to thermally treating sludge. Thermal drying is an energy-intensive process; therefore, dewatering is performed beforehand to reduce the energy consumption [25]. Dewatering of the digestate helps to reduce the volume of the sludge by removing water, simplifying transport, and increasing the calorific value [32]. The pyrolysis of wet digestate generates steam in the reactor, leading to either a higher moisture content in the product or a high production of non-condensable gases. Furthermore, the combustion of high-moisture digestate would not achieve the temperature levels required by waste combustion legislation. Therefore, sewage sludge undergoes mechanical dewatering and thermal drying to reduce the moisture content to 65% and 5%, respectively [33].

Nutrients, mainly nitrogen, are still available for recovery in the reject water as well as the thermal drying fumes, which are directed to the condenser, with the resulting

condensate containing nitrogen. Mass balance calculations are conducted for both dewatering and thermal drying, whereby nitrogen is the key focus in all phases. The nitrogen share in the reject water is about 16% of the total nitrogen contained in the wastewater, which is within the 10–30% range reported in previous studies [34]. In thermal drying, the target moisture removal is 80% [30]. Saud et al. [31] and Deviatkin et al. [30] investigated nitrogen recovery from thermal drying of sewage sludge using established scrubber technology. The mass balance for the WWTP and the dewatering and thermal drying processes is presented in Table 1; a more detailed mass balance is given in the Table S2 of the Supplementary Materialss.

**Table 1.** Mass balance for the WWTP considered in this study [35].

| WWTP | Value | Unit |
|---|---|---|
| Wastewater | 250,000 | $m^3/d$ |
| Nitrogen | 13 | t/d |
| Phosphorus | 1.6 | t/d |
| TS | 79 | t/d |
| **Clean water** | | |
| Mass | 250,000 | t/d |
| Nitrogen | 1.2 | t/d |
| Phosphorus | 0.041 | t/d |
| **Mass to digestion** | | |
| Mass | 2600 | t/d |
| TS | 88 | t/d |
| Nitrogen | 3.6 | t/d |
| Phosphorus | 1.4 | t/d |
| **Dewatered digestate** | | |
| Mass | 180 | t/d |
| TS | 52 | t/d |
| Nitrogen | 1.5 | t/d |
| Phosphorus | 1.4 | t/d |
| **Reject water** | | |
| Mass | 2400 | t/d |
| TS | 1.1 | t/d |
| Nitrogen | 2.1 | t/d |
| Phosphorus | 0.033 | t/d |
| **Thermally dried digestate** | | |
| Mass | 58 | t/d |
| TS | 52 | t/d |
| Moisture content | 20 | % |
| Nitrogen in drying fumes | 0.74 | t/d |
| **Condensate** | | |
| Mass | 120 | t/d |
| Nitrogen | 0.47 | t/d |

*2.2. Pyrolysis Scenario*

In the pyrolysis scenario, presented in Figure 2, wood waste is used together with digestate to produce biochar and pyrolysis gas. The digestate constitutes 70% of the pyrolyzed mass, and the remaining 30% is wood waste. The reason for adding wood waste is to compensate for the lower heating value of the digestate and to introduce more carbon. The pyrolysis process is assumed to be self-sufficient in terms of heat energy through the combustion of the pyrolysis gas and recovering the heat to be used for both thermal drying and pyrolysis. The electricity consumption of the thermal drying process is 0.45 kWh/kg [36], whereas that of the pyrolysis process is another 0.01 kWh/kg [35]. The mass balance of the pyrolysis process is summarized in Table 2.
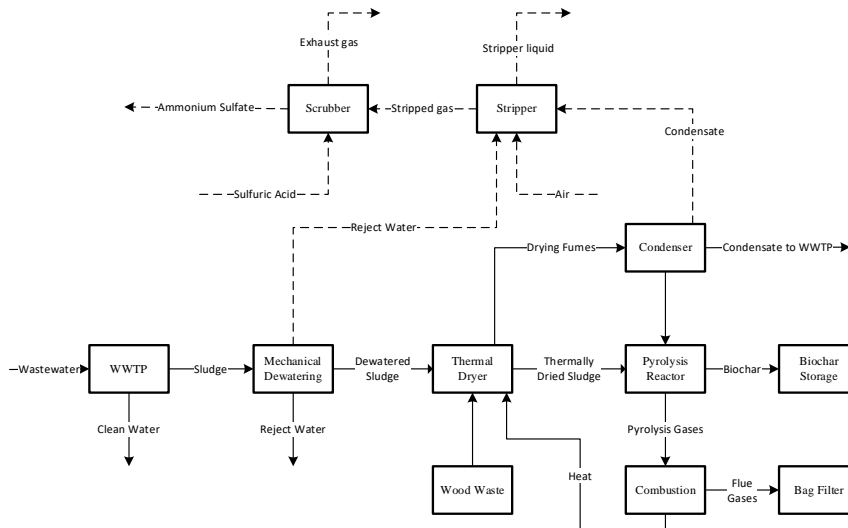
**Figure 2.** Block diagram of the pyrolysis process. Solid lines denote the current process and dotted lines show the possibilities to recover nitrogen from the reject water and thermal drying fumes.

### 2.3. Combustion Scenario

The sludge combustion data are taken from a novel sludge combustion plant, sited in Rovaniemi (Finland) and built by Endev Ltd., which has a throughput of 10,000 t/a. After mechanical dewatering in the WWTP, the sludge enters an integrated fluidized bed drying and combustion process (Figure 3). First, hot sand from a combustion reactor is mixed with the sludge in a dryer to remove moisture, thereby increasing the dry solids content of the sludge to 95–98 wt %. Then, the dried sludge is transferred to a fluidized bed reactor for combustion. The temperature of the reactor is maintained at 850 °C to ensure the destruction of organic pollutants (pathogens, drug residuals, microplastics, etc.). The ash formed in the combustion is carried with flue gases and extracted at two points: the majority of the ash (>95 wt %), the so-called product ash, is collected through a high-temperature cyclone after the air preheater, while the finer ash is removed by a bag filter as byproduct ash. The heat produced in the combustion is used in thermal drying and district heating, whereas the hygienic and nutrient-rich product ash can be used as fertilizer. The water vapor from the fluidized bed dryer is sent to a condenser, and the condensate is directed to the WWTP for wastewater treatment. This condensate contains recoverable nitrogen with concentrations up to 3000 mg/L. The electricity consumption of the combustion plant is 0.147 kWh/t of dewatered sludge, and the thermal efficiency of the plant is 82%.

A wood boiler is also added to the process to make the mass and energy balance comparable with the pyrolysis scenario. The lower heating value (as-received basis) of thermally dried digestate and wood waste is calculated using Equation (1).

$$\mathrm{LHV_{ar}} = \mathrm{LHV_{dry}} \times (1 - w) - l_{25}w \tag{1}$$

where the lower heating value for total solids ($\mathrm{LHV_{dry,D}}$) is 13 MJ/kg for thermally dried digestate and 18 MJ/kg for wood ($\mathrm{LHV_{dry,W}}$). Furthermore, the moisture content, $w$, is 5% for thermally dried digestate and 41% for wood waste, and the heat of vaporization of water, $l_{25}$, is 2.443 MJ/kg. As a result, the lower heating value as received, $\mathrm{LHV_{ar,D}}$ of thermally dried digestate is 12 MJ/kg, and the $\mathrm{LHV_{ar,W}}$ of wood is 9.6 MJ/kg.
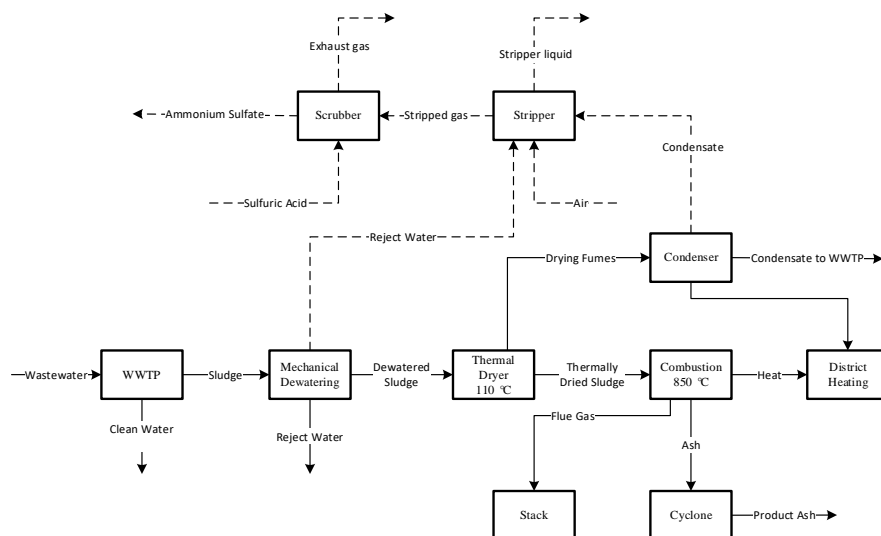
**Figure 3.** Block diagram of the combustion process. Solid lines denote the current process and dotted lines show the possibilities to recover nitrogen from the reject water and thermal drying fumes.

### 2.4. Recovery of Nutrients

In both the pyrolysis and combustion scenarios, 780 t/a of nitrogen ends up in the reject water and can be recovered by adding a stripper and a gas scrubber to the systems. Furthermore, it is assumed, based on Horttanainen et al. [37], that 12% of the total nitrogen in the dewatered sludge is released in the thermal drying fumes in the form of $NH_3$. The high moisture content of the drying fumes would result in a diluted fertilizer. To avoid this, a condenser is added to the process before the stripper–scrubber combination, and thus the drying fumes first pass through this condenser; the condensate is directed to the stripper–scrubber combination together with the reject water.

Air is used to separate gaseous ammonia ($NH_3$) from the liquid in the column in the stripping process. This study assumes a stripping column efficiency of 95% based on the previous literature [38,39]. Subsequently, the stripped gas is directed to an acid scrubber to capture ammonia using sulfuric acid to produce ammonium sulfate. In the scrubber, the liquid to gas (L/G) ratio, acid pH, and mass transfer area play significant roles [31,40]. The scrubber efficiency is also assumed to be 95% based on previous studies [41,42]. The exhaust gas from the scrubber is directed to combustion to destroy possible odorous compounds.

### 2.5. Cost Estimation

The general cost estimation is conducted based on sewage sludge-derived biochar and district heat prices in Finland. Table 2 takes only the sewage sludge-derived biochar values into account in the calculation of the total investment cost. However, other wood-derived biochar values are presented to enable a comparison of the prices with sewage sludge-derived biochar. There is a high variation in the district heat prices due to differences in regional transmission and supply and demand. Therefore, an average of all values is considered to calculate the revenue from district heating. All district heating prices are exclusive of value-added taxes (VAT).

**Table 2.** Biochar and district heat revenue estimation in the pyrolysis and combustion scenarios.

| Biochar (S1.1 and S1.2) | | |
|---|---|---|
| Price (EUR/t) | Country | Reference |
| Sewage sludge derived | | |
| 0 | Finland | [43] |
| 400 | Finland | [43] |
| Wood derived | | |
| 700 | Finland | [44] |
| 800 | USA | [45] |
| **District heat (S2.1 and S2.2)** | | |
| Price (EUR/MWh) | Country | Reference |
| 40 | Finland | [46] |
| 50 | Finland | [47] |
| 60 | Finland | [48] |
| 62 | Finland | [49] |
| 118 | Finland | [46] |

## 3. Results and Discussion

### 3.1. Recovery of Nitrogen from Drying Fumes

The results of the pyrolysis and combustion scenarios with and without the recovery of nitrogen from the reject water are shown in Figures 4–9. Figures 4 and 5 present the base scenarios S1.0 (pyrolysis without N recovery) and S2.0 (combustion without N recovery), respectively. In both scenarios, it is evident that nitrogen could be recovered from the drying fumes and reject water.
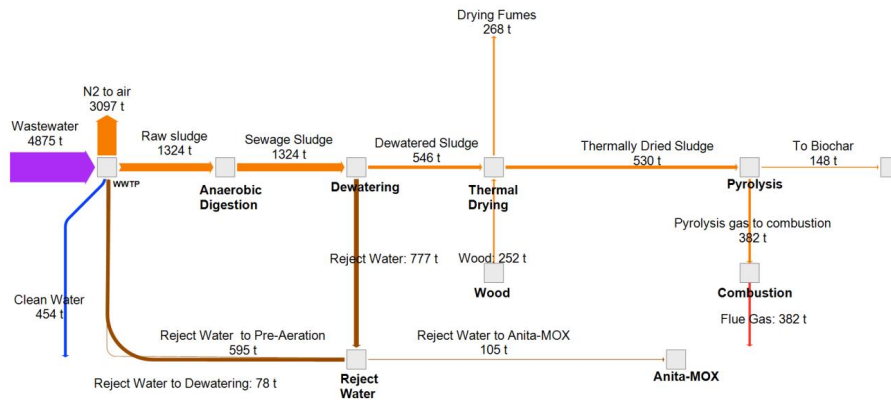


**Figure 4.** Nitrogen balance for S1.0 (pyrolysis base scenario, no N recovery).

The pyrolysis scenario S1.1 and combustion scenario S2.1, where nitrogen is recovered from the drying fumes but not from the reject water, are presented in Figures 6 and 7, respectively. The mass balance calculations reveal that the nitrogen loss in the WWTP process is 3100 t/a in the form of $N_2$ to the atmosphere and 450 t/a in the form of nitrate ($-NO_3$) and ammonium ions ($NH^{4+}$) to clean water, estimated as a 73% loss of total nitrogen. The remaining nitrogen ends up in dewatered digestate, calculated at 550 t/a, or 11%. The remaining 16%, or 780 t/a, is present mainly in the reject water from the mechanical dewatering and can be recovered. Similarly, it is calculated that 3% of nitrogen can be recovered from the drying fumes. The drying fumes from the thermal dryer contain 160 t/a of nitrogen, capable of producing ammonium sulfate fertilizer. The condenser offers two benefits: firstly, it removes moisture from the non-condensable odorous gases that enter

the combustion process. Secondly, it recovers nitrogen from the condensed water. In this case, assuming a scrubber efficiency of 95%, 160 t/a of nitrogen can be captured for use as fertilizer. Odorous gases are directed to combustion. In Figure 5, the combustion of sewage sludge produces 120,000 MWh/a of heat, which can be used for district heating, as explained in a later section. Thermally dried sludge still contains 400 t/a of nitrogen, which is eventually combusted and cannot be utilized. Phosphorus ends up in the ash of sewage sludge combustion. A more detailed nutrient balance is presented in the Tables S4–S6 in the Supplementary Materials. Almost 95% of the phosphorus in thermally dried sewage sludge ends up in the product ash, with the remaining 5% contained in the non-utilizable byproduct ash [50].
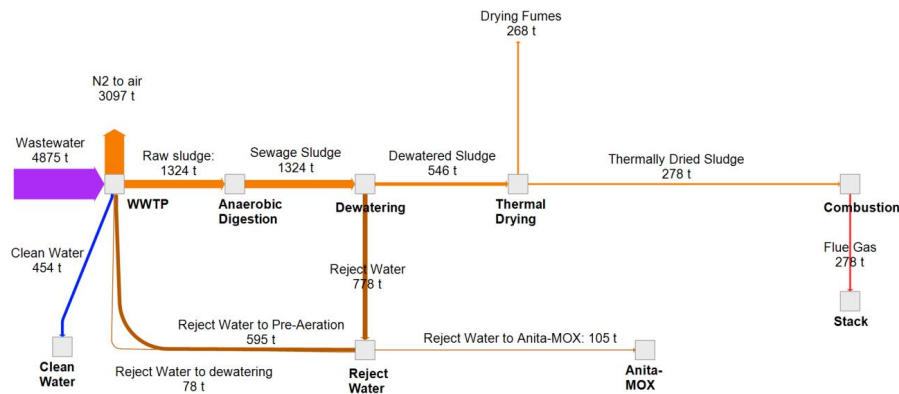


**Figure 5.** Nitrogen balance for S2.0 (combustion base scenario, no N recovery).
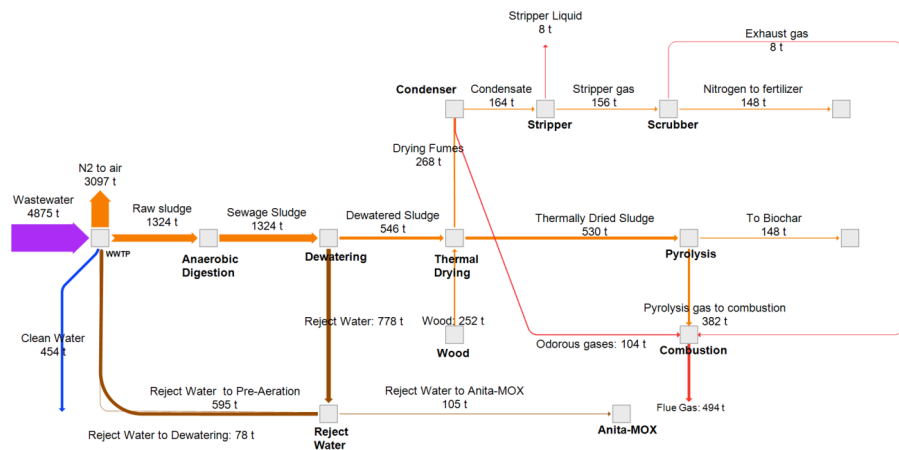


**Figure 6.** Nitrogen balance for S1.1 (pyrolysis with N recovery from drying fumes).
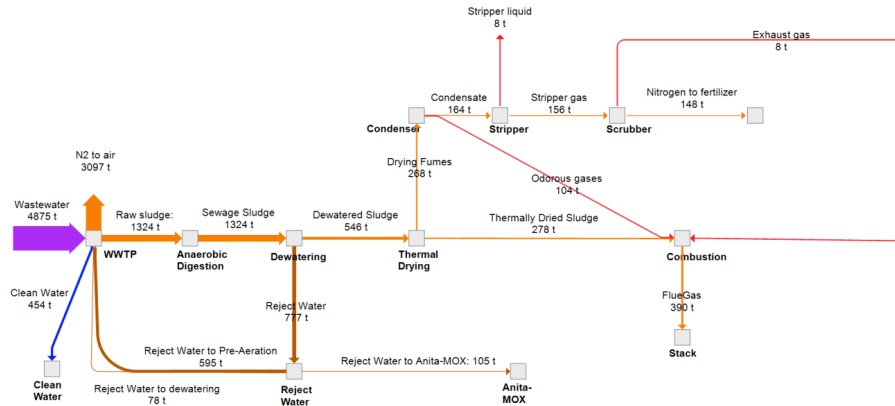
**Figure 7.** Nitrogen balance for S2.1 (combustion with N recovery from drying fumes).
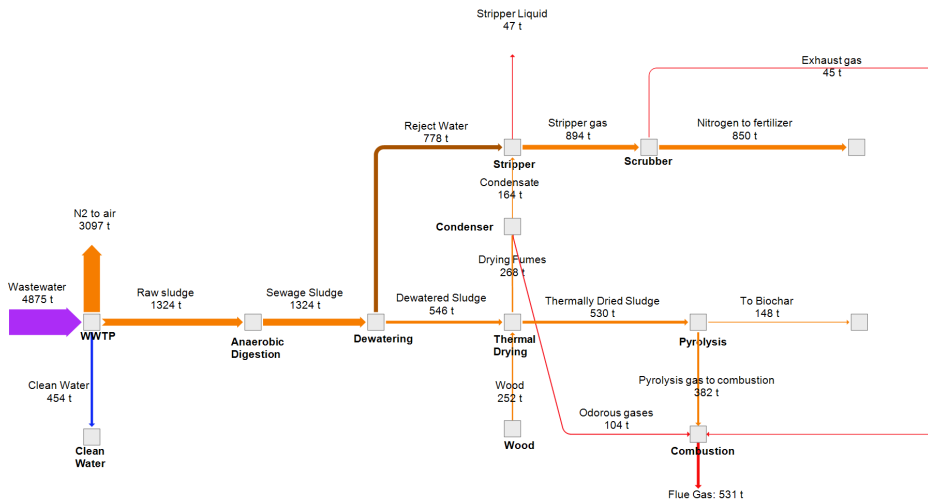


**Figure 8.** Nitrogen balance for S1.2 (pyrolysis with N recovery from drying fumes and reject water).

As shown in Figure 4, 12,000 t/a of biochar, as the main product, contains about 150 t/a nitrogen, and 150 t/a of ammonium fertilizer can be produced in the scrubber as part of the pyrolysis process. The biochar also includes 500 t/a of phosphorus. This biochar has commercial value as a soil amendment and fertilizer due to the presence of nutrients such as phosphorus and nitrogen, although it may be in a less available form for plants [51]. The gases produced during pyrolysis are used for heating in the main plant equipment, such as the pyrolysis reactor and dryer.

*3.2. Recovery of Nitrogen from Reject Water*

Only a small amount of nitrogen (268 t/a) is available for recovery in the thermal drying fumes. It is difficult to justify the investment needed to conduct this recovery alone as the off-gases are irrespectively treated during combustion, whereby the ammonia can

be converted to N$_2$. To increase the overall feasibility of nitrogen recovery, it is possible to combine the recovery from thermal drying fumes with recovery from WWTP reject water, which contains the majority of recoverable nitrogen. In the WWTP reject water, 780 t/a of nitrogen is available for recovery and further utilization as ammonium fertilizer as shown in Figures 8 and 9.
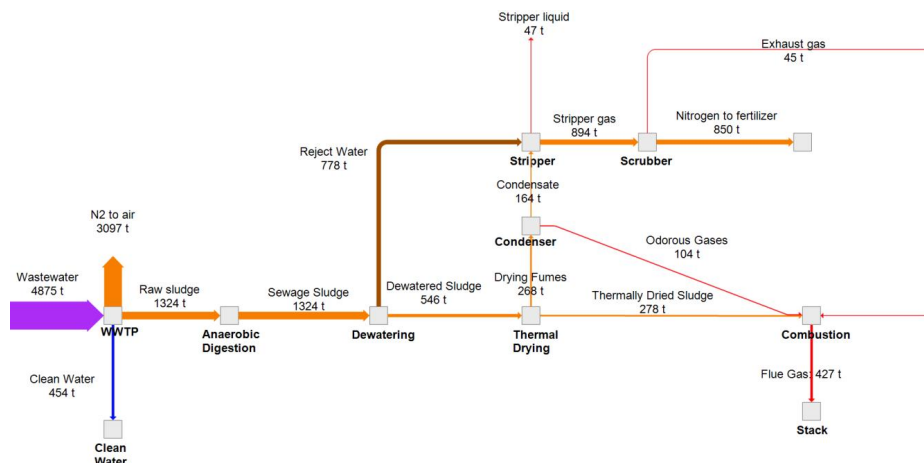


**Figure 9.** Nitrogen balance for S2.2 (combustion with N recovery from drying fumes and reject water).

### 3.3. Comparison of Mass and Energy Balances

The mass balances of the pyrolysis and combustion scenarios (Table 3) use the same amount of wastewater input, and the difference is in the end-products' masses. As shown in Table 3, biochar is produced in pyrolysis at a mass of 12,000 t without ash production, while the combustion process produces 9700 t of ash as a byproduct along with district heat, including 1600 t of wood ash. More than 95% of the ash is separated by a cyclone at a high temperature to reduce the heavy metal concentration. The resulting ash contains phosphorus and can be used in forestry or agriculture, although only 10–25 wt % is available in the form of P$_2$O$_5$ compared to the 5–40 wt % of P$_2$O$_5$ in phosphate ores. The ash can also be used to manufacture lightweight bricks and tiles, as an additive in the manufacture of cement, and as a raw material for Portland cement [52]. Meanwhile, biochar can be used as an adsorbent to remove heavy metals and pollutants from aqueous solutions [53] and as a soil amendment [54]. The heavy metals in biochar and other hazardous compounds, such as dissolved organic carbon which might end up in soil and drinking water after leaching or rainfalls, are still a concern in applicability of biochar in soil. The potential risk of heavy metals in sewage sludge can be reduced by pyrolysis with variation of temperature [55]. The increase in temperature from 300 to 700 °C, increase the concentration of zinc (Zn), chromium (Cr), nickel (Ni), and copper (Cu), whereas the concentration of lead (Pb) and cadmium (Cd) is decreased when temperature increases from 600 to 700 °C. More research and experimental work is needed to completely analyze the heavy metal impact in biochar [56]. The microplastics are also removed at a temperature of 500 °C, which decreases the potential risk to the environment [57]. The detailed mass balance is presented in the Table S2 in the Supplementary Materials.

The pyrolysis and combustion processes differ in their energy balance (Table 4). The production of district heat is the main product in combustion, whereas pyrolysis produces biochar, and the energy of the pyrolysis gas is used to heat the pyrolysis process and dry the sludge. The additional energy consumption in the reject water recovery scenario is

due to the increased mass flow rate of the water, which needs to be heated up to 60 °C for stripping. Consequently, the stripper–scrubber electricity consumption is tripled as compared to the energy balance without reject water recovery. The condensate entering the stripper without reject water recovery has a temperature of 60 °C [50] and further heating is not required. However, with reject water recovery, the reject water is assumed to be 28 °C before heating because the temperature of the digested mass is usually 28–32 °C after dewatering; hence, the liquid needs to first be heated to 60 °C to achieve the optimum efficiency in the stripper [58].

**Table 3.** Comparison of mass balance for pyrolysis and combustion.

| Mass Balance | Pyrolysis | Combustion | Units |
|---|---|---|---|
| **Inputs** | | | |
| Wastewater | 92,000,000 | 92,000,000 | t/a |
| Wood waste | 27,000 | 27,000 | t/a |
| $H_2SO_4$ | 3700 | 3700 | t/a |
| NaOH | 3100 | 3100 | t/a |
| **Intermediate products** | | | |
| Mass to digestion | 950,000 | 950,000 | t/a |
| Biogas | 13,000 | 13,000 | t/a |
| Reject water from mechanical dewatering | 867,000 | 867,000 | t/a |
| Dewatered sludge | 65,000 | 65,000 | t/a |
| Thermally dried sludge | 27,000 | 27,000 | t/a |
| **Outputs** | | | |
| Clean water | 91,500,000 | 91,500,000 | t/a |
| Ash | 0 | 9700 | t/a |
| Biochar | 12,000 | 0 | t/a |

Comparing the end products, in addition to the respective biochar and district heat, both processes also produce 3500 t of ammonium sulfate from the drying fumes and reject water (Table 5). The chemical consumption is also analyzed for both stripping and scrubbing. NaOH is used in the stripper to raise the pH of the liquid to 8–11, depending on the process conditions, while $H_2SO_4$ is used as a scrubbing agent to absorb ammonium ions.

The concentration of ammonium sulfate affects how it can be used in agriculture in combination with other fertilizers. In order to achieve commercial-grade fertilizer, further post-treatment such as crystallization or evaporation may be required, which adds cost to the process [31].

### 3.4. Comparison of Revenues and Costs

The cost and revenue calculations were conducted for both process scenarios and with and without reject water recovery. Biochar and heat, having monetary value, are the main products of the two optional sludge treatment processes. Biochar can have commercial value, but it is subject to a wide range of prices depending on many factors, such as the source of the feedstock, process parameters, amount of equipment used, and operating costs [59]. As there is no difference in biochar production with or without reject water recovery, Table 6 presents different values from the literature to analyze the revenue generation in the pyrolysis scenarios S1.1 and S1.2. The other reasons for biochar's uncertain market value are the unavailability of commercial-scale production methods and the unestablished demand for different quality biochars. The high cost of biochar prevents consumers from using it in large quantities, but this cost will reduce following the commercial production of biochar. Furthermore, biochar made from biomass can be utilized in pyrogenic carbon capture and storage (PyCCS), whereby sequestered carbon dioxide can be sold on the market (Puro Earth 2021), which could offer added value for biochar made of sewage sludge digestate.

**Table 4.** Comparison of energy balance for both pyrolysis and combustion with and without reject water recovery.

| Energy Balance | Pyrolysis (MWh/a) | Combustion (MWh/a) |
|---|---|---|
| **Without reject water** | | |
| **Consumption** | | |
| Reactor electricity consumption | −6500 | −9600 |
| Stripper and scrubber electricity consumption | −680 | −680 |
| **Production** | | |
| District heat sewage sludge | 0 | 56,000 |
| District heat wood | 0 | 62,000 |
| Net balance of heat | 0 | 120,000 |
| Net balance of electricity | −7200 | −10,000 |
| **With reject water** | | |
| **Consumption** | | |
| Reactor electricity consumption | −6500 | −9600 |
| Stripper and scrubber electricity consumption | −3900 | −3900 |
| Stripper heat consumption | −34,000 | −34,000 |
| **Production** | | |
| District heat sewage sludge | 0 | 56,000 |
| District heat wood | 0 | 62,000 |
| Net balance of heat | −34,000 | 84,000 |
| Net balance of electricity | −10,000 | −13,000 |

**Table 5.** Production of ammonium sulfate.

| Ammonium Sulfate Production | Pyrolysis | Combustion | Units |
|---|---|---|---|
| Ammonium sulfate/kg $NH_3$ | 4 | 4 | $kg_{ammsulfate}$/kg $NH_3$ |
| Ammonium sulfate from total off-gases | 600 | 600 | $t_{ammsulfate}$/a |
| Ammonium sulfate (from reject water) | 2900 | 2900 | $t_{ammsulfate}$/a |
| Total ammonium sulfate | 3500 | 3500 | $t_{ammsulfate}$/a |

As shown in Table 7, the commercial value of district heating produced in combustion is also compared with existing district heating prices, excluding VAT, for scenarios S2.1 and S2.2. The main difference between the two scenarios is the larger consumption of heat during reject water recovery, which is necessarily subtracted from the total production of district heat. District heating clearly has the potential to generate revenue. Moreover, the disposal cost of sewage sludge in landfill ranges between 60 and 200 EUR/t of TS in Europe. The treatment cost using pyrolysis and combustion may be higher, but these represent an environmentally safe way to utilize sewage sludge [60].

**Table 6.** Revenue and cost estimation of fertilizer and used chemicals for S1.1 and S1.2 (pyrolysis with nitrogen recovery).

| Pyrolysis | | | | Reference |
|---|---|---|---|---|
| **Costs** | **Mass, t/a** | **Cost, EUR/t** | **Total, M EUR/a** | |
| Chemical used | | | | |
| $H_2SO_4$ | 3700 | 160 | −0.59 | [61] |
| NaOH | 3100 | 380 | −1.2 | [62] |
| Annual investment | | | | |
| Sewage sludge | 65,000 | 35–45 | −2.3–(−2.9) | [63] |
| **Revenues** | **Mass, t/a** | **Price (EUR/t)** | **Total, (M EUR/a)** | |
| Ammonium sulfate-N | 4000 | 660 | 2.6 | [51] |
| Biochar | 12,000 | 400 | 4.8 | [63] |
| **Total** | | | **2.8–3.4** | |

**Table 7.** Revenue and cost estimation of fertilizer and used chemicals for S2.1 and S2.2 (combustion with nitrogen recovery).

| Combustion | | | | Reference |
|---|---|---|---|---|
| **Costs** | **Mass, t/a** | **Cost, EUR/t** | **Total, M EUR/a** | |
| Chemical used | | | | |
| $H_2SO_4$ | 3700 | 160 | −0.59 | [61] |
| NaOH | 3100 | 380 | −1.2 | [62] |
| Annual investment | | | | |
| Sewage sludge | 65,000 | 30–60 | −2−(−3.9) | [63] |
| **Revenues** | **Mass, t/a** | **Nitrogen/Phosphorus (EUR/t)** | **Total, (M EUR/a)** | |
| Ammonium sulfate | 4000 | 660 | 2.6 | [51] |
| Phosphorus | 480 | 1600 | 0.76 | [51] |
| | **MWh/a** | **Price (EUR/MWh)** | **Total, (M EUR/a)** | |
| District heat | 120,000 | 66 | 7.9 | [46–49] |
| **Total** | | | 3.3–5.2 | |

The recovery of nitrogen from reject water increases the amount of ammonium sulfate produced. For both pyrolysis and combustion, Tables 6 and 7 show the revenue generated in terms of nitrogen (in the form of ammonium sulfate) and phosphorus. Ammonium sulfate can generate 3 M EUR/a in revenue from both the pyrolysis and combustion processes, whereas phosphorus can generate 1 M EUR/a in the combustion process. The reason for the high revenue from combustion is the ash produced in addition to district heating.

On the other hand, the recovery of reject water also increases the chemical consumption requirement in stripping and scrubbing. $H_2SO_4$ and NaOH each add 1 M EUR/a to the operational cost.

The economic estimations of the scenarios can only be approximated since both technologies are in the pilot phase and there is no exact information on the investment and operation costs at the full scale. The total investment cost of the Rovaniemi plant is about 4.3 M EUR, and taking the interest rate of 5% and a depreciation period of 20 years, the calculated investment cost is about 30 EUR/t of sludge treated. The fluidized bed combustion plants in Geneva, with a capacity of 55,000 t/a and 28% TS, and the 90,000 t/a plant in Zurich, Switzerland, with 33% TS, have total investment costs of 30–50 M EUR. In Finland, a large fluidized bed plant with a capacity of 70,000 t/a and 25% TS costs about 25 M EUR. Similarly, a pyrolysis plant with the capacity of 30,000 t/a, assuming a 5% interest rate and a 20-year depreciation period, will have an investment cost of 13–17 M EUR, or 35–45 EUR/t of sludge treated [63].

In this study, 65,000 t/a of sludge with 29% TS also falls within the range of the investment costs provided above. In Tables 6 and 7, the estimated investment costs are presented for both the combustion and pyrolysis processes in the different pilot plants. These costs are comparable to the revenues and major chemical costs. The annual investment costs for combustion are 2–4 M EUR, whereas for pyrolysis they range between 2 and 3 M EUR. Adding up the revenues for all the byproducts and subtracting the chemical costs for the reject water recovery scenario, the total revenue would be 3–5 M EUR/a and 3–3.5 M EUR/a for combustion and pyrolysis, respectively. In comparison with the annual investment cost, these revenue values clearly dominate. The two-edged benefits of sewage sludge include the recovery of nutrients from waste and the production of revenue from the resultant products, thereby emphasizing the need to consider these alternatives in sewage sludge management.

## 4. Conclusions

The integration of pyrolysis and combustion processes with a gas scrubbing technology at a large-scale WWTP was investigated through mass and energy balance calculations, in order to maximize the recovery of nutrients and energy from municipal sewage sludge.

It is shown that approximately 73% (3600 t/a) of the total nitrogen within the wastewater entering the WWTP (4900 t/a) is lost to the air and clean water; the rest is divided between different stages of sludge treatment. Nitrogen can be recovered from two stages: (i) mechanical dewatering (16%, 780 t/a) and (ii) thermal drying (3%, 270 t/a). Furthermore, 120 GWh/a of district heat and 9700 t/a of ash with 500 t/a phosphorus are obtained in the combustion scenario and 12,000 t/a of biochar with 500 t/a phosphorus is obtained in the pyrolysis scenario.

It is not possible to recover a large amount of nitrogen from thermal drying alone, and investment for this purpose only would not be cost-effective. However, there is a likelihood that the ammonia in the off-gases of thermal drying is oxidized, producing NOx emissions, which would make the investment for nitrogen removal/recovery more desirable. The high nitrogen content in reject water can represent a high wastewater treatment cost for sludge treatment plants and a nitrogen load problem for WWTPs. In this regard, it would be beneficial to recover nitrogen from two sources instead of one, i.e., reject water and thermal drying fumes, as it would not substantially change the dimensions of the equipment required for recovery. The recovery from thermal drying fumes could be performed with almost the same effort and expense as the recovery from reject water alone, but the recovery rate would increase by more than 20%.

The addition of a stripper and a scrubber for nitrogen recovery increases the total electricity consumption in both scenarios. Combustion and pyrolysis require annual investment costs of 2–4 M EUR/a and 2–3 M EUR/a, respectively, while 3–5 M EUR/a and 3–3.5 M EUR/a would be generated as product revenues.

Both technologies are viable options for nutrient and energy recovery during sewage sludge disposal, having the capability to overtake conventional sewage sludge disposal methods. However, further research is required for high product yields and decreased investment costs.

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# Publication III

Havukainen J., Saud, A., Peltola P., and Horttanainen M.
**Environmental performance of dewatered sewage sludge digestate
utilization based on life cycle assessment**

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*Waste Management*
Vol. 137, pp. 210-221, 2022
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# Environmental performance of dewatered sewage sludge digestate utilization based on life cycle assessment

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ABSTRACT

Due to the global trend of urbanization, the amount of sewage water is increasing in cities. This calls for efficient treatment of the resulting sewage sludge. To date, in the 27 European Union member countries (EU-27), the prevailing treatment method is application on arable land. Anaerobic digestion is one of the treatment methods being increasingly used nowadays. However, the resulting digestate requires further utilization. Therefore, in this study, the environmental performance of composting, combustion, and pyrolysis options for dewatered sewage sludge digestate is evaluated based on a life cycle assessment. The results show that digestate combustion and composting performed better than pyrolysis for most of the selected impact categories. However, pyrolysis of sewage sludge is still under development, and there are, to some degree, uncertainties in the data related to this technology; thus, more information for the performance assessment of pyrolysis is still required.

## 1. Introduction

The world population is still increasing, and together with the widespread trend of urbanization, this means that more and more people are concentrated in large cities. These cities consume vast amounts of resources and produce billions of tons of waste (Kaza et al., 2018) and wastewater. Efficient wastewater treatment is paramount to reduce the impact on the environment. The ever-increasing amount of wastewater and the emergence of more efficient wastewater treatment processes lead to increasing amounts of sewage sludge, which requires further treatment. In the 27 European Union member countries (EU-27), the situation has improved over the last few decades: in 2017, 70% of the population was linked to tertiary-level treatment and 13% to secondary-level treatment (European Environment Agency, 2020). As a result of the Urban Wastewater Treatment (UWWT) Directive 91/271/EC, there was nearly a 50% increase in sewage sludge mass between 1992 and 2005 in EU-15. Generation of sewage sludge varies significantly between European countries, ranging from 0.1 kg per population equivalent and year to 30.8 kg per population equivalent and year (Kelessidis and Stasinakis, 2012). According to data from 2003 to 2006, the dry mass of sewage sludge generated annually in the EU-27 was approximately 10 million tons (Bianchini et al., 2016); it was 11.5 million tons in 2010 (Wiechmann et al., 2013) and was expected to reach 13.5 million tons by 2020 (Durdević et al., 2020).

According to EU statistics, the main treatment method for sewage sludge in the EU-27 is agricultural use, followed by composting and other applications, while disposal into landfills is the least-used method (Eurostat, 2020). Most EU countries have banned disposal into landfills, according to the EU landfill directive 99/31/EC. The regulation on the application of sewage sludge on land is presented in the sewage sludge directive (SSD) (Bianchini et al., 2016). Among the stabilization methods, composting and anaerobic digestion (AD) are the most-used methods, with the former taking place in 20 countries and the latter in 24 countries in the EU, according to Kelessidis and Stasinakis (2012).

AD can be conducted at a wastewater treatment plant (WWTP) to produce biogas for supplying energy to the WWTP (Jenicek et al., 2012). The produced biogas can also be upgraded for use in vehicles (Osorio and Torres, 2009). The remaining digestate can then be dewatered, and the resulting reject water can be directed to the WWTP. The dewatered digestate goes for further treatment. Zhao and Viraraghavan (2004) examined the Regina WWTP in Greece, where dewatered digestate was

| Nomenclature | |
| --- | --- |
| **Subscripts** | |
| ar | as received |
| dry | dry basis |
| | |
| **Acronyms** | |
| AD | anaerobic digestion |
| EASETECH | Environmental Assessment System for Environmental Technologies |
| EU | European Union |
| HSY | Helsinki Region Environmental Services |
| LCA | life cycle assessment |
| LCI | life cycle inventory |
| LCIA | life cycle impact assessment |
| L/G | liquid-to-gas |
| LFO | light fuel oil |
| LHV | lower heating value |
| LPG | liquified petroleum gas |
| SR | sensitivity ratio |
| SSD | sewage sludge directive |
| TS | total solid |
| VS | volatile solid |
| WASSTRIP | Waste Activated Sludge Stripping to Remove Internal Phosphorus |
| WWTP | wastewater treatment plant |

directed to a landfill. Cukjati et al. (2012) studied sewage sludge digestate composting with wood chips, wood bark, and saw dust and found it to be the most economical for use as a landfill cover material. In Prague, dewatered digestate is composted and applied to land (Jenicek et al., 2012).

While application on land is still the main treatment method for sewage sludge and dewatered sewage sludge digestate can also be applied on arable land, there have been concerns related to the safety of utilizing sewage sludge in this manner. Sewage sludge contains heavy metals (Fytili and Zabaniotou, 2008), pathogens (Bibby and Peccia, 2013), and pharmaceutical residues (Malmborg and Magnér, 2015), and can work as a route for microplastics entering into soil (Van der Heyden et al., 2015). Therefore, there has been interest in thermal treatment of sewage sludge (Fytili and Zabaniotou, 2008), which can tackle these issues. While it does not remove heavy metals, it can potentially reduce their mobility.

Thermal treatment options for sewage sludge include direct incineration, gasification, and pyrolysis. In addition, sewage sludge could be utilized in cement kilns to replace conventional fuels, when it is kept in a low share of not more than 5% of clinker capacity (Fytili and Zabaniotou, 2008). Incineration is an effective way to reduce the volume of sewage sludge while producing heat and/or electricity, and the remaining residue is ash. Gasification aims to produce energy-rich syngas that can be utilized in energy, fuel or chemical production, with char and tar as residues (Syed-Hassan et al., 2017). Pyrolysis can produce (i) syngas, which is used mainly as an energy source for the pyrolysis process; (ii) tar, which can be used for energy production; and (iii) biochar, which can be used in energy production or as a raw material (Hospido et al., 2005). Biochar could be utilized as a phosphorus-rich soil amendment (Frišták et al., 2018) or in carbon sequestration, as pyrolysis retains part of the carbon in stabile form (Cao and Pawłowski, 2013). With the help of a solvent, liquefaction can produce oil, gas, charred solids, and reaction water (Leng et al., 2015), of which oil and gas can be utilized in energy production.

Similar thermal treatment technologies can also be utilized for dewatered sewage sludge digestate. Fluidized bed drying, combustion,

and pyrolysis conducted with the aim of producing only biochar are emerging options to tackle the issue of drug residues, bacterial resistance, and spreading of microplastics; application of compost made from dewatered sewage digestate on arable land loses popularity due to these concerns. Besides technological feasibility and economic assessments, life cycle assessment (LCA) has a role in selecting a suitable technology. LCA can be used to quantify the environmental performance of a process or a system by taking into consideration the emissions associated with material and energy flows throughout the entire life cycle.

LCA has been conducted for several treatment technologies, for example, application on agricultural land, AD, incineration of digestate (Yoshida et al., 2018), AD versus thermal processes (Hospido et al., 2005), and AD with or without fast pyrolysis (Cao and Pawłowski, 2013). Furthermore, Teoh and Li (2020) investigated the environmental feasibility of a wide range of treatment technologies, ranging from biological and chemical to thermo-chemical methods. Yoshida et al. (2018) found that human toxicity, non-carcinogenicity, and ecotoxicity are the main impact concerns for the studied technologies, for which the incineration technology showed the least potential impacts. Hospido et al. (2005) concluded that AD followed by application on land is a suitable option considering the low amount of heavy metals in the sludge. Cao and Pawlowski (2013) state that AD with fast pyrolysis performed better than fast pyrolysis only. Findings by Teoh and Li (2020) suggest that the best-performing technologies were AD, pyrolysis, and super-critical water oxidation.

The selection of technology for utilizing dewatered sludge digestate can be, to some extent, based on the perceived risk that bacterial resistance, drug residues, and spreading of microplastics present to nature as well as on the acceptance of products produced on-field where dewatered-digestate-based recycled fertilizers are used. While these impacts are not yet measured by LCA, they can provide valuable information on the other impacts of the emerging treatment options, such as fluidized bed combustion and pyrolysis of dewatered sewage. These impacts may be climate change, eutrophication and acidification potential, mineral scarcity, and toxicity. This information can prove valuable when choosing treatment options.

The purpose of this study is to investigate the environmental performance of three different utilization methods for dewatered sewage sludge digestate generated in municipal WWTPs implementing AD for the treatment of raw sludge. The studied methods are (i) windrow composting and subsequent utilization of compost on arable land and in compost soil production, and two thermal treatment methods, namely (ii) fluidized bed drying and combustion, and (iii) low-temperature pyrolysis to produce biochar.

## 2. Materials and methods

The environmental performance is assessed by using LCA methodology, which is a systematic method for assessing the potential environmental performance of products and services. LCA was conducted using a combination of ISO standards 14040 and 14044 (ISO 14040, 2006; ISO 14044, 2006). This study includes the four steps of LCA: goal and scope definition, life cycle inventory, life cycle impact assessment, and interpretation of results.

### 2.1. Goal and scope definition

This study focuses on the environmental performance of the utilization of dewatered sewage sludge digestate from a municipal WWTP. Three different utilization technologies are investigated, including windrow composting, fluidized bed combustion, and pyrolysis (Fig. 1). The functional unit of this study is 1 metric ton (t) of dewatered sewage sludge digestate. Consequential modeling is used, as the focus is on the long-term impacts of a decision on existing sewage sludge digestate treatment. Marginal electricity and heat are used in the processes, and the production mix calculation is presented in Supplementary
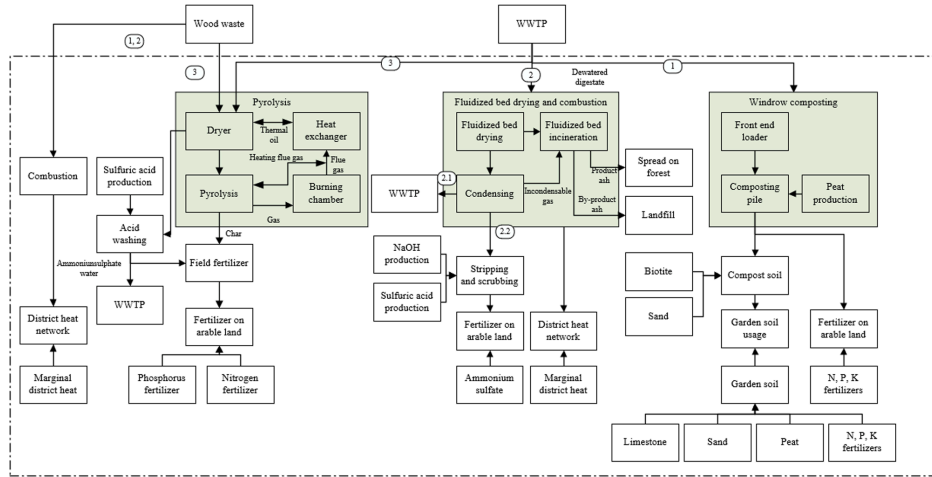
**Fig. 1.** System boundary of sewage sludge digestate treatment.

Information SI-2, Table SI-1, and Table SI-2. Details regarding the background processes are presented in Table SI-5.

The EASETECH (Environmental Assessment System for Environmental Technologies) modeling tool was used for LCA, considering heterogeneous material flows (Clavreul et al., 2014). ReCiPe 2016 with long-term impact was used as the selected life cycle impact assessment (LCIA) method, and the selected mid-point impact categories include climate change due to global importance; particulate emissions and acidification, which are important when energy production is concerned; eutrophication (as nitrogen and phosphorus flows are important in biological treatment); mineral scarcity (to consider the depletion of these reserves); and terrestrial and human toxicity (to estimate the potential impact of heavy metals). The impact categories are described in Supplementary Information SI-6 (Table SI-9).

Three different scenarios for the treatment of dewatered sewage sludge are considered:

1. Windrow composting followed by compost utilization as compost on arable land and compost soil production (S1).
2. Combustion followed by utilization of heat as district heat and ash as a forest fertilizer (S2). Condensate from the thermal drying fumes is directed to:
   a. the WWTP (2.1),
   b. nitrogen recovery (2.2).
3. Pyrolysis with char utilization on arable land (S3).

Fig. 1 shows the system boundary, for which dewatered sewage sludge is considered as an input. Before entering the system boundary, sludge is first digested and dewatered at a WWTP and then transported for further treatment using windrow composting, combustion, or pyrolysis. The reference year is 2020, for which most of the life cycle inventory (LCI) data were collected, and the geographical scope is Finland.

### 2.2. Life cycle inventory

#### 2.2.1. Dewatered sewage sludge digestate and wood

The composition of dewatered sewage sludge is obtained from the Viikinmäki WTTP (HSY, 2019a), located in the Helsinki Region in Finland and operated by the Helsinki Regional Environmental Services

authority (HSY). The properties of the dewatered sewage sludge digestate and wood waste used in pyrolysis are summarized in Table 1.

#### 2.2.2. Sewage sludge digestate composting

The business as usual method (S1) is sewage sludge digestate composting in a windrow (Figure SI-1). The data for S1 are based on the windrow composting of dewatered sewage sludge digestate utilized in the Metsäpirtti composting area owned by Helsinki Region Environmental Services, HSY (Mölsä, 2019). The composting process utilizes peat as the support material (LCI data in Table SI-4), and mechanical

**Table 1**
Properties of sewage sludge and wood (Kainulainen, 2020).

| Parameter | Dewatered sewage sludge digestate | Wood waste | Unit |
|---|---|---|---|
| Total solid (TS) | 28 | 58 | % of mass |
| Volatile solid (VS) | 57 | 90 | % of TS |
| Ash | 43 | 10 | % of TS |
| $LHV_{dry}$ | 13 | 18 | MJ/kg TS |
| $LHV_{ar}$ | 2 | 10 | MJ/kg |
| C | 29 | 45 | % of TS |
| N | 38 | 16 | g/kg TS |
| P | 31 | 1.7 | g/kg TS |
| K | 1.1 | 11 | g/kg TS |
| S | 15 | 2.0 | g/kg TS |
| Ca | 20 | 7.9 | g/kg TS |
| Fe | 140 | 2.8 | g/kg TS |
| Al | 4.7 | 2.1 | g/kg TS |
| Mg | 2.2 | 1.0 | g/kg TS |
| Na | 0.43 | 6.4 | g/kg TS |
| Cl | 0.39 | 10 | g/kg TS |
| Zn | 694 | 68 | mg/kg TS |
| Mn | 354 | 55 | mg/kg TS |
| As | 3.3 | 1.0 | mg/kg TS |
| B | 15 | 13 | mg/kg TS |
| Cd | 0.40 | 0.20 | mg/kg TS |
| Cr | 21 | 17 | mg/kg TS |
| Cu | 310 | 16 | mg/kg TS |
| Hg | 0.32 | 0.060 | mg/kg TS |
| Mo | 4.1 | 2.0 | mg/kg TS |
| Ni | 26 | 8.0 | mg/kg TS |
| Pb | 11 | 5.0 | mg/kg TS |

aeration of the composting pile is done by turning windrows using a front-end loader, which is run by light fuel oil (LFO) (Gareis, 2020). Peat has been proven to work well in the compost process. It reduces odor emissions and the pH value, which can be high in a sludge compost, and the small particle size ensures that there is no screening waste. Furthermore, peat is a necessary component when making a soil product. This means that when peat is used as a supporting material in a composting process it is readily included in the compost, which is directed to soil product manufacturing (Gareis, 2020).

The produced compost is sold mainly as compost soil to be used as a growth medium in gardening and grass-field purposes for individual customers (80%) and, to a smaller extent, as compost to farmers (20%). For preparing compost soil, sand (Wäänänen, 2020) and biotite (Mölsä, 2019) are added to the mature compost to add the required minerals and potassium. It is assumed that the materials are transported by a Euro 6 class truck and that the compost is spread on the field by an agricultural tractor. The main parameters and their values, transfer coefficients for the compost process, and compost utilization and transport distances are summarized in Table 2.

**Table 2**
LCI data of composting.

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Light fuel oil (LFO) demand | 1.57 | l/t sludge | Mölsä, 2019 |
| Electricity demand | 1.56 | kWh/t | Mölsä, 2019 |
| Peat demand | 238 | kg/t sludge | Mölsä, 2019 |
| Biotite demand | 6.8 | kg / t sludge | Mölsä, 2019 |
| N loss | 2.7 | % of tot N | Amlinger et al., 2008; Pradel and Reverdy, 2013 |
| $N_2O$ emission | 42 | % of N loss | Amlinger et al., 2008; Pradel and Reverdy, 2013 |
| $NH_3$ emission | 58 | % of N loss | Andersen et al., 2010; Maulini-Duran et al., 2013 |
| C loss | 53 | % of C | Pagans et al., 2006 |
| C to $CH_4$ | 2.1 | % of C loss | Pagans et al., 2006 |
| Transfer coefficients to compost | | | Nipuli, 2020; Havukainen et al., 2020 |
| Water | 75 | % | |
| Volatile solid (VS) | 55 | % | |
| Ash | 100 | % | |
| C | 47 | % | |
| Compost use | | | |
| Compost soil sand demand | 775 | kg/t compost | HSY, 2017 |
| Compost soil displacement | 43 | %, volume basis | HSY, 2017; Hartikainen, 2020; Boldrin et al., 2010 |
| Diesel for spreading | 0.14 | l /t compost | Havukainen et al., 2020 |
| P usable | 15 | % | Ylivainio et al., 2020 |
| N soluble | 14 | % | HSY, 2017 |
| $N_2O$ emission | 5.2 | % of tot N | Boldrin et al., 2010 |
| $NH_3$ emission | 7.7 | % of tot N | Boldrin et al., 2010 |
| Compost leaching data | | | Boldrin et al., 2010 |
| As | 3.3 | % | |
| Cd | 0.2 | % | |
| Cr | 0.2 | % | |
| Cu | 0.2 | % | |
| Hg | 0.0004 | % | |
| Ni | 0.9 | % | |
| Pb | 0.3 | % | |
| Zn | 0.6 | % | |
| Biotite production | | | Mölsä, 2019 |
| Electricity demand | 163 | kWh/t | |
| Heat from LFO | 0.22 | MJ/kg | |
| Transport | | | |
| Sludge | 0 | km | Mölsä, 2019 |
| Peat | 0 | km | Statistics Finland, 2017 |
| Sand | 19 | km | Statistics Finland, 2017 |
| Biotite | 408 | km | Mölsä, 2019 |
| Compost | 43 | km | Mölsä, 2019 |

### 2.2.3. Sewage sludge digestate combustion

In S2, dewatered sewage sludge digestate is directed to fluidized bed drying and combustion. The data for this technology are gathered from an existing pilot plant in Rovaniemi, Finland. The plant, built in 2019, is the first industrial sludge combustion facility based on Endev's novel technology. The 1 $MW_{th}$ plant (10 000 t/year), owned by a local water and energy utility company, Napapiirin Energia ja Vesi (NEVE) Ltd., is located next to Rovaniemi city's WWTP, which is also operated by NEVE Ltd.

In the pilot plant's fluidized bed drying and combustion processes, mechanically dewatered sludge [20–25% total solid (TS)] from the nearby WWTP is first dried at 110 °C in a circulating mass dryer and then combusted at 850 °C in a circulating mass reactor. The process has to comply with the EU waste incineration directive, which prescribes a minimum combustion temperature of 850 °C and a minimum gas residence time of 2 s (European Parliament and Council, 2010). The process is self-sufficient in terms of energy, i.e., the sludge is the only fuel required during a steady state operation. Furthermore, the reactor is equipped with a propane-fed start-up burner that will automatically ignite if the combustion temperature in the reactor decreases below 850 °C. Auxiliary fuels are only used during plant start-up and shut-down operations to ensure that the minimum temperature of 850 °C is maintained in the reactor.

A simplified flow diagram of the integrated drying and combustion process is shown in Figure SI-2, and the life cycle inventory data are presented in Table 3. The sludge is stored in a silo, which acts as a buffer between the WWTP and the combustion plant to maintain a steady fuel input. In the dryer, sludge is mixed with hot sand, which evaporates the water from the sludge and forms a dry mix of sludge and sand. Dry solid contents of 95–98 wt% are achieved in the dryer. The dryer is maintained at 110 °C, and the heat needed for moisture evaporation is extracted both from hot sand transferred from the reactor and from flue gases passing through a heat exchanger located in the downcomer of the dryer. A part of the water vapor is recirculated back to the dryer for fluidization, and the balance is directed to a scrubber-type condenser. The non-condensable (odorous) gases within the water vapor are separated from the condensed water and injected into the reactor; thus, the issue of odor is minimized. The condensed water is directed to the WWTP, upon which approximately 30% of the total nitrogen within the raw sludge feed is returned to the WWTP.

The mix of dry sludge and sand is fed into the reactor, where the combustion temperature is maintained at 850 °C to ensure complete destruction of unwanted organic compounds (pathogens, drug residuals, microplastics, etc.). The temperature of the insulated reactor is controlled by dividing the internally circulating sand between a cooled and a non-cooled downcomer section. The reactor is fluidized with preheated combustion air. The ash formed in the combustion is carried with flue gases and extracted at two points: the majority of the ash (>95 wt%), the so-called product ash, is collected through a hot cyclone after the air preheater, while the finer ash is removed by a bag filter as by-product ash. A wet scrubber is used to remove sulfur from the flue gas, after which the purified gas is sent to a stack. The discharge water from the scrubber contains $SO_4$ and is directed to a nearby river system according to the environmental permit (PSAVI/891/2017).

Only a fraction of the water in the sludge ends up in the reactor off-gas, reducing the flue gas volume and, therefore, the costs of flue gas handling. The condenser and the reactor both require external cooling; hence, excess heat can be extracted from the process. This heat can be used, e.g., for district heat production. The product-ash is relatively high in nutrients and sufficiently low in heavy metals. Therefore, it can be further processed and used as a fertilizer in forestry or farming.

The drying fumes (water vapor and non-condensable gases) from the fluidized bed dryer are sent to a condenser. The condensate contains 20–30% of the total nitrogen included in the dewatered sludge entering the drying process, and over 90% of this nitrogen is in the form of ammonium ions (Eurofins, 2020). Nitrogen can be recovered to produce

**Table 3**
LCI data for combustion scenarios (S2.1 and S2.2).

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Combustion | | | Endev, 2020 |
| Electricity demand | 147 | kWh/t sludge | |
| LPG demand | 0.804 | kg/t sludge | |
| Fluidizing sand | 7.5 | kg/t sludge | |
| Heat efficiency | 80 | % | |
| Nitrogen to condensate | 30 | % | |
| Ash to product ash | 95 | % | |
| NaOH demand | 4.8 | kg/t sludge | |
| Combustion emissions | | | Endev, 2020; Yoshida et al., 2018, Hermann, 2021 |
| SO$_2$ | 2 | % of S | |
| N$_2$O | 79 | mg/kg TS | |
| Dust | 0.15 | mg/kg TS | |
| HCl | 4.9 | mg/kg TS | |
| HF | 0.39 | mg/kg TS | |
| NO$_2$ | 5224 | mg/kg TS | |
| CO | 20 | mg/kg TS | |
| Dioxin | 0.13 | µg/kg TS | |
| Hg | 0.33 | µg/kg TS | |
| Cd | 0.15 | µg/kg TS | |
| Pb | 0.26 | µg/kg TS | |
| Transfer coefficient to product-ash | | | Eurofins, 2020 |
| As | 84 | % | |
| Cd | 93 | % | |
| Cr | 88 | % | |
| Cu | 92 | % | |
| Hg | 57 | % | |
| Ni | 89 | % | |
| Pb | 93 | % | |
| Zn | 94 | % | |
| P | 94 | % | |
| K | 92 | % | |
| Ca | 90 | % | |
| Condensate to WWTP | | | HSY, 2021 |
| Electricity demand | 0.48 | kWh/t | |
| N removal | 0.48 | kWh/t condensate | |
| Ash treatment | 91 | % | |
| P usability | | | Ylivainio et al., 2020 |
| Granulation electricity | 10 | % | Havukainen et al., 2018 |
| Transport diesel use | 4.25 | kWh/t | Havukainen et al., 2018 |
| Spreading | 2.35 | l/t | Havukainen et al., 2018 |
| Ash landfill diesel use | 5 | l/t | Liikanen et al., 2018 |
| Ash leaching | 0.46 | l/t | Lynn et al., 2018 |
| As | | | |
| Cd | 0.8 | % | |
| Cr | 0.012 | % | |
| Cu | 0.040 | % | |
| Hg | 0.003 | % | |
| Ni | 2.2 | % | |
| Pb | 0.020 | % | |
| Zn | 0.012 | % | |
| Transport distances | 0.011 | % | Statistics Finland, 2017 |
| Fertilizer | | | |
| Sand | 43 | km | |
| NaOH | 19 | km | |
| H$_2$SO$_4$ | 201 | km | |
| Stripping and scrubbing | 201 | km | |
| Electricity demand | 0.48 | kWh/t condensate | Vaneeckhaute et al., 2017; Batstone et al., 2015 |
| NaOH demand | 91 | % | Batstone et al., 2015; Ervasti et al., 2018 |
| H$_2$SO$_4$ demand | | | Batstone et al., 2015 |
| | 10 | % | Eurofins, 2020 |

**Table 3** (*continued*)

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| NH$_4$-N in condensate Recovery efficiency | 4.25 | kWh/t | Provolo et al., 2017; Vaneeckhaute et al., 2017; |

ammonium fertilizers, such as ammonium sulfate. In this regard, a stripper is introduced into the system in S2.1. In air stripping, the condensate enters the stripper and NaOH is added to increase the pH, which is the most vital parameter to maximize the stripping efficiency. However, heat is also significant, and the higher the temperature, the higher the efficiency of ammonia recovery; a temperature range from 30 °C to 70 °C results in an efficiency between 89% and 92% (Guštin and Marinšek-Logar, 2011). Air is added in the stripper to convert ammonium to ammonia gas; eventually, ammonia gas is absorbed in sulfuric acid in a scrubber. For the stripping–scrubbing combination, according to Provolo et al. (2017), a nitrogen recovery efficiency of up to 90% could be achieved with a pH value of 7–9 in the stripping process, whereas Vaneeckhaute et al. (2017) reported an efficiency between 80% and 90%. In this study, heating is not required in the condensate stripping process, as the temperature of the condensate is around 60 °C. The energy and material demand as well as the nitrogen recovery for both the stripper and the scrubber are summarized in Table 3.

### 2.2.4. Sewage sludge digestate pyrolysis

In S3, dewatered digestate is directed to a pyrolysis process (Figure SI-3). The data are collected mainly from laboratory scale tests conducted for the development of the pyrolysis pilot plant for HSY (Huber, 2020; Kainulainen, 2020). Due to lower maturity, data uncertainty is greater in this technology compared with composting and combustion. The uncertainty is managed by conducting a sensitivity analysis.

Wood waste is mixed with digestate to ensure that the pyrolysis process is energy self-sufficient and to increase the carbon content of the produced biochar. First, the feedstock enters a thermal dryer, where most of the moisture is removed and the feedstock has achieved a suitable condition to be processed in the pyrolysis reactor. If wet digestate is pyrolyzed, steam will be generated. This has two main disadvantages: i) the moisture will condense and the end-product will be more diluted and ii) the amount of non-condensable gases will increase (Syed-Hassan et al., 2017). The pyrolysis reactor is designed to operate in the temperature range of 450–650 °C. The pyrolysis gas exiting the reactor, which possibly includes tar, is led to a combustion chamber. The heat recovered from the gas is circulated back to the thermal dryer and pyrolysis reactor. Bag filters are located at the exit of exhaust gases to capture contaminants and to filter the exhaust gas (HSY, 2019b). The main parameters and transfer coefficients, end product utilization, and transport distances are summarized in Table 4.

The drying fumes from thermal drying are led to a scrubber to remove odor and recover nitrogen. In air scrubbing, nitrogen-containing air is introduced from the bottom of the scrubber and liquid acid is either sprayed or passed through a packed column to absorb ammonia and produce ammonium sulfate liquid. In acid scrubbing, the liquid-to-gas ratio, pH, temperature, and concentration of ammonium ions have a substantial effect on the quality of the product. In the reaction of ammonia gas and sulfuric acid, equilibrium needs to shift toward ammonium to maximize nitrogen recovery (Saud et al., 2020). To achieve this equilibrium, a pH range between 1 and 5 plays an important role. Similarly, a suitable liquid-to-gas ratio (L/G) also increases the efficiency of ammonia recovery. The L/G ratio for countercurrent packed-bed scrubbers depends on the flow rate of liquid in such a way that the liquid should wet the packing completely but avoid flooding in the column. It is usually 11–13 times the minimum flow rate (Schnelle and Brown, 2016). For spray scrubbers, an L/G ratio of $2.4 \times 10^{-4}$ is

**Table 4**
LCI data of pyrolysis (S3).

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Electricity use | 100 | kWh/t feedstock | Huber, 2020 |
| Liquified petroleum gas (LPG) use | 0.03 | kg/t dried feedstock | Huber, 2020 |
| Wood waste | 418 | kg/t sludge | Huber, 2020 |
| Nitrogen to vapor | 35 | % of N in sludge | Gareis, 2020 |
| Transfer coefficient to biochar | | | Huber, 2020 |
| VS | 11 | % | |
| Ash | 100 | % | |
| Water | 0 | % | |
| Transfer coefficient to biochar | | | Huber, 2020 |
| As | 54 | % | |
| Cd | 61 | % | |
| Cr | 53 | % | |
| Cu | 57 | % | |
| Hg | 7 | % | |
| Ni | 58 | % | |
| Pb | 60 | % | |
| Zn | 51 | % | |
| C | 33 | % | |
| N | 28 | % | |
| P | 61 | % | |
| K | 100 | % | |
| Ca | 59 | % | |
| S | 0 | % | |
| Cl | 0 | % | |
| Pyrolysis gas | | | Huber, 2020 |
| Flue gas | 7.4 | m$^3$/kg | |
| NaOH demand | 3.5 | kg/kg S | |
| $SO_2$ removal | 96 | % | |
| NOx | 188 | mg/m$^3$ | |
| HCl | 6.7 | mg/m$^3$ | |
| Biochar and ammonium sulfate utilization | | | |
| Biochar P usability | 6 | % | Ylivainio et al., 2020 |
| Biochar N usability | 6.7 | % | Yuan et al., 2016 |
| Field spreading | 0.14 | l/t | Havukainen et al., 2020 |
| Ammonium sulfate | 22 | % | Calculated |
| Transport | | | |
| Biochar | 43 | km | Mölsä, 2019 |
| Fertilizer | 43 | km | Statistics Finland, 2017 |
| $H_2SO_4$ | 201 | km | Statistics Finland, 2017 |
| Ammonia scrubbing | | | |
| $H_2SO_4$ demand | 3.6 | kg/kg $NH_3$-N | Batstone et al., 2015 |
| Electricity demand | 2.5 | kWh/kg $NH_3$-N | Hadlocon et al., 2015 |
| Recovery efficiency | 94 | % of $NH_3$-N | Melse and Ogink, 2005; Tampio et al., 2016 Vaneeckhaute et al., 2017 |

mentioned in the literature (Hadlocon et al., 2015). The sulfuric acid and electricity demands and the recovery efficiency for the scrubbing process are presented in Table 4. A part of the ammonium sulfate liquid is added to the biochar to increase the moisture content; then, the mixture is transported to be spread on arable land (Table 4).

### 2.2.5. Wood waste combustion

The wood waste that is directed to pyrolysis in S3 would otherwise be utilized in district heat production. The produced heat will substitute marginal district heat in Finland. The produced ash is granulated and utilized as a forest fertilizer. The parameters, combustion emissions, and information regarding ash utilization are summarized in Table 5.

### 2.2.6. Substituted processes

The substituted material and energy products as well as the bases for substitution are summarized in Supplementary Information SI-4 in Table SI-6. The compost produced in S1 contains nitrogen and phosphorus, and based on the amount of soluble nitrogen and usable phosphorous, it substitutes a mineral fertilizer when utilized on arable land.

**Table 5**
LCI data of wood waste combustion (Brassard et al., 2014; Havukainen et al., 2018).

| Parameter | Value | Unit |
|---|---|---|
| Comminution diesel demand | 3.8 | l / t wood |
| Heat efficiency | 85 | % |
| Combustion emissions | | |
| $CH_4$ | 5.7 | mg/MJ |
| $NO_2$ | 2.7 | mg/MJ |
| $SO_2$ | 21 | mg/MJ |
| HCL | 0.42 | mg/MJ |
| CO | 574 | mg/MJ |
| $NH_3$ | 2 | mg/MJ |
| Ash utilization | | |
| Granulation electricity demand | 0.0210 | kWh/t |
| Transport diesel demand | 4.3 | l/t |
| Spreading diesel demand | 2.4 | l/t |

The produced compost soil is substituting a regular gardening soil, made by mixing two types of Sphagnum peat (light colored, less decomposed peat and dark colored, more decomposed peat, in total 80–90%), sand (10–20%), limestone (6–7 kg/m$^3$ soil product), and fertilizers (1 kg/m$^3$ soil product). The density of soil product is, on average, 350 kg/m$^3$. The substituted fertilizer contains 9% nitrogen, 4% phosphorus, and 12% sodium (Hartikainen, 2020). The combustion scenarios (S2.1 and S2.2) produce (i) district heat, which displaces marginal heat; (ii) product ash, which is utilized as a forest fertilizer; and (iii) ammonium sulfate liquid (S2.2), which is used on arable land. The pyrolysis scenario (S3) produces biochar and ammonium sulfate liquid, which are utilized on arable land.

### 2.3. Sensitivity analysis

The robustness of the results was analyzed in three parts. First, a contribution analysis was performed to show the main contributing processes. Second, a perturbation analysis was used to show the most sensitive parameters by utilizing sensitivity ratios (SR) according to Clavreul et al. (2012). SR is calculated as a ratio of two relative changes by dividing the relative change of the total result by the relative change of the individual parameter. According to Heijungs and Kleijn (2001), parameters with SR values over 0.8 are important, those with SR values over 1 are especially important, and those with SR values lower than 0.2 have only a minor influence on the overall results. However, according to Bisinella et al. (2016), the value of SR is dependent on the impact category, and therefore the SR results should be evaluated within an impact category rather than compared between them. Third, a "high" and "low" performance sensitivity analysis was used to calculate the range of the net result utilizing the parameter values presented in Table SI-8 for the selected impact categories.

## 3. Results

For the studied scenarios and selected impact categories, the results of the contribution analysis showing the range of net results with error bars are summarized in Fig. 2, whereas Fig. 3 presents the relative contribution of the processes to the produced and avoided emissions. The results are presented in detail in the supplementary material file (Tables SI-10–SI-13). In addition, the results of the contribution analysis for other impact categories are also presented in the supplementary material (Figure SI-5). According to the results, S2.2 shows the lowest net impact for climate change; S1 for fine particulate matter, marine eutrophication, mineral resource scarcity, and human toxicity (cancer); and S2.1 for terrestrial acidification, freshwater eutrophication, and terrestrial ecotoxicity.

The pyrolysis scenario (S3) yields the widest range of results in most impact categories. This is due to the greater uncertainty of the pyrolysis data, which are based on laboratory results, unlike S1 and S2, for which
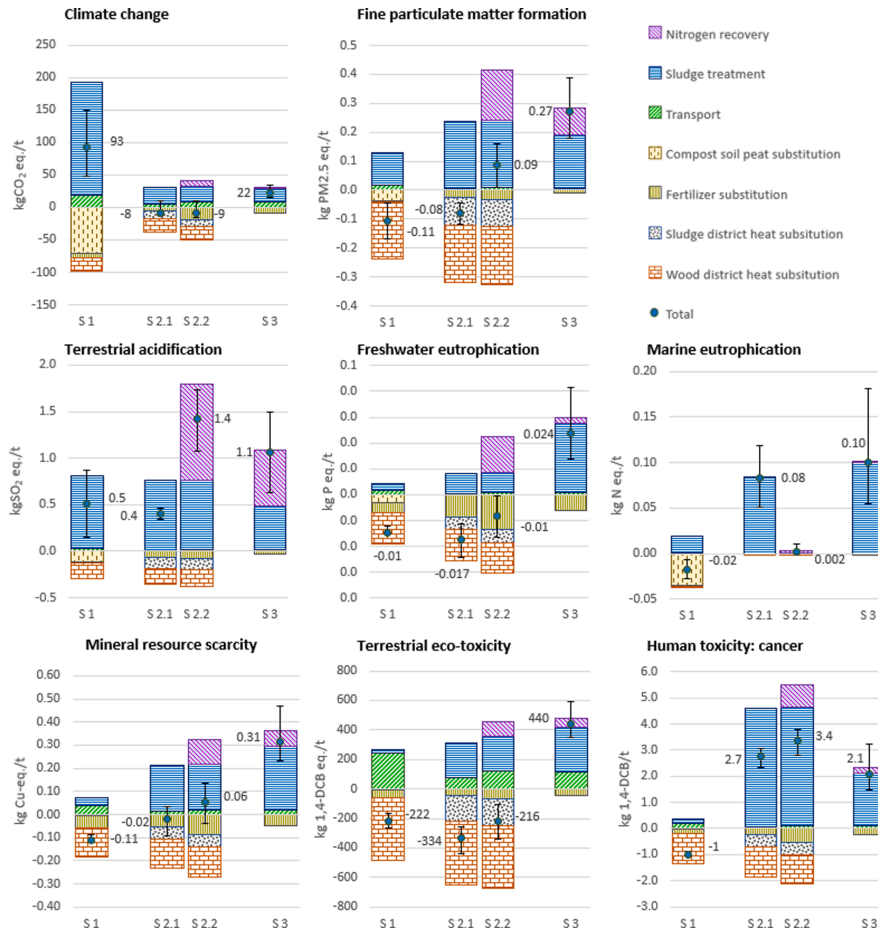
**Fig. 2.** LCIA contribution assessment of the scenarios: S1 – composting, S2.1 and S2.2 – combustion, and S3 – pyrolysis for the selected impact categories of the ReCiPe 2016 methodology (range of net results is shown with error bars).

data were collected from existing facilities. Even when considering the range of results, the results do not suggest that S3 could show a better performance than the other scenarios in most impact categories. The pyrolysis scenario (S3) performs better than both the combustion scenarios S2.1 and S2.2 in the human toxicity (cancer) impact category and better than S1 in the case of climate change. Among the main reasons for the wide range of net results for S3 are the high and low performance values of the parameters related to flue gas emissions, such as emitted flue gas emissions in the case of particle formation and terrestrial acidification and NaOH demand for cleaning the flue gas in other impact categories. Additional reasons for the wide range are the values of the parameters related to electricity consumption in pyrolysis, electricity consumption in the scrubbing process, and change in the production volumes of biochar and ammonium sulfate liquid, which affect the transport emissions.

Then, a question could be raised as to how well a sensitivity analysis,

which is based on high and low performance values for parameters, succeeds in creating a reliable range of net results based on this quite uncertain data for the pyrolysis process. It could be that the most important initial parameter values are already too far from the values attainable in a larger-scale facility and therefore provide a biased starting point for estimating the high and low performance values. For example, if a more elaborate or mature flue gas cleaning process would be in place in a larger-scale pyrolysis facility, perhaps the particle emission impacts would be on the same level as those due to combustion. To tackle this issue, more data would be needed, especially from full scale plants, to further validate the results of this study. It could be argued, however, that this study provides information on the technology and operational parameters that are critical and that more focus should be given to these parameters when obtaining primary data. There might also be potential for the pyrolysis technology to develop more rapidly than the existing technologies, which have been optimized to a greater
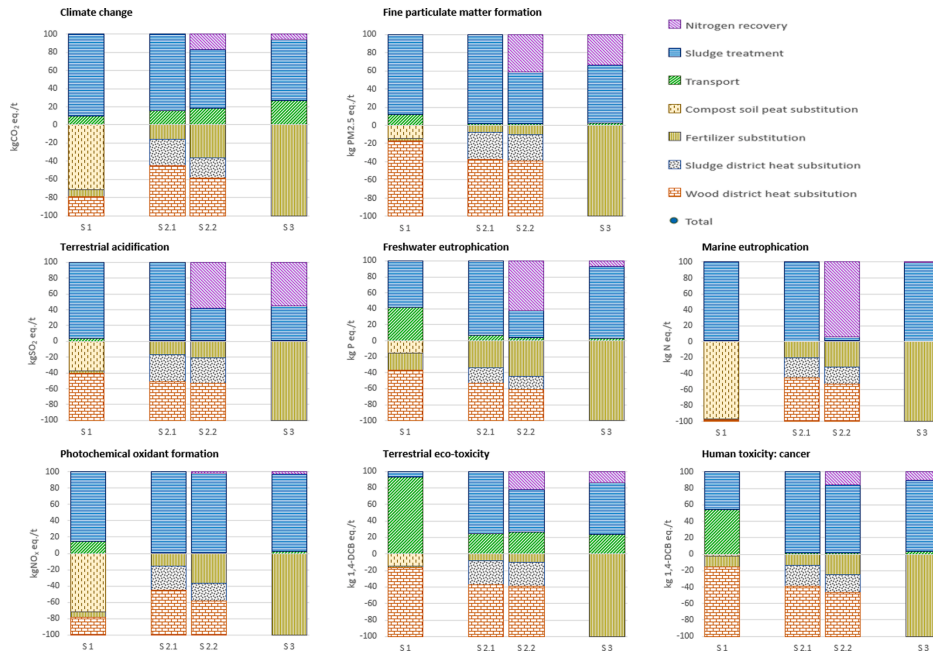
**Fig. 3.** LCIA results: relative contribution of direct emissions and substituted emissions.

extent already. This would then, with time, change the relative performance of these technologies.

In the combustion scenarios S2.1 and S2.2, the impact of the N2O emissions onto the climate change impact category is significant, causing 22% and 16% of the total emissions, respectively. Furthermore, in S2.2, the recovery of nitrogen from the condensate appears to increase the net impacts for most of the studied impact categories as compared to S2.1 without nitrogen recovery. In the case of climate change, the recovery of nitrogen from the condensate increases the emissions by almost 50%, but the avoided emissions increase more because of the use of ammonium sulfate, resulting in a slightly lower net impact for S2.2. More than 80% of the increase in emissions is caused by the production of NaOH and 10% by the production of $H_2SO_4$. The production of these chemicals (NaOH and $H_2SO_4$) is also mainly responsible for the increase in emissions from freshwater eutrophication, scarcity of mineral resources, and the toxicity impact categories (80–94%). In the case of marine eutrophication, the direction of the nitrogen-containing condensate to the WWTP in S2.1 results in emissions that are almost 95% higher compared to S2.2. Therefore, a lower net impact for S2.2 is obtained. If nitrogen recovery could be achieved by adsorption straight from the condensate or from stripping gas, the demand for chemicals and thereby emissions from nitrogen recovery could be reduced.

The relative contribution results in Fig. 3 show the contribution of the process phases separately for the produced and avoided emissions. Treatment of sludge causes the majority of the emissions in almost all the studied impact categories in each scenario, except for toxicity impacts in S1, where transport mainly causes the emissions (more than 90%), and in S2.2, which has the highest impacts caused by nitrogen recovery on the acidification and eutrophication impact categories (60–85%). In the case of avoided emissions, avoiding peat production by displacing the garden soil is important, e.g., for climate change

contributing 70% of the avoided emissions. In S2.1 and S2.2, district heat production from the digestate is important, yielding 20–30% of the avoided emissions across the impact categories. The emission reduction from displacing the marginal district heat by district heat from wood waste has a significant impact in scenarios S1, S2.1, and S2.2 (e.g., 40–50% across the impact categories for S2.1 and S2.2).

S3 could avoid impacts only from fertilizer substitution since there is no energy production due to wood waste being used in the pyrolysis process. The avoided impacts of phosphorus and nitrogen in biochar are responsible for 60% of the avoided impacts, and ammonium sulfate is responsible for the remaining 40%. Of the avoided impacts of biochar, 60% are due to avoiding phosphorus fertilizer and 40% are due to avoiding nitrogen fertilizer production. Frišták et al. (2018) also found that pyrolysis char can be a phosphorus-rich soil amendment. The impact of adding biochar to the ground due to carbon sequestration, the potential of which has been mentioned by Cao and Pawłowski (2013), was not accounted for; however, by applying biochar to the ground, 60 kg of carbon (30% of input carbon) from sewage sludge and wood waste per functional unit would end up going to the ground in a less degradable form, which could provide additional climate impact benefits and potentially improve the soil quality.

In S1, sludge treatment emissions are mainly caused by composting air emissions (30–50% for climate change, particulate emissions, and terrestrial acidification), peat production emissions (60–100% for eutrophication impact categories), and sand production (40–80% for toxicity impacts and mineral resource scarcity). In S2.1 and S2.2, sludge treatment emissions are derived mainly from the production of NaOH (40% for climate change), combustion emissions (80–90% for particle emissions, acidification, and human toxicity), and electricity demand (90–100% for eutrophication, mineral scarcity, and terrestrial ecotoxicity). In S3, the main emissions from sludge treatment are caused by

electricity usage in the pyrolysis process (50–90% for climate change, particulate emissions, acidification, and eutrophication), combustion of the pyrolysis gas (70% for mineral resource scarcity and eco-toxicity) and directing the residual ammonium sulfate to the WWTP (95% for marine eutrophication).

The SRs for the selected parameters that show high values for the studied impact categories can be found in the supplementary material (Table SI-14 and Figures SI-6–SI-9). In S1, the compost soil displacement ratio is the most sensitive parameter across most impact categories. As the displacement was based on volume, the densities of the compost soil and garden soil have a great impact on the results. Besides the share of soil displacement, the emissions from the composting process are important for the climate change impact category. As the information about these emissions was collected from the literature, the results could change significantly when facility-based data are available and used. In the combustion scenarios S2.1 and S2.2, the heat efficiency, electricity demand, and the share of phosphorus ending up in the product ash are sensitive parameters; however, these parameters can be quite accurately determined by measurements. Thus, the degree of uncertainty caused by them is low. When nitrogen recovery is included in the combustion process (S2.2), the share of nitrogen ending up in the condensate, NaOH demand, and recovery efficiency show high sensitivity. In S3, the parameters related to the combustion of pyrolysis gas, such as NaOH demand, NOx emissions, and the sulfur transfer coefficient, are among the most sensitive parameters in addition to the wood demand. The pyrolysis gas combustion parameters are dictated by the operating conditions of the pyrolysis reactor, and the wood demand is determined based on the heat demand of drying and pyrolysis.

## 4. Discussion

### 4.1. Previous studies

The climate impacts obtained in this study are in line with previous studies. Johansson et al. (2008) obtained a climate impact with a rather wide range for composting and subsequent application on land, i.e., from −97 to 3000 $kgCO_2$,eq./$t_{TS}$, whereas in the study of Yoshida et al. (2013), the climate impact for a scenario in which the digestate was first composted and then applied on land was found to be between − 193 and 249 $kgCO_2$,eq./$t_{TS}$. In relation to the TS content in dewatered sludge (Fig. 2 shows the values per 1 t of dewatered sewage sludge digestate), the composting scenario (S1) resulted in a climate impact of 337 $kgCO_2$, eq./$t_{TS}$ (range of 176–536 $kgCO_2$,eq./$t_{TS}$.). The combustion scenarios (the average of S2.1 and S2.2) resulted in a climate impact of −29 $kgCO_2$,eq./$t_{TS}$ (−69 to 6 $kgCO_2$,eq./$t_{TS}$), which is at the lower end of the ranges found by Zhang et al. (2019) and Yoshida et al. (2013), i.e., −32–103 and 36–3183 $kgCO_2$,eq./$t_{TS}$, respectively. The emission of $N_2O$ from mono-incineration of sewage sludge is an important aspect in terms of climate change impact (Svoboda et al., 2006). The possibilities to reduce these emissions have been discussed, for example, by Korving et al., 2010.

In the current study, the climate impact of pyrolysis (S3) was found to be 79 $kgCO_2$,eq./$t_{TS}$ (range of 55–124 $kgCO_2$,eq./$t_{TS}$), which is at the higher end of the range obtained by Cao and Pawlowski (2013), i.e., 12–108 $kgCO_2$,eq./$t_{TS}$. The main difference between this study and the study by Cao and Pawlowski (2013) is that they used a fossil fuel for drying while obtaining an emission reduction from pyrolysis oil; in the current study, the energy was acquired mainly from wood waste.

Based on the recent literature, the technologies investigated in this study have proven to be feasible for sewage sludge treatment. Pyrolysis of dewatered digestate is a promising technology, providing possibilities for energy recovery and sludge volume reduction (Lacroix et al., 2014; Syed-Hassan et al., 2017). According to Li and Feng (2018) and Opatokun et al. (2017), anaerobic digestion followed by pyrolysis of dewatered digestate can generate net energy and alleviate water depletion and climate impacts. The findings by Mancini et al. (2019) suggest that anaerobic digestion and subsequent composting could provide environmental benefits in sewage sludge treatment, while the main concerns for environmental deterioration include human toxicity, freshwater ecotoxicity and marine ecotoxicity. Tonini et al. (2019) investigated the environmental performance of sewage sludge digestate incineration with phosphorus recovery from ashes by the ASH DEC technology (Havukainen et al., 2016b; Hermann and Schaaf, 2019) or by acid recovery and precipitation of phosphorus from sewage sludge after WASSTRIP (Waste Activated Sludge Stripping to Remove Internal Phosphorus) and anaerobic digestion processes. The climate impact of sewage sludge treatment and subsequent recovery of phosphorus was found to be lower than the combined impact of managing sewage sludge by conventional methods (spreading on land or co-incineration) and producing phosphorus from the mining of phosphate rock

### 4.2. Future prospects and contextualization

To date, composting is the most prevalent method for sewage sludge digestate treatment in Finland. Currently, many crop buyers are reluctant to purchase crop that has been grown in a field where sewage-sludge-based renewable fertilizers are used because the sludge contains residual pharmaceuticals and microplastics. Therefore, sewage sludge compost is directed to an increasing extent to soil production or sold for grass soil and gardening soil. Such a development has been an important driver for the search for alternative treatment methods, including thermal treatment. The technologies for thermal treatment of sewage sludge are not yet common in Finland; only one such industrial-scale combustion plant exists, and a pilot plant for pyrolysis is currently being built in the HSY region. The pyrolysis process is partially driven because HSY is building a WWTP and there is need for new sewage sludge treatment capacity when the compost product is becoming less attractive.

Furthermore, the availability of peat to be used as a support medium in composting in Finland can be forecasted to decrease due to the goal to achieve carbon negativity in Finland by 2030 (Finnish Government, 2021). The use of peat in energy production decreased even more rapidly than anticipated by the Finnish government, when they set the goal of ending peat use in energy production by 2030 (YLE, 2021). Currently, supplementing materials for peat in growth medium manufacturing are sought after (Biolan Ltd., 2021). Changing the peat to another support material would reduce the climate impact of composting; e.g., forest residue based wood chips have a climate impact of 20–30 $kgCO_2$,eq./t (Havukainen et al., 2018), while the emissions from peat production were 160 $kgCO_2$,eq./t in the current study.

The selection of suitable technology could incorporate the consideration of the fate of nutrients and carbon included in sewage sludge digestate. The composting process can retain both nitrogen and phosphorus. Although pyrolysis and combustion would release most of the nitrogen, phosphorus would be left in the residual biochar or ash, albeit in a less suitable form for plants (Ylivainio et al., 2020). Furthermore, the composting process would retain most of the carbon in digestate since biodegradable carbon is mainly consumed in the AD process. Pyrolysis would retain part of the carbon in the form of biochar. The carbon in biochar could potentially provide carbon sequestration benefits (Cao and Pawlowski, 2013). Combustion would consume almost all the carbon but destroy all harmful organic compounds.

The fate of heavy metals is more uncertain, and it is not clear how the solubility of these metals differs between compost, biochar, and ash. In the composting process, all the heavy metals end up in the compost, and in the production of biochar, most of the heavy metals end up in biochar. In combustion with a staged removal of dust, a higher concentration of heavy metals is found in the by-product ash. If sewage sludge contains a significant concentration of heavy metals, the ashes can be treated with a phosphorus recovery technology (Havukainen et al., 2016a).

Additional environmental emissions or impacts that could be potentially important in dewatered sewage sludge digestate treatment

are dispersion of drug residues, bacterial resistance, and spreading of microplastics. These are not, at the moment, fully addressed in LCA, but they can be important from the technology choice point of view since the application of products based on sewage sludge, such as compost on land, is losing popularity due to the concerns related to these potential impacts.

It should be kept in mind that there is not such an ultimate disposal method for sewage sludge treatment that applies to each and every situation (Ding et al., 2021). Composting is feasible when the produced compost can be utilized and the content of hazardous contaminants, drug residues and microplastics in the sludge is low enough. Combustion, on the other hand, is efficient in terms of volume reduction, and the phosphorus in the remaining ash can be utilized. Furthermore, additional benefits can be obtained with heat production. Pyrolysis can be feasible when the carbon in the dewatered digestate is considered valuable and there is additional waste biomass or other biomass to be used in providing energy self-sufficiency for the pyrolysis process and to increase the carbon content of the produced biochar.

## 5. Conclusions

The results of this study indicate that in a Finnish context, combustion or composting of dewatered sewage sludge performed environmentally better than pyrolysis. Composting could be feasible when there is a possibility to utilize the remaining compost. In combustion, the volume of the sludge is significantly reduced and the resulting ash can be utilized as a nutrient-rich fertilizer. Overall, the main impacts were caused by the sludge treatment processes itself, and transport emissions were less important (except for toxicity impact categories). In the case of composting, the associated air emissions and impacts from peat consumption contributed with significant impacts across several categories but also provided significant emission savings from substitution of gardening soil containing peat. The added emissions from the recovery of nitrogen from the condensate in the combustion scenario completely outweighed the benefits achieved from substitution of fertilizer production, which indicates that nitrogen recovery efficiency should improve while process chemistry requirements should reduce. For pyrolysis, wood waste removes the requirement for additional fuel in the pyrolysis process; however, alternative utilization of the same wood waste for district heating is lost. Combined with uncertainties of full-scale process performance, the fate of heavy metals, and carbon, pyrolysis was the least preferred option in several impact categories. In the case of pyrolysis, important technology parameters are related to the ratio of wood waste and sludge so that enough pyrolysis gas is produced to provide heat for the drying and pyrolysis processes, pyrolysis gas combustion, cleaning of flue gas, and electricity demand of the processes. It is recommended that further process data are provided to improve the environmental assessment of pyrolysis. So far, several environmental consequences that are of potentially critical importance have not been considered in LCA, e.g., dispersion of drug residues, bacterial resistance, and spreading of microplastics. If these potential impacts are to be avoided, composting processes may not be feasible options.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

## Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2021.11.005.

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# Publication IV

Saud, A., Havukainen J., Peltola P., and Horttanainen M.
**Environmental Performance of Nitrogen Recovery from Reject Water of Sewage Sludge Treatment Based on Life Cycle Assessment**

*Article*

# Environmental Performance of Nitrogen Recovery from Reject Water of Sewage Sludge Treatment Based on Life Cycle Assessment

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**Abstract:** Recovering and recycling nitrogen available in waste streams would reduce the demand for conventional fossil-based fertilizers and contribute toward food security. Based on life cycle assessment (LCA), this study aimed to evaluate the environmental performance of nitrogen recovery for fertilizer purposes from sewage sludge treatment in a municipal wastewater treatment plant (WWTP). Utilizing either air stripping or pyrolysis-derived biochar adsorbent, nitrogen was recovered from ammonium-rich reject streams generated during mechanical dewatering and thermal drying of anaerobically digested sewage sludge. A wide range of results was obtained between different scenarios and different impact categories. Biochar-based nitrogen recovery showed the lowest global warming potential with net negative GHG (greenhouse gas) emissions of $-22.5$ kt $CO_2$,eq/FU (functional unit). Ammonia capture through air stripping caused a total GHG emission of 2 kt $CO_2$,eq/FU; while in the base case scenario without nitrogen recovery, a slightly lower GHG emission of 0.2 kt $CO_2$,eq/FU was obtained. This study contributes an analysis promoting the multifunctional nature of wastewater systems with integrated resource recovery for potential environmental and health benefits.

**Keywords:** adsorption of ammonia; biochar; Life Cycle Assessment (LCA); nitrogen recovery; sewage sludge; waste to energy

## 1. Introduction

Resource conservation and recovery have gained immense attention globally in the past few years. The concerns related to the growing population, excessive extraction and utilization of raw materials, irresponsible consumption, and scarcity of basic materials have gathered researchers and scientists to find solutions that benefit humans and help to mitigate the environmental burden. As a crucial component in the urban sewage system, wastewater treatment plants (WWTPs) extract organic and inorganic pollutants that would otherwise leak into the environment and create potential hazards to the ecosystem and human health. The ever-increasing demand for wastewater treatment also increases emissions and generates piles of sewage sludge [1], which, on the other hand, emphasizes the need to recover and reuse the resources available in wastewater. Global nutrient needs and waste-to-energy potential among the main drivers; future WWTPs as "ecologically sustainable" technological systems are expected to strengthen the energy–nutrient–water nexus and, thus, become an integral part of the circular economy [2].

Nutrient recycling from WWTPs reduces the demand for conventional fossil-based fertilizers and contributes toward food security. Nitrogen is the most limiting nutrient to crop production [3], yet its recovery from wastewater treatment has become a research focus only recently. Nitrogen fertilizers are manufactured through the energy-intensive Haber–Bosch process using natural gas, from atmospheric nitrogen to plant-available ammonium nitrogen ($NH_4$-N). Contributing up to 2% of global energy consumption and causing significant greenhouse gas (GHG) emissions, the extensive production of mineral

nitrogen for fertilizers via chemical synthesis has raised economic and environmental concerns. Globally, food production can utilize only 17% of the applied nitrogen fertilizer, while the rest is lost to water bodies and the atmosphere [4]. Moreover, part of the elemental nitrogen transformed into mineral fertilizer ends up in human waste in the form of urea and ammonium, and municipal WWTPs are required to remove this nitrogen to avoid eutrophication. The European Union (EU) Urban Waste Water Directive (91/271/EEC), established to prevent adverse effects of wastewater discharge into natural water streams, specifies a minimum reduction of 80% for phosphorus and 70–80% for nitrogen [5]. However, albeit widely employed in modern WWTPs, nitrogen removal brings no additional benefits besides complying with effluent concentration limits [4]. Instead, recovering nitrogen would allow better utilization of anthropogenic nitrogen sources while saving energy and raw materials.

Inlet wastewater streams are characterized by high volumes but low concentrations; hence, without a concentration step, they are too dilute for profitable resource recovery [4]. Nitrogen, nevertheless, accumulates in the activated sludge generated in the wastewater treatment process. The nitrogen is subsequently released back to the aqueous phase during anaerobic digestion, a method commonly used for sludge stabilization, and when the anaerobically digested sludge is mechanically dewatered for further processing, a nitrogen-rich liquid fraction (reject water) with $NH_4$-N concentrations of up to 1.5 g/L is formed [6,7]. Reject water is the most nitrogen-rich stream at a WWTP, containing 15–25% of the total nitrogen content, but less than 5% of the total volume of the influent wastewater [8]. Furthermore, the remaining solid fraction (sewage sludge) carries a notable amount of nitrogen, up to 8% (dry basis), among other major plant nutrients [9]. Targeting both these waste streams for nitrogen recovery would improve the total recovery rate and allow maximal utilization of the nitrogen sources available in WWTPs [10].

Handling excess sewage sludge produced during wastewater treatment is a common problem worldwide. In the EU alone, the amount of sewage sludge has increased enormously, by around 70% from 6.5 Mt to 10.9 Mt of dry matter during 1992–2015 [11]. Currently, sewage sludge is disposed of and reused in different ways in EU member countries, including landfilling (6%), composting and other applications (12%), agricultural use (35%), and incineration (37%) [12]. Because of increasingly stringent legislation, limited space available in landfills, and soaring environmental and health issues due to the presence of harmful contaminants, e.g., heavy metals, microplastics, pharmaceutical waste, pesticides, and substances found in personal care and household products, traditional methods such as landfilling and agricultural application after biological treatment are considered problematic [13,14]. Instead, thermochemical conversion processes, e.g., pyrolysis, gasification, and incineration, have attracted significant attention as an alternative route for sludge disposal. Via thermal processing, the quantity and toxicity of sewage sludge can be reduced with simultaneous recovery of the embedded energy and chemical assets [15,16]. While requiring advanced equipment and operations, the thermochemical conversion could provide superior economic performance, efficiency, and volume reduction compared to competing sludge management technologies [17].

Pyrolysis, a thermal degradation process under inert or anoxic conditions at moderate to high temperatures (300–700 °C), converts different types of sewage sludge (raw, digested, and waste-activated) into products with added value [18]. The process results in the production of liquid pyrolytic oil (bio-oil), solid biochar, and non-condensable gases (syngas) [19]. Bio-oil is considered a potential source of energy that can fuel boilers, combustion engines, and turbines. Alternatively, bio-oil can be upgraded and refined for specialty chemicals. Biochar has shown potential benefits as a phosphorous-rich soil amendment, a carbon-neutral fuel, a low-cost adsorbent, and a replacement for carbon black, among other applications promoting environmental remediation [20]. The yield and properties of pyrolysis products depend on several factors, such as temperature, residence time, pressure, and feedstock composition. Even after mechanical dewatering, sewage sludge contains a substantial amount of moisture (73–84%). Pyrolyzing wet sewage sludge will generate a

steam-rich atmosphere inside the reactor; consequently, the liquid product will be more diluted, and the amount of non-condensable gases will increase [13]. To avoid complexities and improve pyrolysis performance, the water content in sludge can be reduced to 5–10% via thermal pre-drying. During thermal drying, a considerable proportion of the nitrogen is released within the drying fumes, which, after condensation, results in a nitrogen-rich liquid stream (condensate) that can be directed to nitrogen recovery together with the reject water from mechanical dewatering [10].

The concepts of sustainability, resource recovery, and climate change mitigation have developed a growing interest in the modelling of sewage sludge treatment systems. Besides technoeconomic assessments, life cycle analysis (LCA) has a pronounced role in selecting suitable sludge management strategies in terms of different spatial and temporal scales [21]. LCA aims to quantify the environmental performance of a process or a system by accounting for the emissions associated with material and energy flows throughout the entire life cycle. Recently, Ding et al. [22] reviewed the progress in LCA research performed on sewage sludge management and compared the environmental sustainability of existing and emerging technologies with the purpose of nutrient recovery and energy saving. Lam et al. [23] provided a summary of 65 LCA studies with different methodological practices and different scopes of nutrient removal/recovery. Clearly, the focus has been on various strategies for phosphorous recovery, while few studies only have concentrated solely on nitrogen. Kar et al. [24] and van Zelm et al. [25] examined the life cycle environmental impact of nitrogen recycling from WWTPs, considering air stripping to recover ammonia from side streams generated during sludge dewatering. Despite the different conditions and assumptions, both studies showed overall environmental benefits of the integrated removal, recovery, and fertilizer production over ammonia removal-only systems.

This study aimed to evaluate the environmental performance of nitrogen recovery for fertilizer purposes from sewage sludge treatment in a municipal WWTP. Three different scenarios, one without and two with nitrogen recovery, were investigated and compared in terms of nitrogen recovery rate and potential environmental impacts. The base case scenario without nitrogen recovery included anaerobic digestion as a conventional method to stabilize raw sludge. Since biological sludge processing alone is insufficient to remove harmful substances, pyrolysis with thermal pre-drying was considered as a post-treatment method for sludge disposal. Utilizing either air stripping or pyrolysis-derived biochar adsorbent for nitrogen recovery, the reject water generated during mechanical dewatering and the condensate generated during thermal drying were targeted as a combined source of nitrogen.

## 2. Results

The results for the studied scenarios and selected impact categories (climate change with and without biogenic carbon, terrestrial acidification, and marine and freshwater eutrophication) are summarized in Figure 1. Figure 2 illustrates the relative contribution of the processes to the produced and avoided emissions. Moreover, the results are compiled and presented in detail in Tables S5 and S6 (Supplementary Material).

Concerning environmental performance, definitive conclusions of superiority cannot be straightforwardly drawn. Scenario S3 (AdBC) performs better in three of the five impact categories, including climate change with biogenic carbon, freshwater eutrophication, and marine water eutrophication, whereas S1 (CWWTP) shows the lowest net impact for the remaining two categories, namely, climate change without biogenic carbon and acidification. Scenario S2 (S&S) remains in the middle, except for yielding the highest net impact in two categories: climate change with biogenic carbon and freshwater eutrophication.

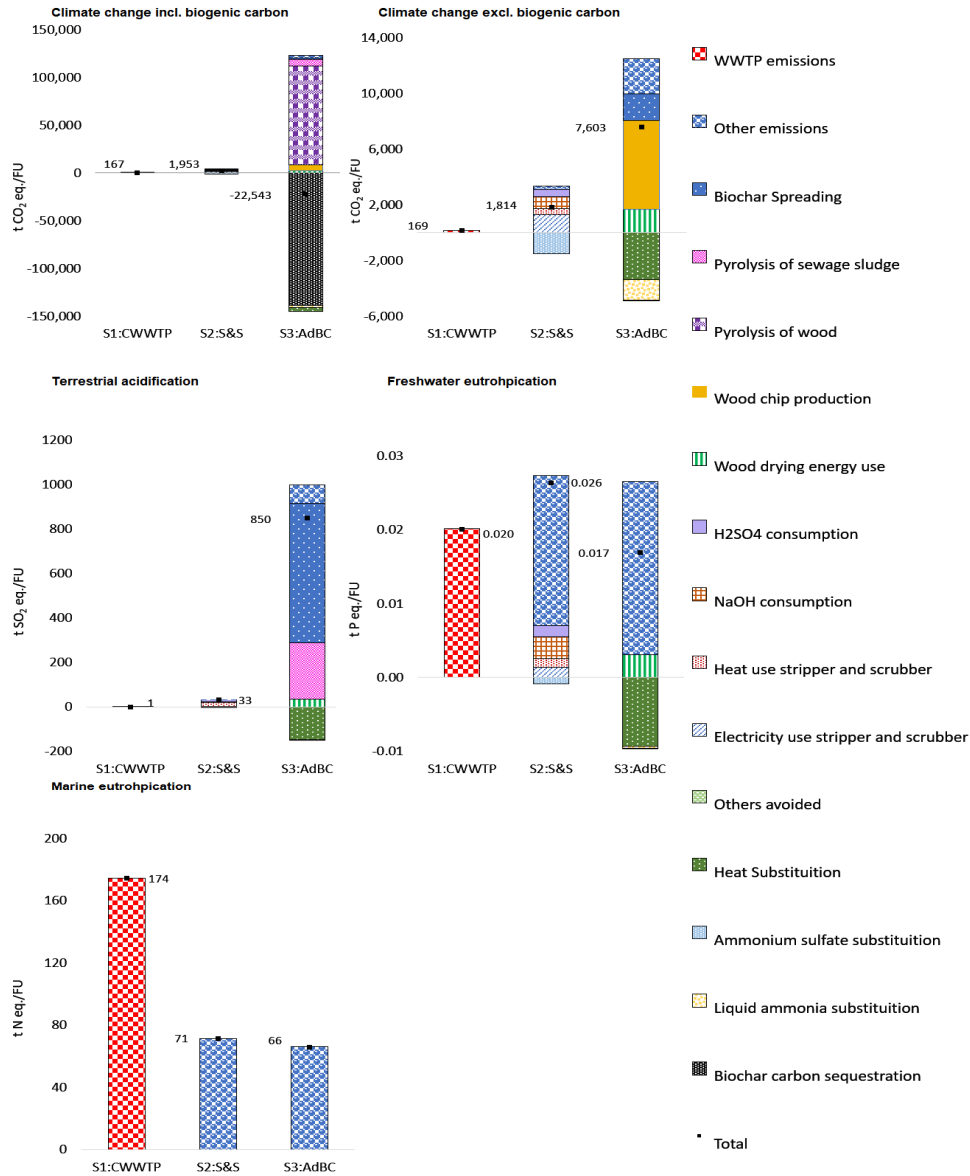**Figure 1.** LCIA results for the selected impact categories in S1 (CWWTP), S2 (S&S), and S3 (AdBC).
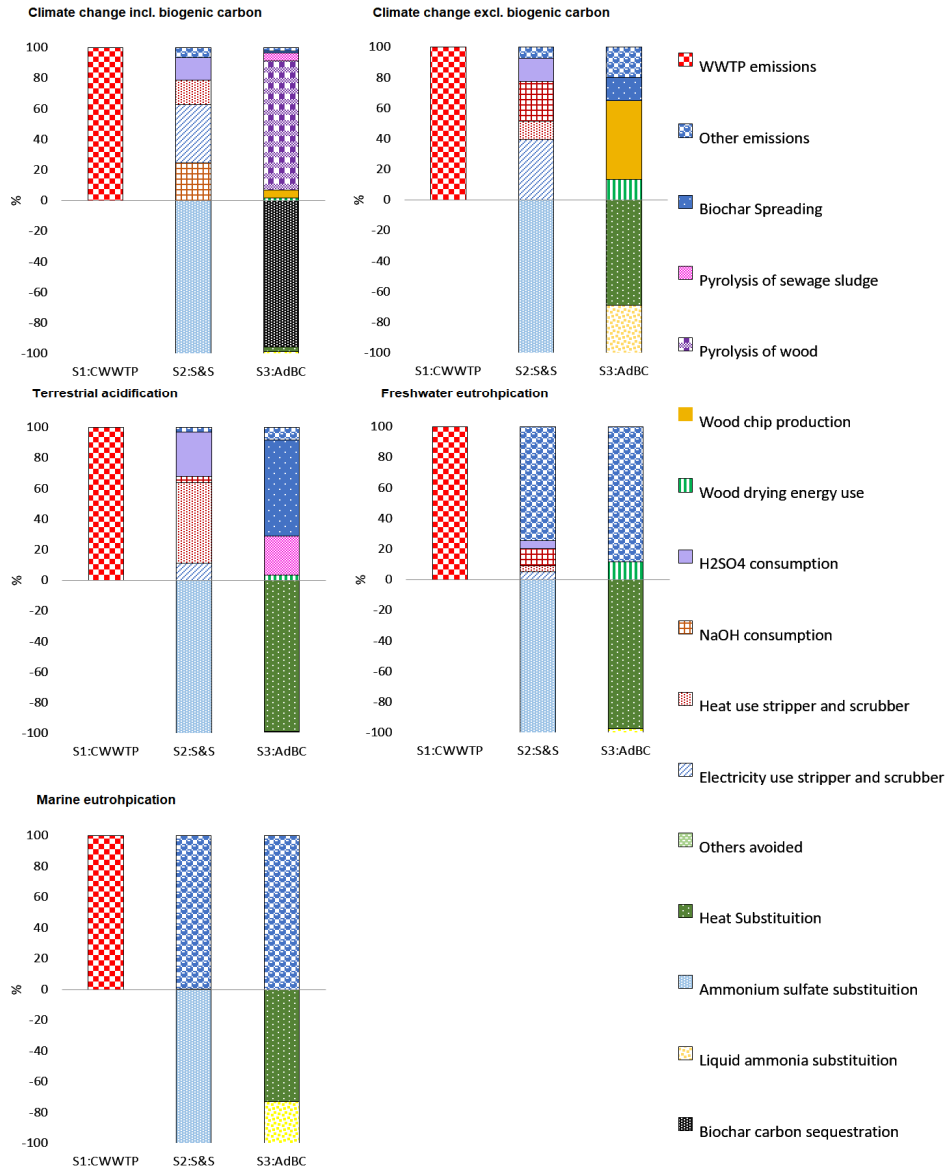
**Figure 2.** LCIA contribution assessment of scenarios S1 (CWWTP), S2 (S&S), and S3 (AdBC) for the selected impact categories.

Considering climate change with biogenic carbon, the total produced and avoided emissions from S3 (AdBC) are 120 kt $CO_2$,eq./FU and 145 kt $CO_2$,eq./FU, respectively. The

production of wood biochar yields significant emissions because wood pyrolysis requires a high amount of energy. On the other hand, 139 kt $CO_2$,eq./FU can be avoided because biochar is utilized for carbon sequestration, and a further 6 kt $CO_2$,eq./FU is avoided by substituting heat and fossil-based nitrogen fertilizers. In total, S2 (S&S) produces 3.5 t $CO_2$,eq./FU, but avoids 1.5 t $CO_2$,eq./FU. Notable emissions originate from the combined stripping and scrubbing process, which consumes electricity (1 kt $CO_2$,eq./FU) and chemicals (NaOH 0.9 kt $CO_2$,eq./FU, sulfuric acid 0.5 kt $CO_2$,eq./FU). The baseline scenario S1 (CWWTP) generates only direct emissions (0.2 kt $CO_2$,eq./FU) in this category.

In the case of climate change without biogenic carbon, the direct emissions from S3 (AdBC) are 12 kt $CO_2$,eq./FU, while a total of 5 kt $CO_2$,eq./FU is avoided. When comparing climate change with biogenic carbon, a very different net impact is obtained due to biochar-based carbon sequestration. In S2 (S&S) and S1 (CWWTP), the net impacts to the climate remain almost the same.

Table 1 shows the nitrogen recovery rate in each scenario. S3 (AdBC) with biochar adsorption yielded a nitrogen recovery rate of 540 t/a, which is 3.8% higher than that obtained via air stripping in S2 (AdBC). Nitrogen was not recovered in S1 (CWWTP).

**Table 1.** Nitrogen recovery rates.

| Scenario | Recovered Nitrogen (t/a) |
|----------|--------------------------|
| S1 (CWWTP) | 0 |
| S2 (S&S) | 520 |
| S3 (AdBC) | 540 |

*2.1. Contribution Analysis*

The environmental impact of each process was further assessed through a contribution analysis. In the case of climate change with biogenic carbon, wood biochar production and biochar spreading cause most of the total emissions in S3 (AdBC), up to 68% and 21%, respectively (Figure 2). The high electricity demand for wood biochar production results in significant emissions. On the other hand, wood pyrolysis provides excess heat that can be used as a substitute for district heat production. However, biochar-based carbon sequestration contributes 96% of the avoided emissions, so the total impact of ammonia and heat substitution is rather limited—only 4%.

Regarding S2 (S&S), the electricity consumption in stripping and scrubbing is one of the main contributors to the climate change impact category with biogenic carbon, accounting for 38% of the total emissions. The consumption of sulfuric acid and sodium hydroxide (NaOH) contributes 15% and 25% of the total emissions, respectively. Determined by input mass flows, the electricity demands of the stripper and the scrubber are critical parameters [10]. The high input flows of reject water and condensate are the main reasons for the high electricity consumption obtained here. All the avoided emissions are due to nitrogen recovery, which enables the substitution of fossil-based fertilizers.

The main impact on freshwater and marine eutrophication in S2 (S&S) is caused by "other emissions", which include, e.g., transport of different chemicals, spreading of nitrogen fertilizer (ammonium sulfate), water consumption for acid dilution, and recycling of residual stripping liquid. Correspondingly, "other emissions" in S3 (AdBC) include transporting and spreading sewage sludge/wood biochar. Compared to S2 (S&S), S1 (CWWTP) performs better for freshwater eutrophication, which is mainly due to the consumption of NaOH and sulfuric acid in stripping and scrubbing, and, to some degree, due to fuel consumption for spreading fertilizers.

With respect to terrestrial acidification, the environmental impact of S2 (S&S) is mainly from the consumption of electricity, heat, and sulfuric acid in the stripping and scrubbing process. In S3 (AdBC), the main impact originates from biochar spreading, but also the pyrolysis of sewage sludge causes a notable impact. Furthermore, the substitution of district heat from wood pyrolysis generates 150 t $CO_2$,eq./FU of avoided emissions.

### 2.2. Sensitivity Analysis

The most important sensitivity ratios for each impact category (climate change including and excluding biogenic carbon, marine eutrophication, freshwater eutrophication, and terrestrial acidification) are shown in Table 2. Regarding S1 (CWWTP), nitrogen removal efficiency is the only parameter showing considerable variation in any impact categories, indicating high sensitivity for marine eutrophication. By increasing the nitrogen removal efficiency, the SR became negative; so, the impact on marine eutrophication will decrease because of the effective removal of nitrogen. Other parameters, such as electricity and heat demand, show only a minor influence on any impact category ($|SR| < 0.7$).

**Table 2.** Sensitivity ratios (SRs) for parameters in each scenario against selected impact categories. ▨ $|SR| > 1$ Particularly important, ▨ $|SR| = 0.8-1$ Important parameter, ▨ $|SR| = 0.2-0.8$ Slightly important, ▢ $|SR| < 0.2$ Minor importance.

| S1 (CWWTP) | CC incl. Biogenic | CC excl. Biogenic | FWE | ME | TA |
|---|---|---|---|---|---|
| Parameter | | | | | |
| Electricity | 0.58 | 0.58 | 0.65 | 0.00 | 0.66 |
| Heat | 0.28 | 0.28 | 0.35 | 0.00 | 0.34 |
| Lime | 0.15 | 0.15 | 0.00 | 0.00 | 0.00 |
| N removal efficiency | 0.00 | 0.00 | 0.00 | −8.49 | 0.00 |
| Electricity biogas | 0.02 | 0.02 | 0.09 | 0.00 | 0.06 |
| Heat biogas | −0.23 | −0.22 | 0.35 | 0.00 | 0.30 |
| **S2 (S&S)** | **CC incl. biogenic** | **CC excl. biogenic** | **FWE** | **ME** | **TA** |
| Parameter | | | | | |
| Electricity use | 0.71 | 0.76 | 0.05 | 0.00 | 0.12 |
| Heat use | 0.29 | 0.21 | 0.04 | 0.00 | 0.54 |
| H$_2$SO$_4$ use | −0.26 | −0.31 | 0.02 | 0.00 | 0.21 |
| NaOH | 0.32 | 0.32 | 0.08 | 0.00 | 0.03 |
| Water use | 0.00 | −0.01 | 0.00 | 0.00 | 0.00 |
| Stripper–Scrubber efficiency | −0.16 | −0.27 | −0.01 | −1.44 | −0.01 |
| Distance of fertilizer spreading | 0.00 | −0.01 | 0.00 | 0.00 | 0.00 |
| **S3 (AdBC)** | **CC incl. biogenic** | **CC excl. biogenic** | **FWE** | **ME** | **TA** |
| Parameter | | | | | |
| Nitrogen adsorption capacity, SS biochar | 0.12 | −0.11 | 0.03 | 0.0001 | −0.07 |
| Electricity demand, SS biochar production | 0.02 | 0.07 | 0.03 | 0.0001 | 0.01 |
| Heat demand, SS biochar production | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Electricity demand, wood biochar production | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Heat demand, wood drying | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| SO$_2$ removal | 0.00 | 0.00 | 0.00 | 0.0000 | 0.29 |
| Carbon share in biochar | −0.31 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Biochar nitrogen usability | 0.00 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Nitrogen adsorption capacity, wood biochar | 0.52 | −0.50 | 0.13 | 0.0002 | −0.31 |
| Electricity demand wood biochar production | 0.04 | 0.12 | 0.06 | 0.0002 | 0.00 |
| Heat demand biochar | 0.05 | 0.10 | 0.13 | 0.0002 | 0.04 |
| Wood processing emissions | 0.28 | 0.84 | 0.00 | 0.0000 | 0.00 |
| Yield of wood biochar | −6.83 | −0.43 | 0.26 | 0.0004 | 0.09 |
| Excess heat production | −0.19 | −0.40 | −0.51 | −0.0009 | 0.15 |
| Substituted district heat emissions | 0.00 | 0.00 | 0.00 | 0.0000 | 0.01 |
| C share remaining in soil | −11.37 | 0.00 | 0.00 | 0.0000 | 0.00 |
| Carbon content of wood biochar | 9.57 | 0.00 | 0.00 | 0.0000 | 0.00 |
| CF (Carbon footprint) | 5.77 | −0.09 | 0.02 | 0.0000 | −0.06 |
| Steam from biomass and natural gas | 4.06 | 12.62 | −0.01 | −0.0002 | −0.04 |

**CC incl. biogenic** = Climate change including biogenic carbon; **CC excl. biogenic** = Climate change excluding biogenic carbon; **FEW** = Fresh water eutrophication; **ME** = Marine water eutrophication; **TE** = Terrestrial acidification

Indicated by the highest and the lowest SR values, the results in S2 (S&S) are most sensitive to two parameters, i.e., electricity use and stripper–scrubber efficiency. The impact on global warming potential increases by increasing the electricity demand, whereas other impact categories remain unaffected. On the other hand, the efficiency of the stripping and scrubbing process has a negative SR in the marine eutrophication impact category, so a

decreasing efficiency increases the eutrophication impact significantly (e.g., a 10% decrease increases the impact by 14%).

Among all variables in S3 (AdBC), the following parameters showed the highest influence on global warming potential: wood biochar yield, carbon share remaining in soil, total carbon in wood, CF (carbon footprint), and steam obtained from biomass and natural gas. Biochar yield with SR < −6 implies considerable variation in results; if the yield is increased, the impact on the climate change categories will decrease (e.g., a 10% increase will decrease the impact by 68%). However, the range of biochar yield is typically broad and it can be controlled by varying the conditions during pyrolysis. Thus, the conditions should be carefully set to produce a high yield of char.

Furthermore, the amount of steam obtained from biomass and natural gas combustion significantly impacts both climate change categories. Therefore, the parameter is considered extremely sensitive, because replacing a small amount of biomass with natural gas as the source of steam generation would result in significant variation in results. On the other hand, the higher the biochar yield from pyrolysis, the higher the need for external energy from biomass or natural gas. Consequently, it is necessary to pursue an optimum yield from pyrolysis. The share of carbon remaining in the soil is in turn the most sensitive parameter for the climate change category with biogenic carbon. If this variable decreases by 10%, the emissions will increase by 110%. Nevertheless, the parameter is considered highly uncertain because the long-term experimental data on carbon persistence in different soil conditions is rather limited.

## 3. Materials and Methods

The potential environmental impacts in each scenario are evaluated through a systematic LCA methodology based on ISO standards 14,040 and 14,044 [26,27]. The study includes the four steps of LCA: goal and scope definition, life cycle inventory, life cycle impact assessment (LCIA), and interpretation of results. The following impact categories were selected based on a literature review [28]: global warming potential, terrestrial acidification, marine water eutrophication, and freshwater eutrophication. Environmental performance modeling was carried out using the GaBi 10.5.1.124 software and employing the ReCiPe 2016 v1.1 (midpoint hierarchist timeframe) technique. ReCiPe indicators, which provide information on the environmental issues related to the inputs and outputs of a product system [26], are commonly utilized due to their reliability [29].

### 3.1. Goal and Scope Definition

The goal of this work is to assess the environmental performance of recovering nitrogen from sewage sludge treatment in a municipal WWTP. Nitrogen is recovered from liquid waste streams generated during mechanical dewatering and thermal drying of anaerobically digested sewage sludge. The digestate from anaerobic digestion is first dewatered and then directed to thermal drying to further reduce the water content before post-treatment via pyrolysis. The resulting water streams are rich in ammonium ($NH_4$-N) and combined for effective nitrogen recovery. Three different scenarios to manage these streams are evaluated and compared in terms of nitrogen recovery rate and potential environmental impacts:

- Scenario S1 (CWWTP) incorporates conventional treatment of reject water and condensate in a municipal WWTP; accordingly, nitrogen is not recovered but mostly removed and released into the atmosphere as $N_2$ through nitrification/denitrification.
- Scenario S2 (S&S) utilizes air stripping in combination with gas scrubbing to recover nitrogen. The two streams with recoverable nitrogen (reject water of mechanical dewatering and condensate from thermal drying) enter a stripper, and air is added in the stripper to convert ammonium to ammonia gas; subsequently, ammonia gas is absorbed in sulfuric acid in a scrubber to produce ammonium sulfate fertilizer.
- Instead of air stripping, Scenario S3 (AdBC) considers nitrogen recovery from reject water and condensate through ammonia adsorption on biochar derived from sewage

sludge and wood pyrolysis. The biochar doped with ammonia is applied to land for soil enhancement and carbon sequestration, substituting fossil-based nitrogen fertilizers.

The functional unit of this study is 870 kt/a (thousand metric tons per year) of reject water and 45 kt/a of condensate. The functional unit is based on a case study on Viikinmäki WWTP of Helsinki Region Environmental Services Authority (HSY), which was studied earlier by Havukainen et al. [21] and Saud et al. [10]. Illustrated in Figure 3, the system boundaries include the treatment of nitrogen-rich water streams (reject water and condensate).
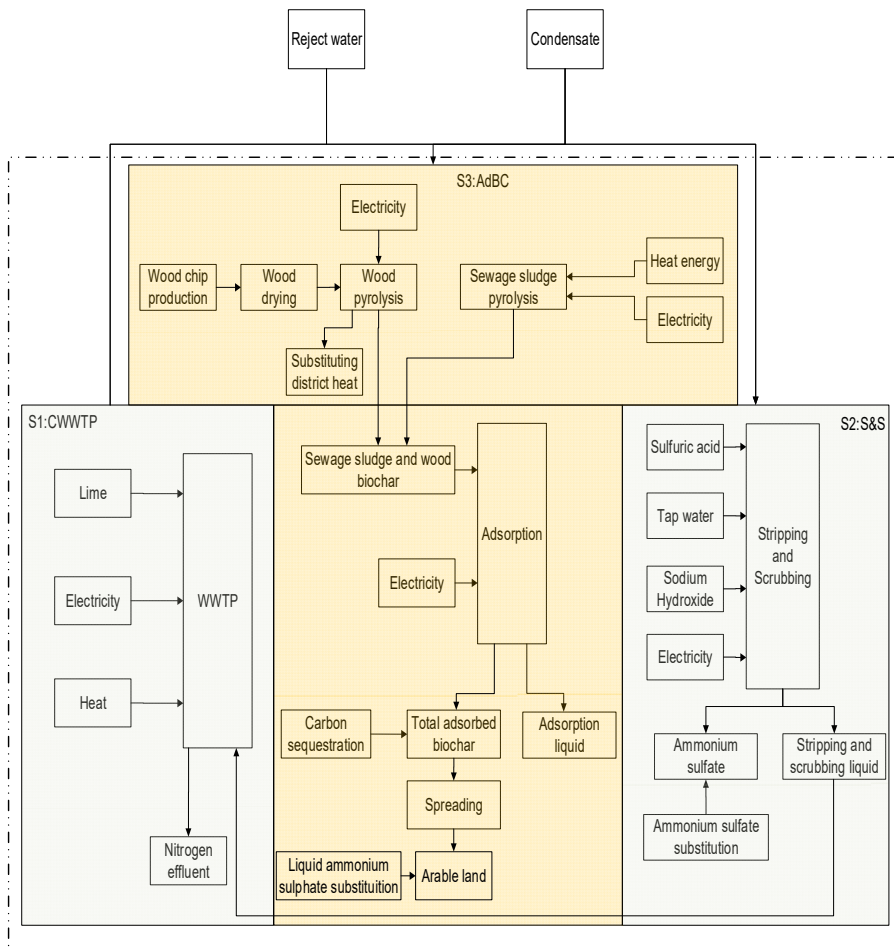


**Figure 3.** System boundaries and processes in each scenario.

*3.2. Life Cycle Inventory*

3.2.1. Reject Water and Condensate

Sewage sludge is directed to anaerobic digestion and the anaerobically digested sewage sludge (digestate) is mechanically dewatered for further processing, and an ammonium-

rich liquid stream (reject water) is formed. In the baseline configuration in Scenario S1 (CWWTP), the reject water is recirculated back to the wastewater treatment process for nitrogen removal via nitrification/denitrification. After dewatering, the digestate is directed to thermal drying to further reduce the water content. Up to 12% of the total nitrogen ($N_{tot}$) in the digestate is released within the drying fumes [30], and after condensation, another ammonium-rich liquid stream (condensate) is formed. By combining these streams, a considerable amount of nitrogen can be recovered and recycled. The properties of the reject water and the condensate are presented in Table 3.

**Table 3.** Reject water and condensate properties.

| Reject Water | | | |
|---|---|---|---|
| **Parameter** | **Value** | **Units** | **References** |
| $N_{tot}$ | 1 | kg/t | [31] |
| $NH_4$ | 0.8 | kg/t | |
| $NH_4$-N | 0.62 | kg/t | |
| **Condensate** | | | |
| **Parameter** | **Value** | **Units** | **References** |
| $N_{tot}$ | 0.09 | kg/t | [32] |
| $NH_4$-N | 0.09 | kg/t | |

### 3.2.2. Wastewater Treatment Plant (WWTP)

The WWTP considered in this work is based on data obtained from Viikinmäki WWTP in Helsinki, Finland, operated by the HSY [30]. The properties and parameters of the plant are given in Table 4.

**Table 4.** WWTP properties.

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Electricity | 1.52 | MJ/t of water | [31] |
| Heat | 1.33 | MJ/t of water | |
| Lime | 0.030 | kg/t of water | |
| N removal efficiency | 85 | % | |

The required heat for the WWTP is produced by biogas and the electricity to the WWTP is supplied partially by biogas (64%). The remainder of the electricity is assumed to be supplied either by the grid mix in Finland or renewable electricity (wind power). Table S4 (Supplementary Material) shows the power grid mix in Finland, including production and importation. A sizable portion of renewable electricity (47%) and electricity with low emissions (34.7%) are produced in Finland, and less than 20% of the whole energy mix comes from fossil fuels. Sweden accounts for most (18%) of the imported electricity, with fewer emissions than the Finnish energy production mix. Only 8% of the imported power comes from Russia, but due to the high emissions intensity of Russian electricity, it is responsible for 25% of the total emissions [33]. Since wind power in Finland has the lower emission factor compared to other renewable energy sources, it has been chosen as an example of renewable energy [34].

### 3.2.3. Stripping and Scrubbing

In S2 (S&S), a stream combined of reject water and condensate is introduced in the stripper to convert dissolved nitrogen (ammonium) to gaseous ammonia. After stripping, the nitrogen-containing air is delivered to the scrubber from the bottom, and liquid acid is either sprayed on top of or through a dense column to take in ammonia and create liquid ammonium sulfate. The inventory data of the stripping and scrubbing process is summarized in Table 5.

**Table 5.** Stripping and scrubbing parameters.

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| Electricity use | 0.028 | MJ/kg | [35] |
| Heat use | 0.188 | MJ/kg | [35,36] |
| $H_2SO_4$ use | 3.5 | kg $H_2SO_4$/kg $NH_4$-N | [37] |
| NaOH | 3.3 | kg NaOH/kg $NH_4$-N | [21] |
| Water use | 2.1 | kg water/kg $NH_4$-N | Calculated |
| Stripper–Scrubber efficiency | 95 | % | [38,39] |
| **Transport** | | | |
| Biochar | 43 | km | [40] |
| Fertilizer | 43 | km | [21] |
| $H_2SO_4$ | 201 | km | [21] |

3.2.4. Adsorption on Biochar

In S3 (AdBC), biochar derived from sewage sludge digestate and wood pyrolysis is used as an adsorbent. The amount of biochar from sewage sludge alone is insufficient; therefore, it is necessary to pyrolyze wood to fulfill the adsorbent requirement. Before pyrolysis, wood must be chopped and dried, and electricity is required in the pyrolysis reactor. Wood drying requires electricity and heat (steam), which is produced using biomass and natural gas. In wood biochar production, excess heat originating from non-condensable pyrolysis gas is utilized for district heating and it is substituting the Finnish average district heating mix [41].

Ammonia adsorption is carried out by introducing the streams of reject water and condensate into an adsorption column filled with biochar. The energy data of sewage sludge biochar are obtained from Refs. [10,21], and the mass flow rates of sewage sludge and biochar are collected from Ref. [10]. Additional parameters, e.g., the biogenic carbon footprint of biochar recovery caused by biochar land application, were obtained from Ref. [41]. Table 6 summarizes the parameters considered in the pyrolysis process.

**Table 6.** Parameters of the adsorption system.

| Parameter | Value | Unit | Reference |
|---|---|---|---|
| **Sewage sludge biochar** | | | |
| Mass of SS biochar | 12,000 | t | [10] |
| Mass of sewage sludge | 65,000 | t | [10] |
| Nitrogen adsorption capacity | 0.004 | kg N-$NH_4$+/kg biochar | [42–44] |
| Electricity demand, SS biochar production | 0.827 | MJ/kg biochar | [21] |
| Heat demand, SS biochar production | 0.003 | MJ/kg biochar | [21] |
| Electricity demand, wood biochar production | 0.750 | MJ/kg biochar | [21] |
| Heat demand, wood drying | 0.003 | MJ/kg biochar | [21] |
| $SO_2$ removal | 0.021 | kg $CO_2$, eq./kg biochar | [21] |
| Carbon share in biochar | 34% | | [41] |
| Biochar nitrogen usability | 64% | | [45] |
| Carbon footprint biogenic | 0.45 | kg $CO_2$, eq./kg $CO_2$ | [41] |
| **Wood biochar** | | | |
| Mass of wood biochar | 97,000 | t | Calculated |
| Mass of wood | 280,000 | t | Calculated |
| Nitrogen adsorption capacity | 0.005 | kg N-$NH_4$+/kg biochar | [46–48] |
| Electricity demand wood biochar production | 0.252 | MJ/kg removed water | [41] |
| Heat demand biochar | 4.504 | MJ/kg removed water | [41] |
| Moisture (wet wood) | 28% | | [41] |
| Moisture (dry wood) | 10% | | [41] |
| Wood processing emissions | 0.018 | kg $CO_2$/kg wood | [49] |
| Yield of wood biochar | 0.34 | kg biochar/kg dry wood | [50] |
| Excess heat production | 4.9 | MJ/kg wood | [41] |
| Carbon content of wood biochar | 34% | | [41] |
| C share remaining in soil | 68% | | [41] |
| CF (carbon footprint) biochar land application | 0.45 | kg $CO_2$,eq./kg $CO_2$ | [51] |
| CF wood pyrolysis gas combustion | 0.45 | kg $CO_2$,eq./kg $CO_2$ | [51] |

### 3.3. Life Cycle Impact Assessment

The results for selected impact categories (global warming potential with and without biogenic carbon, terrestrial acidification, marine water eutrophication, and freshwater eutrophication) are analyzed through a life cycle impact assessment. First, a contribution analysis is performed to show the main contributing processes. The results of each individual process are presented discretely in terms of direct and avoided emissions. In this way, the least and the most important processes can be identified, which helps to better understand the results [52].

Second, a sensitivity analysis is used to show the most sensitive parameters. Ref. [53] suggests using sensitivity ratios (SR). SR is calculated as a ratio of two relative changes by dividing the relative change of the total result by the relative change of the individual parameter. Ref. [54] asserts that parameters with SR values greater than 0.8 are significant, and those with SR values greater than 1 are particularly significant. Parameters with SR values less than 0.2 have just a minor impact on the overall results. The usefulness of SR, however, is reliant on the effect category. Hence the SR results should be assessed inside an impact category rather than being compared between them [55]. Third, the range of the net result was determined using a "high" and "low" performance sensitivity analysis using the parameter values shown in Table S7 (Supplementary Material) for the chosen impact categories.

## 4. Discussion

A 90%/10% mixture of wood/sewage sludge biochar is generated during pyrolysis in S3 (AdBC). When applied to soil for carbon sequestration, one metric ton of this mixture removes 1.41 metric tons of $CO_2$,eq. from the atmosphere. Depending on the biochar source, this value is commonly between 0.8–2.9 t $CO_2$,eq./t [56]. According to Ref. [57] biochar derived from sewage sludge has a carbon sequestration value of 0.8 t $CO_2$,eq./t, while Ref. [58] suggest that biochar from forest residue could sequester carbon in the range of 2–2.6 t $CO_2$,eq./t.

The biochar-based carbon capture in S3 (AdBC) resulted in net negative $CO_2$ emissions. It has been estimated that the amount of carbon sequestered by biochar could increase globally to 0.3–2 Gt $CO_2$ per year by 2050. On the other hand, the yield and properties of pyrolysis products depend on several factors, such as temperature, residence time, pressure, and feedstock composition. Thus, the results obtained for S3 (AdBC) are considered highly sensitive, and further data would be needed to reliably assess the global warming potential of ammonia adsorption on biochar [59]. However, despite different methodologies and different conditions in biochar production, recent studies suggest that biochar could potentially neutralize greenhouse gas emissions while facilitating carbon capture [60].

The electricity consumption and the use of chemicals ($H_2SO_4$ and NaOH) in the stripping and scrubbing process are the main contributors to the environmental impacts in S2 (S&S). In the current work, an electricity consumption of 0.2 MJ/kg $NH_4$-N was assumed, but values as low as 0.01 MJ/kg $NH_4$-N can be found in the literature [35]. Generally, the electricity demand is determined by equipment design, operational conditions, and efficiency [61]. In addition, the environmental performance is greatly affected by the source of electricity generation—when using renewable electricity (wind power), the net emissions will reduce by 95%.

The consumption of chemicals can also be optimized. The selection of acid in the scrubbing process is determined by the requirement of the final product. Here, sulfuric acid ($H_2SO_4$) was used to produce ammonium sulfate. In addition to its use as nitrogen fertilizer in agriculture, ammonium sulfate has a wide range of potential applications. For example, it is used as a wood preservative and as a chemical in flame retardants [62]. However, as a source of key macronutrients N and S, the main target for the ammonium sulfate recovered from waste streams is the substitution of synthetic ammonium sulfate produced by the energy-intensive Haber–Bosch process. Alternatively, sulfuric acid could be replaced by organic acids such as citric acid or acetic acid to recover ammonium [63].

Furthermore, it is possible to reduce the environmental impacts by reutilizing spent sulfuric acid from petroleum refineries [64,65]. By using nitric acid (HNO$_3$), the end product would be ammonium nitrate, the most widely used nitrogen fertilizer after urea. Ammonium nitrate has been used in mining, construction, and yeast production industries. Moreover, it can be utilized as a component in insecticides or as an adsorbent for nitrogen oxide [66].

## 5. Conclusions

This study aimed to evaluate the environmental performance of nitrogen recovery for fertilizer purposes from sewage sludge treatment. Three different scenarios, one without and two with nitrogen recovery, were investigated and compared in terms of nitrogen recovery rate and potential environmental impacts. Utilizing either air stripping/scrubbing or pyrolysis-derived biochar adsorbent, nitrogen was recovered from ammonium-rich side streams generated during mechanical dewatering (reject water) and thermal drying (condensate) of anaerobically digested sewage sludge. The results show that targeting both these streams for nitrogen recovery would improve the total recovery rate and allow efficient utilization of the nitrogen sources available in WWTPs.

Scenario S3 (nitrogen recovery via ammonia adsorption) performed better in three of the five impact categories considered in this work, including climate change with biogenic carbon, freshwater eutrophication, and marine water eutrophication. The baseline scenario S1 (conventional treatment without nitrogen recovery) showed the lowest net impact for two categories, namely, climate change without biogenic carbon and acidification. In terms of environmental impacts, S2 (nitrogen recovery via air stripping and subsequent scrubbing) remained between these two scenarios. Overall, the main climate impact was caused by biochar production and utilization for carbon capture. Ammonia capture and substitution of nitrogen fertilizers appeared to cause only minor effects on climate change.

The stripping and scrubbing process for nitrogen recovery could be further improved. Specifically, the production of electricity and chemicals (NaOH, sulfuric acid) caused significant emissions. Potential improvements could be obtained by using renewable sources for electricity or replacing some of the chemicals. Furthermore, alternative process designs, e.g., steam stripping and subsequent condensation instead of air stripping and scrubbing, could provide additional benefits and reduce the net environmental impact.

The use of pyrolysis-derived biochar in ammonia recovery and further utilization for soil improvement and carbon sequestration appeared highly beneficial. While carbon sequestration is not directly related to nutrient recycling, it is shown to be relevant for sludge-based nutrient recycling pathways. However, massive quantities of biochar would be required due to the possibly low adsorption capacity, affecting the total cost of nitrogen recovery.

Solutions that simultaneously address global concerns as well as local human and ecological health are increasingly needed. This study contributes an analysis promoting the multifunctional nature of wastewater systems with integrated resource recovery for potential environmental, economic, and health benefits. The results can be used, e.g., by WWTP utilities in planning for approaches to climate change mitigation.

**Author Contributions:** Conceptualization, A.S., J.H. and M.H.; methodology, A.S.; validation, A.S. and J.H.; data curation, A.S. and J.H.; writing—original draft preparation, A.S.; writing—review and editing, A.S., J.H., P.P. and M.H.; visualization, A.S.; supervision, J.H. and M.H.; project administration, M.H. All authors have read and agreed to the published version of the manuscript.

**Data Availability Statement:** The data presented in this study are available in supplementary material.

**Conflicts of Interest:** The authors declare no conflict of interest.

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