Department of Built Environment

Waste nutrients harvested

Design and evaluation of nitrogen and phosphorus recovery processes utilizing membrane contactor and adsorption techniques

Juho Uzkurt Kaljunen

Aalto University

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Abstract

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Abstract

Global food production relies on industrial fertilizers. The explosive growth of the human population would not have been possible without processes that bind nitrogen from the atmosphere to ammonia and mined phosphate rock to easily soluble products. However, the linear consumption pattern (as opposed to circular) has backfired on us. As a result, the nutrient systems we rely on consume significant amounts of energy and create an array of environmental problems. This dissertation focuses on the question: 'How can we close the nutrient loop?' Furthermore, special attention is paid to managing nutrient recycling in an environmentally and economically sustainable way. There is a vast quantity of nutrients available in different concentrated waste streams. However, these nutrients are not recycled efficiently because it is economically unfeasible or there are concerns about pollutants. These concerns apply to both liquid and solid waste streams.

This dissertation is built around NPHarvest, a nutrient recovery technology developed at Aalto University. NPHarvest recovers both nitrogen and phosphorus from liquid waste streams with high efficiency. The end products are clean ammonia salt and solid material that contains phosphorus, calcium and carbon. The dissertation found that NPHarvest as a technology is economically competitive. The recovery process's novelty is how well it is designed to tolerate suspended solids in a wastewater environment and operate with low energy consumption, which decreases operational costs. Upon studying the life cycle assessment of the process, NPHarvest environmental performance was found to be positive or neutral in most impact categories. The process has the potential to be climate-positive (carbon-negative) with further optimization. Additionally, this dissertation examines P recovery from chemically precipitated sludge. It is technically possible, but not feasible enough, to be implemented on a large scale with currently available technologies. Waste-based biosolid materials proved to be a suitable pathway to capture phosphorus after acidleaching. The phosphorus-loaded biosolid can function as organic fertilizers.

Finally, the dissertation reflects on the implications of systemwide nutrient recovery. Nutrient recovery is a shift towards transforming treatment plants to resource recovery plants, in addition to enabling the facilities to reach better cost-effectiveness. Furthermore, nutrient recovery enables decentralized treatment systems should they be desired. To finish the discussion, the true meaning of sustainable technology is discussed.

Keywords Wastewater treatment, nutrient recovery, circular economy, NPHarvest

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Tiivistelmä

Maailmaanlaajuinen ruoantuotanto nojaa teollisiin lannoitteisiin. Ihmiskunnan räjähdysmäinen kasvu ei olisi ollut mahdollista ilman prosesseja, jotka sitovat typpeä ilmakehästä ammoniakiksi ja maaperästä louhittua fosforikiveä helposti kasveille saatavaan muotoon. Mutta ihmisten lineaariset (kiertävän vastakohta) kulutustottumukset ovat kostautuneet meille. Tuloksena ravinnejärjestelmät, joihin turvaudumme, käyttävät merkittävän määrän energiaa ja luovat joukon ympäristöongelmia. Tämä väitöskirja pyrkii vastaamaan kysymykseen "kuinka voimme sulkea ravinnekierron?". Erillistä huomiota on kiinnitetty siihen, kuinka ravinteiden kiertoa voi hallita kestävästi sekä ympäristön että talouden kannalta. Erilaisissa jätevirroissa on huomattava määrä ravinteita, joita on mahdollista hyödytää. Näitä ravinteita ei kuitenkaan hyödynnetä tehokkaasti, koska se ei ole taloudellisesti kannattavaa tai kiertolannoitteiden (sekä nesteiden että kiinteiden jakeiden) haitta-ainepitoisuudet ovat liian korkeat.

Tämä väitöskirja on tehty NPHarvestin ympärille. NPHarvest on ravinteiden talteenottoteknologia, joka on kehitetty Aalto-yliopistossa. NPHarvest ottaa talteen sekä typen että fosforin nestemäisistä jätevirroista. Lopputuotteet ovat puhdas ammoniumsuola sekä kiinteä materiaali, jossa on fosforia, kalkkia ja hiiltä. Tuloksien pohjalta on selvää, että NPHarvest on taloudellisesti kilpailukykyinen teknologia. Talteenottoprosessin uutuus on siinä, että se on suunniteltu sietämään jätevesien korkeaa kiintoainepitoisuutta kuluttamatta suurta määrää energiaa. Tämä pitää prosessin operaatiokulut matalina. NPHarvestin elinkaarianalyysin mukaan prosessi on ympäristön tilaa parantava tai neutraali useimmissa vaikutuskategorioissa. Ilmastonmuutoksen suhteen NPHarvestilla on potentiaali olla positiivinen (hiilinegatiivinen). Lisäksi tämä väitöskirja tarkastelee fosforin talteenottoa kemiallisesti saostetusta lietteestä. Talteenotto siitä materiaalista on tekniseltä kannalta mahdollista, mutta tällä hetkellä se ei ole tarpeeksi kannattavaa suuren mittakaavan sovellusta varten. Jätepohjaiset biomateriaalit osoittautuivat sopivaksi keinoksi ottaa fosfori talteen sen jälkneen, kun se oli liuotettu hapolla irti lietteestä. Fosforilla ladattu biomateriaali voi toimia orgaanisena lannoitteena.

Lopuksi tämä väitöskirja pohdiskelee ravinteiden talteenoton vaikutuksia laajempaan kokonaisuuteen. Ravinteiden talteenotto edustaa siirtymää kohti resurssien talteenottolaitoksia, sen lisäksi että se mahdollistaa näiden laitosten toiminnan kustannustehokkaammin. Lisäksi ravinteiden talteenotto mahdollistaa hajautetut käsittelyratkaisut, mikäli siihen suuntaan on tarpeellista mennä. Pohdiskelun lopuksi mietin, mitä tarkoittaa kestävä teknologia.

Avainsanat Jäteveden käsittely, ravinteiden talteenotto, kiertotalous, NPHarvest

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This dissertation is the culmination of the first steps of my professional career. I am the creation of many sources of influence, and I will list them in chronological order.

The first and largest impact comes from my parents Timo and Laura for the support in pursuing whatever I wanted. They had a philosophy where I (along with my brother) was provided with tools rather than objectives. These tools have enabled me to develop to any direction desired and for that I'm thankful.

To complement the acquisition of these tools, The Helsinki High School of Mathematics in Maunula (MAYK) was very useful. They set me on a path of higher education, of course, but not by their curriculum alone. The true strength of that school, at least for me and the class of 08, is that they group up like-minded students together. Spending 2-3 years with that class made me see education, learning and argumentative interaction as an enjoyable endeavour. These traits have served me well during this doctoral journey.

After high school I started studying Bioproduct Technology at Aalto School of Chemical Engineering. Towards the end of my Batchelor studies the School of Chemical Engineering decided to pull the rug under my feet and renew the Master's program without Bioproduct Technology in it. Instead, they allowed me to completely customize my Master's studies. Everything was acceptable as long as the package of courses made sense. This resulted in me finding water engineering in the School of Engineering.

Water engineering led me to discover NPHarvest and I started working with my supervisor Anna Mikola. She has guided me through my Master's thesis as well as this dissertation and I want to thank her for her phenomenal job in mentoring me while allowing a high degree of freedom in research while battling for administrative survival and funding on my behalf.

During my dissertation work Anna hired Raed Al-Juboori. He became integral part of NPHarvest (and the Water Lab in Aalto). He brought great analytical knowledge that enhanced the quality of my publications significantly. In addition, we have enjoyed many uninhibited philosophical and academic discussions that I so thoroughly enjoyed with my high school class of 08.

During the time I was working on my dissertation, I met my wife Nesli in a summer school in Palermo, Italy. She has given her constant (and persistent) support during the last three years and without her the dissertation might have taken a bit longer.

The working environment has also been important. The people of Vesitalo (Tietotie 1E) have been crucial in creating the most wonderful workplace where I, once again, have felt like anything is possible. A special mention to Antti Louhio for his genius ideas and Aino Peltola for having patience with my (messy) lab adventures.

This work has been supported by several funding organizations. Maa- ja vesitekniikan tuki (MVTT) has supported me and research for Paper III in this dissertation. Finnish Ministry of Environment has supported NPHarvest through several stages of RAKI funding. BusinessFinland stepped in during the later phases of NPHarvest to fund the commercialisation phase of the research. Finally, the School of Engineering has also funded my graduation work.

For the final steps of the I want to thank the pre-examiners Dr. Maite Pijuan and Dr. Mari Winkler for the quick evaluation and very positive comments on my work.

Dr. Maite Pijuan also was my opponent. I deeply thank you for being a part of the process, without you I would not have been able to graduate.

Espoo, March 2023
Juho Uzkurt Kaljunen

Contents

List of Abbreviations and Symbols

$(NH_4)_2SO_4$	Ammonium sulphate
ADPe	Abiotic depletion of elements
AP	Acidification potential
CPR	Chemical phosphorus removal
Cr	Chromium
Cu	Copper
EBPR	Enhanced biological phosphorus removal
EP	Eutrophication potential
ETP	Environmental toxicity potential
EU	European Union
GWP	Global warming potential
HTPc	Human toxicity potential – cancer
HTPnc	Human toxicity potential – non-cancer
K	Diffusion coefficient
LCA	Life cycle assessment
N	Nitrogen
N_2	Nitrogen gas molecule
N_2O	Nitrous oxide
P	Phosphorus
PAH	Polycyclic aromatic hydrocarbons
Pb	Lead
PCB	Check this
PED	Primary energy demand
PFAS	Check this
PO_4^{3-}	Phosphate ion
SS	Suspended solids
WWTP	Wastewater treatment plant

List of Publications

This doctoral dissertation consists of a summary and the following publications, which are referred to in the text by their numerals.

I. Uzkurt Kaljunen, J., Al-Juboori, R.A., Mikola, A., Righetto, I., Konola, I., 2021. Newly developed membrane contactor-based N and P recovery process: Pilot-scale field experiments and cost analysis. Journal of Cleaner Production 281, 125288. https://doi.org/10.1016/j.jclepro.2020.125288

II. Al-Juboori, R.A., Uzkurt Kaljunen, J., Righetto, I., Mikola, A., 2022. Membrane contactor onsite piloting for nutrient recovery from mesophilic digester reject water: The effect of process conditions and pre-treatment options. Separation and Purification Technology 303, 122250. https://doi.org/10.1016/j.seppur.2022.122250

III. Uzkurt Kaljunen, J., Al-Juboori, R.A., Mikola, A., Khunjar, W., Wells, G., 2023. Phosphorus recovery alternatives for sludge from chemical phosphorus removal processes – Technology comparison and system limitations. Sustainable Materials and Technologies 34 (2022) e00514. https://doi.org/10.1016/j.susmat.2022.e00514

IV. Uzkurt Kaljunen, J., Yazdani, R., Al-Juboori, R.A., Zborowski, C., Meinander, K., Mikola, A., 2022. Adsorptive behavior of phosphorus onto recycled waste biosolids after being acid leached from wastewater sludge. Chemical Engineering Journal Advances 11, 100329. https://doi.org/10.1016/j.ceja.2022.100329

V. Högstrand, S., Uzkurt Kaljunen, J., Al-Juboori, R.A., Jönsson, K., Kjerstadius, H., Mikola, A., Peters, G., Svanström, M., 2023. Life cycle assessment of nutrient recovery from reject water using a novel pilot-scale membrane-based technology. Submitted to Journal of Cleaner Production on October 14, 2022

Author’s Contribution

I. Newly developed membrane contactor-based N and P recovery process: Pilot-scale field experiments and cost analysis

This study was conducted with the NPHarvest field scale pilot. I had a major role in designing the pilot equipment with external collaborators. I also designed the field experiments (including sample collection and analysis) for this study, and I was the primary operator. Al-Juboori and Righetto also operated the pilot. Al-Juboori and I performed the membrane characterization analysis and pilot performance (recovery efficiency) calculations. I performed the cost analysis and comparison for the process while Al-Juboori estimated the investment costs. I also contributed to reviewing and editing the manuscript, while Al-Juboori prepared the initial manuscript draft.

II. Membrane contactor onsite piloting for nutrient recovery from mesophilic digester reject water: The effect of process conditions and pre-treatment options

This paper was also conducted with the NPHarvest field pilot, with upgraded equipment compared to the tests in Paper I. I was directly involved in designing the new equipment. I planned the field tests with the aims of this paper. I set up and operated the pilot with Al-Juboori and Righetto. The data processing that led to drawing the conclusions was mainly done by Al-Juboori, but Righetto and I were also contributing.

III. Phosphorus recovery alternatives for sludge from chemical phosphorus removal processes – Technology comparison and system limitations

This paper was the first paper I started writing. All the co-authors (Al-Juboori, Mikola, Wells and Khunjar) and I created the concept of the paper together. I collected the literature data, developed the methodology, performed the modelling and calculations to draw the conclusions and wrote and edited the paper based on co-author and reviewer comments. I dare say that I did almost everything.

IV. Adsorptive behaviour of phosphorus onto recycled waste biosolids after being acid leached from wastewater sludge

This paper is built on the results of Paper III. I created the concept of this paper with Yazdani and then designed the experiments together with Yazdani and Al-Juboori. Al-Juboori and I created the methodology and performed the experiments together. I handled the data processing and created the kinetic, temperature, diffusion and isotherm model fits for the adsorption process. I collected the literary data and wrote and edited the manuscript. Yazdani, Al-Juboori and Mikola provided comments and edits to the manuscript.

V. Life cycle assessment of nutrient recovery from reject water using a novel pilot-scale membrane-based technology

The work behind this paper was divided into the NPHarvest process operation and LCA modelling. I was responsible for the NPHarvest process operation (like in papers I and II) and data production for the LCA model. I also curated the data for the LCA model. Högstrand created and operated the LCA model and wrote the initial manuscript. I took over the manuscript review, editing and correspondence after Högstrand began her maternity leave. Al-Juboori and Mikola contributed to piloting operation. Peters and Svanström guided the LCA modelling. Jönssön and Kjerstadius administrated the project.

1. Introduction and knowledge gaps

This dissertation is a product of several research projects at Aalto University aimed at developing sustainable recovery processes suitable for Finnish conditions. These include:

- NPHarvest technological development, with the goal to upscale and commercialize nitrogen (N) and phosphorus (P) recovery technology suitable for liquid waste streams
- P recovery from chemically precipitated sludge, with the goal to identify and/or create suitable P recovery technology for local conditions
- NPHarvest life cycle assessment (LCA), with the goal to understand the environmental impacts of the nutrient recovery processes on a system-wide scale and identify hotspots for further process development

The work could be described in two layers. The reasons to develop a technology form the conceptual layer. This includes topics such as why nutrient recovery should be applied and what kinds of benefits society and nature can reap by applying this technology. The second layer is technical, which focuses on how the technology functions and how it performs in terms of economic feasibility and environmental impact.

The nutrients N and P are both a problem and a necessity for us (Robertson and Groffman, 2007). We would not live without these elements (Sengupta et al., 2015), but we have also changed the natural ecosystems greatly with them (Steffen et al., 2015) by discharging the excess. This work explores the necessary steps to reach a situation where the growing human population can co-exist in the natural world without inflicting permanent change. I do not argue for the survival of the natural environment. I doubt that humanity can extinguish life itself even if we tried. No, the grand objective is to ensure that we do not snuff ourselves out by making our very favourable living environment less favourable.

There are three key players: general public/consumers, wastewater treatment utilities and fertilizer production. The general public is the link between utilities and fertilizer producers as people consume food that is grown with fertilizers and produce the wastewater treated at treatment plants. Farmers should be included in the group of key players, but unfortunately, farmers and farming practices fall outside the scope of this work. Cooperation between all players is necessary to reach a sustainable state.

1.1 Global changes in nutrient cycles

Both nutrients, N and P, have their natural cycles through Earth's system (Bouwman et al., 2013). Nitrogen's great and inert storage is the atmosphere, from which some bacteria and plants as well as lightning strikes transform N2 into its reactive forms, such as ammonia (NH3) (Galloway et al., 2004). The reactive forms of ammonia propagate through organic environments and are returned to the atmosphere by a biological denitrification process (Galloway et al., 2003). P, on the other hand, is an element that is naturally in the crust of the Earth (Tiessen, 2008). It is leached off by erosion and other natural phenomena, cycles through the organics and is eventually deposited to the bottom of the water bodies (Smil, 2000).

In the pre-industrial era, flows of these elements were minor and slow (Galloway et al., 2004; Smil, 2000). When the Haber-Bosch process and P-rock mining were widely applied to produce fertilizers, the magnitude of these flows increased significantly (Galloway et al., 2004; Smil, 2000). Granted, this industrialized fertilizer production enabled the feeding of the growing population, but the amount of N and P in natural ecosystems increased manyfold, which has resulted in environmental problems such as eutrophication (Murray et al., 2019). The impact of the change in nutrient flows is visible in the physiochemical flows of planetary boundaries, where the threshold of safe operating space has been already exceeded (O'Neill et al., 2018).

Wastewater treatment plants have had – and still have – a crucial role in controlling the nutrient flows to water bodies. Globally, most wastewaters still go untreated (Sato et al., 2013), but also non-point sources such as agricultural areas produce a nutrient burden on natural areas (Wu and Chen, 2013). We need to improve the nutrient cycles to maintain the current population. However, we also need to maintain the health and stability of the environment that we live in. For this, the nutrient loops need to be closed and discharges controlled more efficiently.

1.2 Wastewater treatment process and environmental issues

Currently, the primary purpose of wastewater treatment is to prevent pollutants and unwanted substances from entering natural aquatic environments (Salgot and Folch, 2018). Nitrogen removal is most commonly achieved with biological activities (labelled the 'activated sludge process') that transform reactive N to N_2 gas (Dold et al., 1981). Phosphorus is removed either by chemical phosphorus removal (CPR) or enhanced biological phosphorus removal (EBPR), or both combined (Tomei et al., 2020). Chemical phosphorus removal requires precipitation chemicals, which are typically iron or aluminium salts (Hauduc et al., 2015). In addition, lime can be used to adjust alkalinity (Teichgräber, 1990). Most plants in Finland utilize CPR; this work focuses primarily on this treatment.

A small fraction of nitrogen is also converted to nitrous oxide in the biological nitrogen removal process, which is also emitted into the atmosphere, significantly contributing to global warming (Vasilaki et al., 2019). Furthermore, the aeration needed to maintain the activated sludge process consumes a considerable amount of energy (Mamais et al., 2014). In some cases, the process also requires an external carbon source if there is not enough carbon in the wastewater, and it is common to use methanol (Cherchi et al., 2009). Methanol is also problematic from an environmental point of view since methanol production has a high carbon footprint (Rumayor et al., 2019).

Wastewater treatment plants often treat concentrated waste streams, such as waste flows from biogas production or other industries. Similarly, if the treatment plant has a digester that produces reject water, that stream is returned to the mainstream process. These concentrated flows contribute significantly to the load even though they are volumetrically small (Koskue et al., 2021; van Loosdrecht and Salem, 2006).

No prior studies have focused on understanding the effects of nutrient recovery on the overall environmental impact of the wastewater treatment process. This research gap is investigated in Paper V. As already mentioned, when a side-stream nutrient recovery process is treating reject water or a concentrated industrial stream, the nutrient load on the mainstream process decreases (Paper V). This is beneficial for the process, not because this would affect the plant's effluent quality (since that is dictated by the environmental permit), but because the resources needed to reach the effluent limits would decrease, making the plant operation more economically viable (Paper V).

1.3 Fertilizer production

In the spring of 2022, the political landscape of Europe changed drastically when Russia attacked Ukraine. Prior to this conflict, the global price of N and P fertilizers had already been increasing (Baffes and Koh, 2022), but the consequences of the conflict made these resources even more scarce. This is a good example of how fertilizer production and distribution are problematic.

The availability of both N and P fertilizers have different issues. P is a limited resource controlled by only a few nations (Walan et al., 2014). The EU is especially vulnerable since most of the P used in agriculture is imported (El Wali et al., 2019). This vulnerability culminates in food price volatility, as is the case in the EU in 2022 (Carbonaro, 2022). Nitrogen, on the other hand, is plentiful in the atmosphere. It is bound to reactive forms through the Haber-Bosch process, which consumes a significant amount of global energy (Smith et al., 2020). The top four countries, China, Russia, the United States and India, produced 55% of ammonia in 2021 (Statista, 2022). The energy supply of these countries consists of 76–89% of coal, natural gas and oil in 2019 or 2020 (IEA, 2022). This leads to significant carbon emissions from N fertilizer production.

A recent idea is green ammonia (Salmon and Bañares-Alcántara, 2021), in which ammonia is produced from the atmosphere's N2 using renewable resources. This approach would solve the problems related to carbon balance. However, this method cycles N through the atmosphere, which still consumes a great deal of energy. Recycling N directly from waste to use circumvents this issue. Yet, on the practical level, the sheer volume of ammonia production is so large that replacing fossil-based Haber-Bosch production is likely to require all available tools, including both recycled and green ammonia.

1.4 Carbon cycle in agriculture and organic fertilizers

The use of nutrient fertilizers has provided increased crop yield but at the same time has contributed to a decrease in carbon content in agricultural soils (Grønlund et al., 2008; Vasbieva, 2019). Carbon is beneficial in soil as it affects the efficiency of fertilizers and decreases their run-off (Lehmann, 2007). In addition, carbon in soil acts as carbon storage, and losing this storage contributes to climate change (Marris, 2006). Organic fertilizers are an option to improve soil health while providing nutrients to the crops. There have been many laboratory-scale studies on creating fertilizers with a carbon structure, which is useful for P adsorption processes (inter alia Khan et al., 2008; Yao et al., 2013). Biochar is not the only porous material that functions as a P adsorbent. In Paper IV, we tested novel waste-based materials that could provide low-cost organic P fertilizer. (Organic means that it contains carbon in its structure.) (Paper IV).

1.5 Nitrogen and phosphorus pathways in the current system

Currently, in Finland, nutrients are recycled only to a limited degree. Digested wastewater treatment sludge is typically composted or otherwise stabilized before use in agriculture or as a soil amendment (VVY, 2019). However, the use of recycled products originating in wastewater is questionable due to pollutant concerns (Gherghel et al., 2019; Seleiman et al., 2020) and is a much-debated topic (Bengtsson and Tillman, 2004). Furthermore, if the sludge originates from a CPR process, P is bound in metal complexes and considered poorly available (Ylivainio et al., 2021). Regardless, the supposed unavailability of phosphorus as well as concerns about pollutants and hygiene have started a discussion on how to extract phosphorus from sludge and use it as fertilizer.

To bypass the pollutant concerns, the negative image and the P availability issue, several technologies have been developed to recover nutrients from waste flows as cleaner products and separate compounds. Several reviews have been written about the recovery technologies for wastewater nutrients (Egle et al., 2016; Van der Hoek et al., 2018; Ye et al., 2020). To transform bound P into soluble phosphate (PO_4^{3-}), wet chemical leaching is a known method (Meyer et al., 2018) that was studied further in papers III and IV.

Besides the solid sludge, there are liquid waste streams that carry nutrients. The most common recovery technologies are ammonia stripping and scrubbing, which captures N in an ammonium salt (Kinidi et al., 2018), and struvite precipitation for P (Mavhungu et al., 2021). A brief overview on the different recovery technologies can be found from Kaljunen (2018). Munasinghe-Arachchige and Nirmalakhandan (2020) published a review comparing different N recovery technologies from liquid waste stream and concluded that gas permeable membranes are the best option, followed by struvite precipitation. Other reviews have arrived to the same conclusion (Al-Juboori et al., 2023; Beckinghausen et al., 2020), using their energy efficiency as an argument. Our process, NPHarvest, relies on this the same phenomena, in addition to the tolerance for suspended solids. It was designed to offer a more profitable option than the above-mentioned ammonia stripping & scrubbing and struvite precipitation that exist in the market.

1.6 NPHarvest

My journey for a doctorate started with NPHarvest, a nutrient recovery process that took a new twist to N stripping with membranes and P precipitation with lime. The goal was to develop an economically feasible process with low energy consumption and easy use of standard, readily available chemicals. The technical journey of the process is presented in Figure 1. The first trial of NPHarvest used hydrophobic gas-permeable ePTFE membranes to strip ammonia from source-separated urine (Pradhan et al., 2017). However, we wanted to explore more abundant waste streams that are more relevant to a wastewater treatment environment, such as reject water from digesters. The best option would, of course, be to recover ammonia directly from sewage, a.k.a. mainstream. However, current recovery technologies are not suitable for the low N concentrations in the mainstream (Cruz et al., 2019).

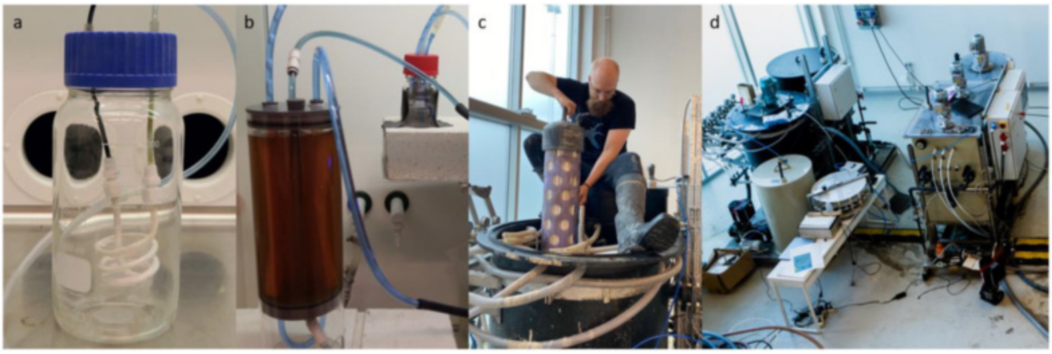

Figure 1. NPHarvest's journey: (a) first batch tests in 2016 (Pradhan et al., 2017), (b) continuous process tests in 2017 (Kaljunen, 2018), (c) field-scale membrane contactor in 2019 (Righetto et al., 2022; Paper I) and (d) full field-scale equipment including pre-treatment in 2020 (papers II and V). The process was scaled up because we realized that a lab-scale environment does not properly represent field conditions, and larger equipment was needed for field trials.

Reject water typically has a high suspended solids (SS) concentration. Membrane contactors on the market are not suitable for wastewater with high SS content since they require microfiltration (10 μm), according to their own manual (3M, 2022). Based on our own estimations and discussion with other researchers, intensive pre-treatment would increase operational costs to an unprofitable level. We believe that the nutrient recovery process must be profitable for it to be an attractive option for industrial applications. In most of the world, there are no legislative requirements for nutrient recovery. Thus, for water utilities to implement nutrient recovery, economic profitability is a necessity, according to our experience. We designed our own membrane contactor so that it tolerates high SS concentrations without harming the membranes or clogging, as shown in Paper I. This allows a greater variety of wastewater to be treated by NPHarvest while maintaining low operating costs (Paper I).

Still, some wastewater streams have such a high SS concentration that the designed NPHarvest tolerance of approximately 300–500 mg-SS/l is not sufficient, and the membrane stripping of ammonia is disturbed (Paper I). In addition, there can also be a considerable amount of phosphorus in waste streams (Paper I). We developed a lime-based ballasted sedimentation process to complement the membrane stripping (Paper I). An up-to-date process schematic can be found in Paper II.

NPHarvest was designed to solve the problems that have been presented in earlier sections. Recovering nutrients from concentrated waste flows allows the mainstream process (and the entire system, including the nutrient recovery technology) better operation from both the economic and environmental points of view (papers I and V) by decreasing existing operational costs, unwanted emissions and producing high-quality recycled nutrient products.

NPHarvest produces two products (Paper I). Ammonia is captured in an inorganic acid, forming an ammonia salt, while P is captured by lime precipitation into a product with P, Ca and C (Paper I). Ammonia salt is a liquid product that can be concentrated or crystallized if desired (Paper V). The membrane that separates ammonia is highly selective (papers I and II). This results in practically pollutant-free ammonia salt, which was confirmed in (Konola, 2019) and our own later analysis results, which are yet to be published. P-product is hygienized by the high pH of the process, but any pollutants that may be present in the source waste stream may escape in the P-product (Konola, 2019).

We evaluated the environmental performance of NPHarvest using a life cycle assessment (LCA) and an established N and P recovery process as a benchmark (Paper V). An LCA is a useful tool to understand larger systems and the impacts of different choices or alternatives that resonate through the system (Finnveden et al., 2009). This was an important step because environmental sustainability is paramount for a process that aims to improve the world's state of sustainability. The motivation for conducting an LCA early in the research lies in the possibility to integrate the results into the process's development.

1.7 Research questions and study-specific focus

Research questions:

1. How to design a membrane stripping process for ammonia so that it is suitable for wastewater? (Paper I)
2. What are the key parameters that affect ammonia recovery with membrane stripping and the optimization of that process? (Paper II)
3. Which technology is most suitable for P recovery from CPR sludge, and how feasible are they? (Paper III)
4. Which waste or natural material (containing carbon) is the most suitable to bind acid-leached P onto organic fertilizers? (Paper IV)
5. What are the environmental impacts of the NPHarvest process, and what needs to be considered in further process development? (Paper V)

The research is focused – on the practical level – on the wastewater treatment field, while the implications of the research are discussed in a greater context. As the overall purpose of my research is to figure out how nutrients from waste could be reused efficiently and safely, the basis for the research was to work with wastewater streams that are suitable for nutrient recovery. This means nutrient-rich waste streams and not mainstream wastewater. Most of the tests and papers are about reject water, but urine and landfill leachate were also investigated.

Figure 2 shows an illustrated map of where investigated recovery techniques fit within wastewater treatment and sludge post-treatment processes, and how the obtained knowledge is distributed accordingly in the produced publications. The blue boxes are the existing infrastructure in the current wastewater treatment system, and the green boxes are research items presented and discussed in my publications. The wastewater system, without NPHarvest or other research items in Figure 2, works by taking in wastewater from a sewage network (upper right corner) and treating it in the treatment process. The treatment process releases a fraction of N into the atmosphere while transferring N and P into sludge (both primary sludge and waste-activated sludge). The sludge is dewatered, usually with a centrifuge. The separated liquid fraction returns to the treatment process while the solid fraction (sludge) is typically stabilized and used. This is not visible in Figure 2. The pyrolysis box represents an ongoing pilot project in the Helsinki area where municipal wastewater treatment plant (WWTP) sludge is pyrolyzed to improve the end-use possibilities of the sludge. We used this pyrolyzed sludge in one of our studies.

Figure 2 also shows how the system could be altered when nutrient recovery technologies are implemented. Papers I and II focus on nutrient recovery from reject water. Paper I is a broad overview of NPHarvest technology functioning on the field scale, including performance, fertilizer end products and cost estimations. The paper answers the very simplified question, ‘Does nutrient recovery like this make sense?’ It is noteworthy that NPHarvest’s applicability is not limited to municipal reject water. Industrial waste streams rich with N and P can also be treated before they reach the municipal treatment process. Paper II takes a deeper dive into the N recovery part of NPHarvest, evaluating different operational parameters and how they affect N recovery performance. This paper has a complementary contribution to the dissertation since Paper I makes the argument for Research Question 1 and builds the foundation for Paper V. Nevertheless, it is included in the thesis to provide further details and form part of the foundation for discussing further product development.

The research gap that NPHarvest fills is specifically about membrane stripping and tolerance to suspended solids. The NPHarvest membrane contactor was designed and built in such a way that membranes are loosely placed inside a container and there is no pressure gradient through the membrane. This means that the suspended solids present in the wastewater being treated do not enter the pores of the hydrophobic membrane; they float by. Ammonia is driven through the membrane by the concentration difference, which does not require pressure manipulation.

Reject water (and some industrial wastewater flows) are rich in N but might be poor in P. Most of the P in municipal (chemical) treatment systems is bound to solid sludge. The P in the sludge can be recovered. Paper III evaluates different technologies to extract P from the chemical P sludge and transform it into a safe fertilizer product. We wanted to find a suitable process for Finnish/Nordic conditions, as stated in Research Question 3. The technologies were selected to represent both chemical and thermal processes and were evaluated based on their operational data (i.e., cost, recovery efficiency and product quality). The Paper III box in Figure 2 has only one output: Paper IV.

We picked one of the five processes evaluated in Paper III for Paper IV and modified it to fit local conditions to answer Research Question 4. We chose to leach P off sludge and then bind it to different biosolids. The existing recovery method produced struvite (Meyer et al., 2018). However, we felt that the material was not the best possible end product. We wanted to develop a fertilizer product that contains carbon in its structure. This meant that instead of struvite precipitation, we took natural or waste-originating materials, such as biochar, lignin, sludge char (pyrolyzed sludge) and humus, and explored the possibility of phosphorus adsorption onto those materials. This was a lab-scale study (unlike papers I and II). This process generates P-loaded organic fertilizers.

Finally, we wanted to also know how installing NPHarvest impacts socio-technical systems around NPHarvest. These include the mainline wastewater treatment and mineral fertilizer production. Paper V examines the environmental impacts of deploying NPHarvest through an LCA study. The discoveries from this study were also used to guide the NPHarvest process development, reflecting on both aspects of Research Question 5.

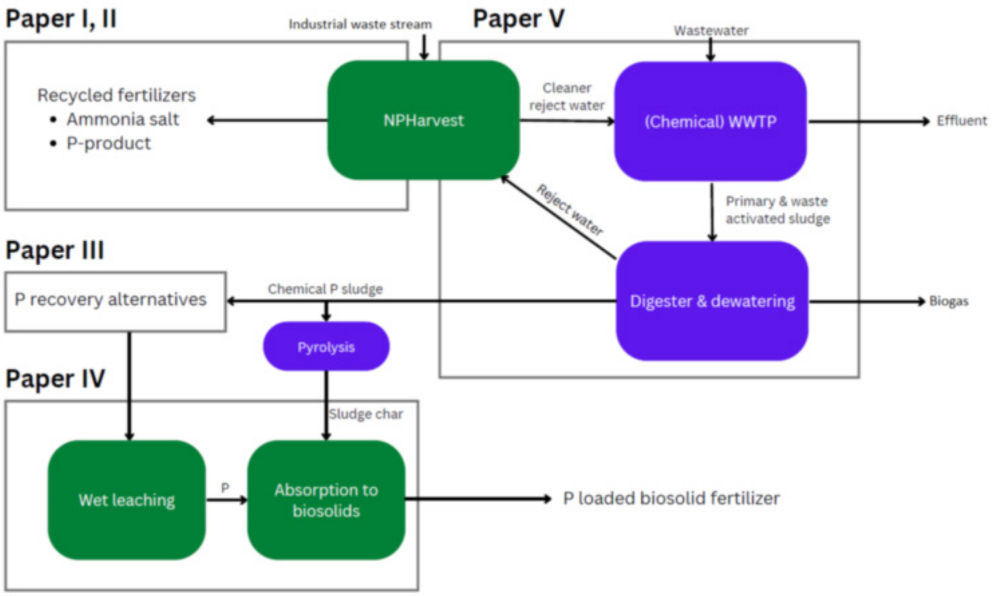

Figure 2. A visual representation of the specific focus areas for each of the papers in this dissertation. The WWTP effluent and biogas from the digester are out of the scope of this dissertation.

2. Methods

This chapter concisely presents the methods used in each paper. Figure 3 summarizes the methodological approaches for each paper. Papers I, II, IV and V are based on experimental lab- and field-scale work, while Paper III is a combination of literary analysis and lightweight modelling. Paper V combines experimental work and modelling through LCA methods.

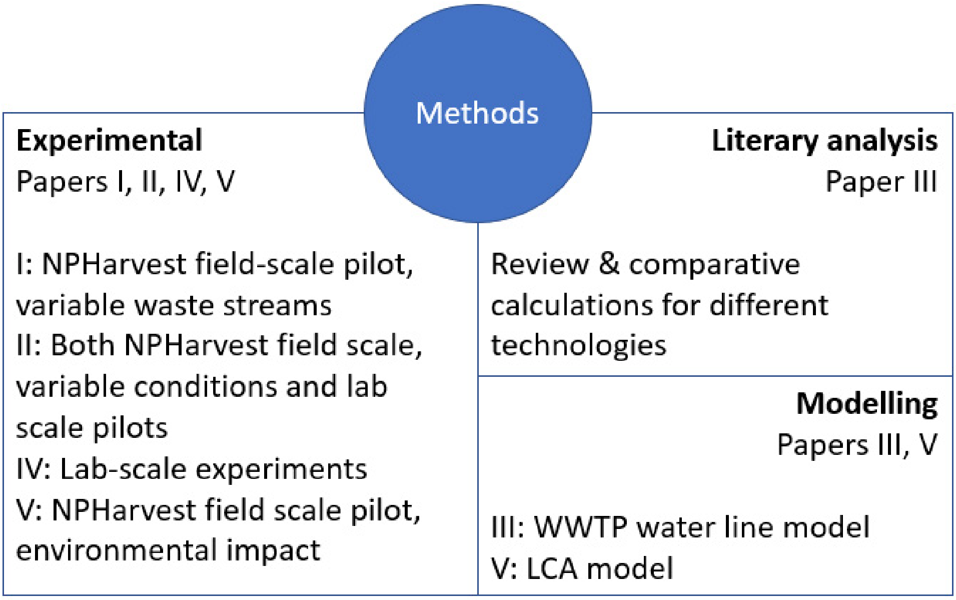

Figure 3. The methodological approaches for the studies included in this dissertation.

Papers I and II

The studies were conducted with the NPHarvest field pilot shown in Figure 1d. In addition, Paper II augments information with lab-scale studies using equipment that is similar to the one in Figure 1b. The field-scale process (in the order of the water flow) includes a collection tank, pre-treatment (slow and fast mixing & sedimentation) and N recovery (levelling tank, membrane contactor and acid tank). While the process equipment is the core that enabled these studies, many analytical methods were necessary. Characteristics of water and solid samples as well as membrane samples were analysed. These methods are explained in detail in papers I and II.

Paper III

Initially, Paper III was conducted purely as a review article using the available literature, namely the process data from several P recovery technologies suitable for chemical P sludge. However, as the work progressed, we created a representative wastewater plant in SUMO modelling software to allow for examining the recovery technologies on the same scale and including the water process in the evaluation. Thus, the resulting methodology for this paper was a literary review that was augmented by modelling, explained in detail in Paper III.

Paper IV

The selected P recovery method (developed based on the analysis in Paper III) was acid leaching from sludge and adsorption on different biosolids. The study was conducted on a lab scale using normal laboratory equipment. P was leached off sludge with sulphuric acid, and the biosolids used in the adsorption were sludge char (pyrolysis), lignin powder (forestry industry), humic substance from black liquor (pulp & paper mill) and commercial biochar. The liquid and solid samples were analysed thoroughly for their characteristics to assess the process recovery efficiency and end-product quality.

Paper V

LCA methodology is defined by ISO 14040:2006. The NPHarvest field pilot was operated to produce data for the LCA model in a similar manner as in papers I and II. A benchmarking technology was also evaluated in parallel with NPHarvest using literary data. The effects of nutrient recovery on the main water line treatment were systematically included in the model. The model was built on the assumption that a decrease in chemical consumption decreases the production of these chemicals, and the recycled nutrient products substitute mineral fertilizers at a 1:1 ratio.

3. Findings

This chapter briefly summarizes the results. Table 1 summarizes all experiments that produced data for this dissertation.

Table 1. Experiments, their locations, results and conditions.

Experiment	**Site/stream**	**Condition(s)/purpose**	**Result**	**Reference(s)**
1	Viikinmäki WWTP	Field test, continuous operation	Process works satisfactorily	Paper I
2	Gasum biogas plant	Field test, extremely high SS	Process underperformed	Paper I
3	Ämmässuo landfill	Only ammonia recovery, can membranes tolerate inorganic pollutants?	Process works spectacularly	Paper I
4	Urine		Process works well	Paper I
5	Viikinmäki WWTP	Longer field tests, parameter-specific tests	Parameter effects identified	Paper II
6	Aalto lab with sludge	P recovery from sludge	P recovery on biosolids successful	Paper IV
7	Helsingborg	Several months-long piloting, LCA, extensive product quality analysis	Process is robust in longer time frames, better understanding of the process	Paper V

3.1 NPHarvest recovery performance (Papers I, II and V)

The principles of recovery for NPHarvest are P precipitation with lime and possibly other chemicals and ammonia (that includes N) stripping with hydrophobic membranes (Paper I) The most detailed process description, along with a process schematic, can be found in Paper II. The process was tested with reject water from the Viikinmäki (Helsinki) treatment plant, Gasum (Riihimäki) biogas plant and Öresundsverket (Helsingborg) treatment plant, landfill leachate from Ämmässuo landfill, source-separated urine in the Aalto University lab and digested black water in Öresundsverket (Helsingborg) (Paper I). The results have been good in all locations except the Gasum biogas plant, where the suspended solids load was too much for our small pretreatment equipment at that time (Paper I).

Nitrogen recovery in the membrane contactor is between 70 and 80% (Paper I). The ammonia mass transfer coefficients (K) depend on the wastewater characteristics: Viikinmäki WWTP reject water had the highest K value despite having the lowest ammonia concentration (Paper I). In addition, the mass transfer is positively affected by the high flow rate (low hydraulic retention time), efficient solids removal in pre-treatment (which functions best at pH 10), low acid (in which ammonia is captured) strength and use of strong acid (nitric or sulphuric acid) (Paper II). Furthermore, the ammonia recovery process is the most efficient when the membrane material is PTFE and the membrane porosity is high (Righetto et al., 2022). The tests conducted in Helsingborg, Sweden, observed similar recovery efficiency in the membrane contactor (Paper V). However, due to ammonia emissions in the air in the pre-treatment process, the overall N recovery efficiency was ~50% (Paper V).

P recovery was over 80-90% at the Gasum biogas plant (Riihimäki), the Viikinmäki WWTP and Öresundsverket (Helsingborg) (papers I, II and V). The Viikinmäki WWTP required a mix of several chemicals to precipitate SS and P (papers I and II), whereas Öresundsverket (Helsingborg) only required calcium hydroxide (Paper V).

This summarizes the answer to research questions 1 and 2. The design principle for the membrane stripping process that allows the process to be feasible for wastewater is high SS tolerance without compromising the ammonia recovery efficiency. The key parameters for this process are flow rate (and hydraulic retention time), membrane material, pH and acid type and strength.

3.2 P extraction and adsorption on biosolids (Papers III and IV)

Phosphorus recovery from sludge can be done using several methods or technologies (Paper III). Paper III examined five different technologies that represent different recovery pathways (chemical and thermal). While conducting the study, we realized that it was necessary to compare the technologies on the same scale. This was not possible by simply comparing the original data, so a new method of comparison was developed for the paper by linearly scaling the processes to the same representative size. Figure 4 illustrates the methodological approach in Paper III.

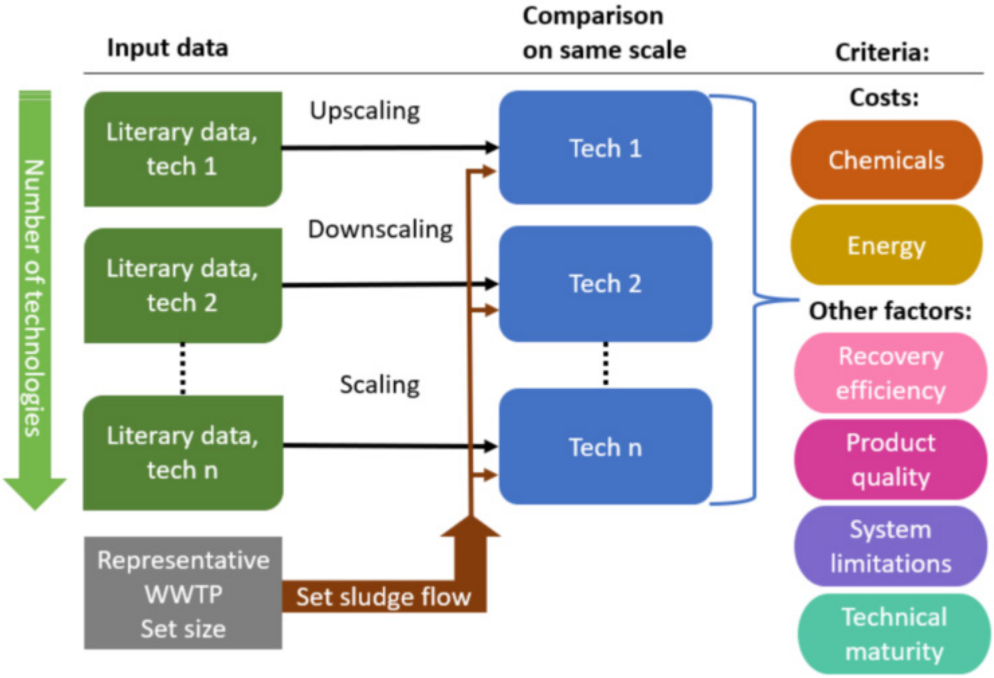

Figure 4. The methodological approach to comparing P recovery technologies on the same scale. The set size for WWTP and sludge flow means that it was set as constant in the analysis. Five technologies were analysed in Paper III, but any number of technologies is possible.

The technologies that we evaluated allow P recovery for 6–38 €/kgP (Paper III). A significant portion of the recovery cost is related to the precipitation metal. It was included in the analysis since the chosen metal affects the choice of the recovery technology. It is noteworthy that due to the political unrest at the time of writing (in 2022), the prices of some of the chemicals that are used in the water or the recovery process are volatile, resulting in recovery costs doubling between May and October (Paper III). The results of this paper are based on May 2022 data as they more accurately represent a stable economy.

A critical factor affecting the recovery cost is the availability of a mono-incinerator, which lowers the recovery cost because it allows the recovery from sludge ash (Paper III). The recovery cost for Thermochemical sodium sulphate and White P recovery processes were 6 and 12 €/kgP, respectively. However, not all locations in the world have access to mono-incineration. This is the case in Finland, where (we assume) investing in a mono-incinerator does not make financial sense. P extraction from sludge (as opposed to sludge ash) is the most feasible option in these cases (Paper III). The recovery costs for wet leaching and struvite precipitation, magnetic vivianite precipitation and sludge melt gasification are 38, 15 and 28 €/kgP, respectively (Paper III). The most feasible, however, is not sufficient, as these recovery costs are significantly higher than the market prices for fertilizers (Paper III). Thus, these processes are not economically feasible (Paper III).

However, the choice of P recovery technology should not be simplified to a single cost value. Recovery efficiency and end-product type and quality are important factors to consider (Paper III). We proposed a decision-making tool to help in finding the most suitable process for each reader's conditions (Paper III)

We chose wet leaching as the process to be developed further for Paper IV. The majority of the costs in the recovery itself (excluding the water process metal cost) are tied to citric acid and sodium hydroxide (Paper III). The P recovery process we designed for Paper IV, struvite precipitation, was replaced with adsorption onto biosolids, which swaps expensive chemicals for waste-based material to lower economic costs, and improved environmental performance. However, Paper IV did not include an economic evaluation or an environmental assessment as it focused on a proof-of-concept laboratory study.

The biosolid materials tested were biochar, commercial lignin, sludge char and humus extracted from black liquor (Paper IV). The material that performed the best was sludge char when adsorption capacity is examined. The sludge char adsorption capacity was ~50 mgP/g, while the other materials reached ~40 mgP/g in 70 hours in the kinetics experiments (Paper IV) The adsorption process is dependent on initial P concentration and temperature (Paper IV). In isotherm and temperature tests, the adsorption capacity went as high as 300 and 500 mgP/g, respectively (Paper IV). Modelling the adsorption process showed that the biomaterials obey a pseudo-first-order kinetic model and that adsorbents are heterogenous, meaning that the adsorbate physiochemically bonds with the functional groups of adsorbents with different adsorptive energies (Paper IV). All loaded materials contained ~10% P after adsorption, and the overall recovery efficiency from sludge to loaded adsorbent was 21%. Thus, all tested waste materials would be acceptable materials to recycle leached P. However, sludge char is the most attractive option since it functions best as an adsorbent and has no other purpose as a waste flow (at least at the time of writing Paper IV).

3.3 Fertilizer end products

The characteristics of the fertilizer end products generated by the NPHarvest process are not covered in any of the research questions, but they are still relevant for this dissertation. NPHarvest generates two products, one for phosphorus and one for nitrogen (Paper I). The quality and the boost to plant growth were evaluated by Konola in her master's thesis (Konola, 2019). Her work showed that the end products have good hygienic quality, low heavy metal concentrations and that they improve plant growth (Konola, 2019).

A more thorough evaluation of the quality of the end products was performed while conducting the LCA study (Paper V). The quality of the products is reported in a Swedish Water Research (Svenska Vatten Utvecking, SVU) report (Högstrand et al., 2022). The nitrogen product is pure ammonia salt that is liquid as it exits the process but when dried, it forms white crystals with 20% nitrogen content (Högstrand et al., 2022) when produced with sulphuric acid. The resulting salt is ammonium sulphate in diammonium form ($(NH_4)_2SO_4$) at pH 3 (Paper II). This is comparable to commercial ammonium sulphate, which has 21% N content. If nitric or phosphoric acids are used, they generate ammonium nitrate or ammonium phosphate (Paper II). Ammonium phosphate is the most valuable of the three salts, but it is not the most effective ammonia recipient at a low pH range (between 1 and 3) (Paper II). Using diluted acids (0.5 mol/l instead of 1 mol/l) have no impact on the end-product quality (Paper II).

The phosphorus product reflects the source material quality to some degree, but it generally has 1–3% phosphorus, ~8% calcium and ~10% carbon (Högstrand et al., 2022). The ammonia salt is very pure since the membrane separates ammonia very selectively, but the P-product may contain impurities that are present in the raw wastewater (Högstrand et al., 2022). The high pH of the process sanitizes the biological activity in the product, but organic pollutants persist (Högstrand et al., 2022; Paper I). We bought external services to analyze 320 different substances in the products (Högstrand et al., 2022). Low heavy metals mass concentrations were found in the end products (for example ammonium sulphate: Pb 6.6 mg/kgTS, Cu 3 mg/kgTS, Cr 0.3 mg/kgTS; and P-product: Pb 0.6 mg/kgTS, Cu 0.67 mg/kgTS, Cr 1.1 mg/kgTS). These are comparatively lower than similar values for the sludge from the wastewater treatment process and much lower than the legal limits in Finland and Sweden (Högstrand et al., 2022). Other micropollutants, like PAH, PCB, nonylphenols and ibuprofen, were not detected in the majority of samples (Högstrand et al., 2022).

3.4 NPHarvest environmental performance (Paper V)

The LCA study (Paper V) was conducted to steer the NPHarvest process development to the most sustainable path already from an early development phase. A proper analysis was necessary to understand the connected impacts between the nutrient recovery technology (NPHarvest) and its surrounding environment. Thus, how the nutrient recovery affects the main water line treatment process (mainline impact, MLI) was included in the LCA model. In other words, how much does energy and chemical consumption decrease due to the lower nutrient load? This novel approach has been documented only partially in the previous literature, with each paper focusing only on some aspects of MLI (Paper V). The mainline variables form a substantial contribution to the overall LCA results (Paper V).

The impact categories included in Paper V were Global Warming Potential (GWP), Eutrophication Potential (EP), Acidification Potential (AP), several Toxicity Potentials (ETP, HPTc, HPTnc), Primary Energy Demand (PED) and Abiotic Depletion of Elements (ADPe). The benefits of nutrient recovery were clear. The substituted materials in mineral fertilizer production, reduced N2O emissions and reduced precipitation metal consumption form the main beneficial effects (Paper V). These effects are present in GWP, EP and ADPe impact categories (Paper V). However, ammonia emissions dominated EP and AP impact categories (Paper V).

Overall, both nutrient removal (reduced load on the main water line) and recovery (substitution of mineral fertilizers) are needed to reach a climate-neutral state (Paper V). However, if the electricity mix leans towards fossil fuel production, this decreases the climate category performance (Paper V). Further process development needs to solve the ammonia emissions issue and reduce energy consumption related to ammonium salt drying (Paper V).

4. Discussion

4.1 NPHarvest as a process

The NPHarvest results have been positive through the years (Pradhan et al., 2017; Kaljunen, 2018; Righetto et al., 2022; papers I and II). Recovering nutrients from concentrated flows with this process is a good way to produce recycled nutrients with relative ease and at a low cost while improving the environmental aspects of the treatment facility. However, it is important to understand that nothing is simple when it comes to the development of industrial-scale processes.

When transitioning from the lab to the field, the number of variables increases and the amount of control decreases. This makes our tests less reliable. On the field scale, we have compensated for the loss of accuracy by increasing the number of samples taken and conducting long test runs. As an example, calculating the ammonia mass balance during a test run is difficult to conduct accurately, since the balance is based on lab analysis of samples that were taken once per day, combined with online liquid flow measurements that could be in error by up to 15% (Li et al., 2009). We did not have an option to directly measure the ammonia emission to the air at piloting sites, so other flows need to be calculated and the offset is assumed to be ammonia emission. This calculation is bound to be inaccurate despite the high number of samples and/or analyses.

Uncertainties are not limited to the process performance. We dared to delve into the realm of economics already in an early phase of the project. We needed to understand financial feasibility if we would build something sustainable in the future. But this also meant guesswork and estimations. Upscaling costs are especially subject to change in the current politically volatile environment. Paper I utilizes a scientific method to estimate upscaling costs (Paper I). It is a reliable method, but the calculations are based on the manufacturing costs of the field equipment, which included a great deal of manual assembly. This inflated the upscaled cost. To find a more realistic estimation of the economics, we also relied on external experts. Furthermore, the profitability of the process, which defines the return on investment, is calculated through parameters that are undergoing massive changes due to the Russian invasion of Ukraine in Spring 2022.

The most useful findings besides the process performance itself are the shortcomings of the process. Ammonia emissions are a problem with a simple fix on a larger scale since it is possible to seal the process containers so that there are no gaseous emissions from the process. In addition, 70-80% recovery efficiency for nitrogen is not good enough for competitive economic and environmental performance. As shown in Paper V, nitrogen recovery efficiency affects the maximum environmental benefit reaped from decreased N2O emissions. In addition, ammonia salts are the most valuable product and thus a key aspect in the process's financial sustainability. Improving the recovery efficiency above 90% is possible and necessary when moving towards commercial application (Paper V).

A reader could also question the entire point of recovering nitrogen with NPHarvest (or any other technology). After all, the atmosphere is full of N2, and the real problem is the Haber-Bosch process that is operated with fossil-based energy sources (Smith et al., 2020). If we could replace fossil electricity with renewable and natural gas with a renewable alternative, that would create a solution that is called green ammonia (Salmon and Bañares-Alcántara, 2021). Well, yes and no. It would solve the climate issue in ammonia production. But a significant amount of energy would still be consumed, which could be used for something else. In addition, if the concentrated side streams are not treated with nutrient recovery, this maintains energy consumption and N2O emissions in the wastewater treatment process at the current level. Finally, nitrogen (and phosphorus) flows are already past the safe operating space in planetary boundaries (Steffen et al., 2015). The nitrogen cycle that involves the atmosphere as a major component (on a conceptual level) does not mitigate the plentiful nitrogen flows in the natural environment. Thus, I believe recycling nitrogen offers more benefits than focusing on green ammonia. However, both pathways are most likely needed to replace all fossil-based ammonia.

4.2 Competing processes

There are other processes that treat concentrated waste streams, either for removal (Ma et al., 2016) or recovery (Egle et al., 2016; Van der Hoek et al., 2018; Ye et al., 2020). Removal processes are typically biological and aim to decrease the energy consumption of the treatment plant by returning N to the atmosphere with a less energy-consuming process (Ma et al., 2016). In a world where resource recovery is increasingly important, this approach seems short-sighted.

The true competitors are the ones that aim at nutrient recovery. The industrial standards are air stripping (Katehis et al., 1998) and evaporation stripper (Bonmatí and Flotats, 2003) for N and precipitation as either struvite or other precipitates for P (Egle et al., 2016). Several papers have arrived at the conclusion that gas permeable membranes are the most economical solution (Munasinghe-Arachchige and Nirmalakhandan, 2020; Al-Juboori et al., 2023; Beckinghausen et al., 2020). This represents NPHarvest, which differs from these processes in that the nutrients are recovered using fewer resources, also by utilizing SS tolerant design. There is no need to increase pressure or the temperature, and P recovery occurs spontaneously while increasing pH for ammonia recovery (Paper I). This is the competitive edge that NPHarvest has over other technologies.

4.3 Products

There are three products included in this dissertation. NPHarvest products are ammonia salt and P-product, and loaded biosolids are the third product. The ammonia salt's quality is practically the same as a similar commercial product, but the P-product is more difficult to classify since it is a mix of different solids and precipitates (Paper I).

The P-loaded biomaterials are more accurately defined. The motivation to use waste-based biomaterial as P fertilizer is to provide a recycled carbon source to the agricultural environment

(Paper IV). However, in the case of sludge char, the same sludge was the origin of the leached phosphorus and the adsorbent. One could wonder if the WWTP sludge could be pyrolyzed and used as fertilizer. However, the raw sludge char contains only 2–3% P, and the availability of P after pyrolysis is poor (VVY, 2019), so the loaded material is more concentrated and thus better suited for fertilizing purposes.

Throughout the tests, the products have been clean (Högstrand et al., 2022; Konola, 2019). The sludge and wastewater we have been treating have not had noticeable heavy metal concentrations (Högstrand et al., 2022; HSY, 2020), which may be unique to the Nordic environment. Bacterial activity has been eliminated by low (wet leaching for loaded biosolids) (Paper IV) or high (NPHarvest products) pH (Paper I). Organic pollutants and pharmaceuticals remain in the P-product. For fertilizing purposes, the top-most worry is the presence of pollutants in the natural environment rather than the contamination of food, since the pollutants are rarely taken up by plants (Levén et al., 2016).

The European Union updated the regulation for fertilizer products in 2022 (European Commission, 2022). This enables recycled products of certain qualities to receive the CE marking. Ammonia salt falls in this category, but the other two products are more difficult to categorize. However, new legislation aims to add new material categories as needed. This means that recycling products containing P are not ruled out for acceptable materials in the long term.

4.4 Wastewater treatment plant performance improvement

Both the economic and environmental performance of the wastewater treatment plant is the result of assumptions (Paper V). Nutrient recovery through NPHarvest influences the system it is a part of. One of the goals of this work was to find or develop suitable technology for the Nordic environment, and our assumptions reflect this. Sweden, for example, has an electricity mix that is practically only renewables and nuclear energy – meaning fossil-free. This affects the GWP impact category performance of the process and the WWTP attached to it. Also, the nitrous oxide emission factor is uncertain, not just for us but for the entire academic world (Delre et al., 2017).

However, the work in papers I and V definitely shows that there is significant potential for improving the environmental and economic performance of WWTPs. Another idea for optimization is to apply only the N recovery part of NPHarvest to CPR WWTPs, since their reject water has very little P (Paper I) and recover P from digestate using, for example, with the technology presented in Paper IV. Then, the full NPHarvest technology could be applied to an EBPR plant's reject water since it has significant P concentrations (Högstrand et al., 2022).

A rough case can be calculated for the economic benefits if NPHarvest were applied to a CPR WWTP (calculating only N) that generates 2250 m3/d with 850 mgNH3-N/l. Viikinmäki WWTP personnel gave me an estimate that their process consumes methanol at about 2.8 kg/kgN, aeration energy at 4.5 kWh/kgN and calcium hydroxide at 2 kg/kgN. Assuming the N recovery is 90%, the amount of ammonium salt generated is 8100 kg/d ammonium sulphate, 9800 kg/d ammonium nitrate or 6106 ammonium phosphate. For this calculation, I assumed the following prices: methanol 550 €/ton, energy 0.1 €/kWh, calcium hydroxide 300 €/ton, ammonium sulphate 400 €/ton, ammonium nitrate 900 €/ton and ammonium phosphate 750 €/ton. These prices are based on information on various Internet sites such as Alibaba.com. Using these numbers, the savings in the main wastewater treatment process are 5400 €/d, and the value of ammonium salts generated is 3200 €/d with ammonium sulphate, 8900 €/d with ammonium nitrate and 4600 €/d with ammonium phosphate. If we further assume that NPHarvest operational costs (for energy and chemicals) are 3 €/m3 (which is realistic when using sulphuric acid), the profit (savings + ammonia salt price – operational costs) is between 1900 and 6500 €/d. The calculation is the most accurate for ammonium sulphate, and this would yield a created value of ~700,000 € per year.

4.5 Central or local nutrient recovery

NPHarvest can be applied at a small housing complex to treat source-separated waste flows as well as large treatment facilities (Paper I). Currently, a central application is the only sensible option since source-separated flows do not exist apart from a few example cases, like the Hiedanranta area in Tampere, Finland (Lehtoranta et al., 2022). Since the conflict in Europe has changed the world, there is a critical need for fertilizers. In the short term, securing agricultural production is the top priority. The benefits of nutrient recovery can only be realized by investing in recovery technologies. It is an obvious point to make, but plant owners/operators should invest in nutrient recovery technology as soon as possible (probably yesterday). In practice, reject water is an easy target for capturing nitrogen and phosphorus, and it is a question of will and funding to see it through and start producing recycled nutrients to fulfil the needs of society.

In the longer term, it is interesting to speculate on the future of wastewater treatment. Is a central treatment solution better than local systems? The central treatment option offers an efficient way to continue what we have been doing for the past decades, but major improvements are more elusive. It is difficult to imagine how source separation could be done in a central system since the sewer infrastructure does not support separation. Source separation offers undiluted waste flow, urine and black water, which are straightforward to treat and extract nutrients from. But it requires local process control and a new distribution system for the products.

For example, if NPHarvest were implemented in every household in Finland and every toilet was separating urine and faeces, urine would be treated by NPHarvest to produce ammonia salts and calcium phosphates. The effluent (high pH liquid void of nitrogen and phosphorus) would be directed to the sewer network (possibly after dilution/neutralization to avoid calcium build-up and corrosive effects) and to the treatment plant. Faeces could be flushed down with this high pH effluent if the toilet did not have composting functionality. The person who was maintaining this system would have the NPHarvest products to sell or distribute. However, this imaginary local system relies on the activity of the person maintaining it. An interested person can make it work, but odds are the whole society is not interested. If this kind of system was built by interested people, over a longer time, the system would most likely fall into ruin because it would be maintained by an uninterested person. In this sense, a centralized system is more reliable.

4.6 How should the entire society change?

It feels like we humans have fallen victim to our own cleverness. Let us examine organic pollutants like PFAS compounds. Someone figured out a useful purpose for them (coating for cookware) but failed to foresee that these are very toxic compounds that are difficult to get rid of once they are released into the natural environment (Sunderland et al., 2019).

The main hindrance in recycling nutrients is the concern over pollution (or the high cost of recovering the nutrients in a pure form – this is where NPHarvest has an edge) (Paper III). If society would not consume all the unwanted compounds and materials, they would not be present in wastewater sludge, and they could be more readily recycled. In other words, the ideology of the circular economy should start with material and product design rather than reusing the existing waste because it is very expensive to reuse if it is contaminated by several unwanted compounds. However, having a wide variety of pollutants in the wastewater is inevitable since many different products are necessary to maintain human health or personal hygiene. Further research is required on both fronts: how to separate more efficiently and design products and materials that are not causing long-term issues for the natural environment. This will require a collective effort by all stakeholders: scientists, industrial actors, consumers, farmers and politicians.

Another change that would make a more sustainable society would be to move nutrients from places with an excess to places where they are needed. For example, 60% of the nitrogen that is applied to European agricultural areas is not taken up by plants, causing various environmental problems (de Vries et al., 2021). Agricultural waste streams, however, are too easy to deposit on adjacent farmland for nutrient recovery to be considered in this context.

These are just examples of practical actions that can be taken to improve society's sustainability. But how can we truly build a sustainable society from a philosophical point of view? NPHarvest holds the flag of sustainability high, but are we just maintaining a culture of consumerism and painting a shade of green on it by telling everyone that this is sustainable? Allow me to elaborate. First, the core issue of an unsustainable society is that the human population on Earth is consuming more than what the Earth's systems can provide without compromising the ability to provide life-supporting functions in the future (reflecting on Brundtland Commission, 1987). In this light, the total amount of consumption should decrease. Manufacturing and consuming a recycled product still utilize a great amount of resources along the cycle. If we brand recycled products like NPHarvest fertilizers as sustainable products that can be consumed without any care in the world, does that only encourage society to consume more although the goal was to consume less?

I used to think that it was difficult to build a profitable business by decreasing consumption. On one hand, this is true in the current capitalistic system. But on the other hand, it's not true at all, at least for businesses that are built on recycling material. If we as a society manage to decrease the total consumption of virgin natural resources while having our needs fulfilled, that must mean that most of the consumption is reused materials. I believe we can build a profitable business that focuses on supporting the recycling of critical materials, like nutrients, without compromising the future of the species.

5. Conclusions

This dissertation set out to find solutions for nutrient recycling in the Nordic environment. The work was conducted through several Aalto University research projects that focused on developing NPHarvest as a technology, finding methods to recover P from sludge and understanding the environmental effects of nutrient recovery.

NPHarvest set out to develop ammonia recovery using a membrane stripping application. Research questions 1 and 2 are focused on this topic: how to design the process and what are the key parameters for it. It turns out that the key is to design the process so that it tolerates high suspended solids concentrations so that the process becomes economically feasible in a wastewater environment. The key parameters are membrane material, hydraulic flow rates and acid type and strength.

P recovery from sludge is not feasible with any existing technology when considering the price per P mass (Research Question 3). However, different recovery processes are applicable in different situations, depending on the water process type, the desired recovery efficiency and the end-product type and purity. By selecting a part of one recovery technology suitable for the Finnish environment, P acid leaching, and developing that further, we discovered that the best waste-based biosolid to bind acid-leached P is sludge char (pyrolyzed sludge), which answered Research Question 4.

The NPHarvest environmental impact was evaluated through an LCA, as we needed to understand the current impacts and develop the process in a truly sustainable direction (Research Question 5). A significant weight in the assessment originated from the novel mainline impact examination, where the impacts of nutrient recovery on the wastewater treatment process performance were included. The process is climate-neutral already in this development phase, but ammonia emissions contributed to eutrophication and acidification. The next step in NPHarvest development is to mitigate ammonia emissions and improve N recovery efficiency. When fully implemented, these actions will result in an environmentally and economically sustainable nutrient recovery process. In general, further research should be focused on decreasing the cost of nutrient recovery and improving the purity of the recovered material.

While the untapped potential in nutrient recovery is considerable, tradition (how nutrients in waste streams are now used), the cost of technology and a negative perception (of the waste-based nutrient products) stand in the way of fast implementation. However, I hope it is clear that fast implementation is desperately needed to overcome the issues caused by both the consumption patterns of our oversized population and world leaders who have a few screws loose. I want to act as I preach. That is why the research presented here is currently in the process of commercialization. This way, we can have an immediate and true impact on society – as soon as 2024!

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The explosive growth of the human population would not have been possible without processes that bind nitrogen from the atmosphere to ammonia and mined phosphate rock to easily soluble products. However, the linear consumption pattern (as opposed to circular) has backfired on us. As a result, the nutrient systems we rely on consume significant amounts of energy and create an array of environmental problems. This dissertation focuses on the question: 'How can we close the nutrient loop?'

This dissertation is built around NPHarvest, a nutrient recovery technology developed at Aalto University. NPHarvest recovers both nitrogen and phosphorus from liquid waste streams with high efficiency. The end products are clean ammonia salt and solid material that contains phosphorus, calcium and carbon.

Nutrient recovery is a shift towards transforming treatment plants to resource recovery plants, in addition to enabling the wastewater treatment facilities to reach better cost-effectiveness. The true meaning of sustainable technology is also discussed on these pages.

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