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Environmental and economic sustainability of the nitrogen recovery paradigm: Evidence from a structured literature review.

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Abstract

Our economy drives on reactive nitrogen (Nr); while Nr emissions to the environment surpass the planetary boundary. Increasingly, it is advocated to recover Nr contained in waste streams and to reuse it 'directly' in the agri-food chain. Alternatively, Nr in waste streams may be removed as N_2 and refixed via the Haber-Bosch process in an 'indirect' reuse loop. As a systematic sustainability analysis of 'direct' Nr reuse and its comparison to the 'indirect' reuse loop is lacking, this structured review aimed to analyze literature determining the environmental and economic sustainability of Nr recovery technologies. Bibliometric records were queried from 2000-2020 using Boolean search strings, and manual text coding. In total, 63 studies were selected for the review. Results suggest that 'direct' Nr reuse using Nr recovery technologies is the preferred paradigm as the majority of studies concluded that it is sustainable or that it can be sustainable depending on technological assumptions and other scenario variables. Only 17 studies compared the 'direct' with the 'indirect' Nr reuse route, therefore a system perspective in Nr recovery sustainability assessments should be more widely adopted. Furthermore, Nr reuse should also be analyzed in the context of a 'new Nr economy' that relies on decentralized Nr production from renewable energy. It is also recommended that on-par technology readiness level comparisons should be carried out, making use of technology development and technology learning methodologies. Finally, by-products of Nr recovery are important to be accounted for as they are reducing the environmental burdens through avoided impacts.

Keywords: life cycle and economic assessment, circular economy, nutrient reuse, blue/green ammonia, urine, manure

List of acronyms

AD	Anaerobic Digestion
AS	Activated Sludge
CO_2-eq.	CO_2 equivalent
HB	Haber-Bosch
K	Potassium
LCA	Life Cycle Assessment
N/DN	Nitrification/Denitrification
Nit/DNit	Nitritation/Denitritation
Nr	Reactive Nitrogen
P	Phosphorus
PN/A	Partial Nitritation/Anammox
SM	Supplementary Material
TRL	Technology Readiness Level
Tg	Teragram

1 Introduction: Nr in the Anthropocene

Nitrogen is an essential element for all living organisms. The biggest pool of nitrogen on our planet is the atmosphere. By volume, dry air consists for 78% of dinitrogen (N_2), or an estimated 3,878,000,000 Teragram (Tg, million ton N)[1]. Since the early 20th century, about 48% of the global population have been depending on this atmospheric pool by industrially converting N_2 to ammonia (NH_3) and derived products like urea and nitrate through the Haber-Bosch (HB) process (Erisman et al., 2008). On a global scale, this process uses large amounts of fossil energy (1% of global energy and 2% of the global natural gas use) (Cherkasov et al., 2015) and is responsible for significant CO_2-equivalent (CO_2-eq.) emissions (1.2%) (Smith et al., 2020). Haber-Bosch further accounts for roughly one third (100-165 Tg N/year) of the total global reactive nitrogen (Nr) generation, with the remained being derived from agricultural biological nitrogen fixation (50-70 Tg N/year), fossil fuel combustions (27-33 Tg N/year) and natural biological nitrogen fixation (58-128 Tg N/year) (Galloway et al., 2004; Scheer et al., 2020). The massive increase in anthropogenic Nr flows and its associated emissions to the environment are profoundly altering the global biogeochemical N cycle. The landmark publications of Rockström et al. (2009) and Steffen et al. (2015) show that among all global environmental impacts, Nr is most severely exceeding the carrying capacity of our planet (about 3 times), with agricultural fertilizer use, livestock wastes and urban wastewater having major responsibility for this (Campbell et al., 2017; Fowler et al., 2013). Nr production is therefore one of the most important elements through which humans are altering the global geochemical balance and leaving their footprint on what can informally be referred to as the Anthropocene (National Geographic, 2022).

[1] dry air mass as $5.1352 \pm 0.0003 \times 10^{18}$ kg in atmosphere, 75.5% nitrogen by mass in air. https://www.cs.mcgill.ca/~rwest/wikispeedia/wpcd/wp/e/Earth%2527s_atmosphere.htm

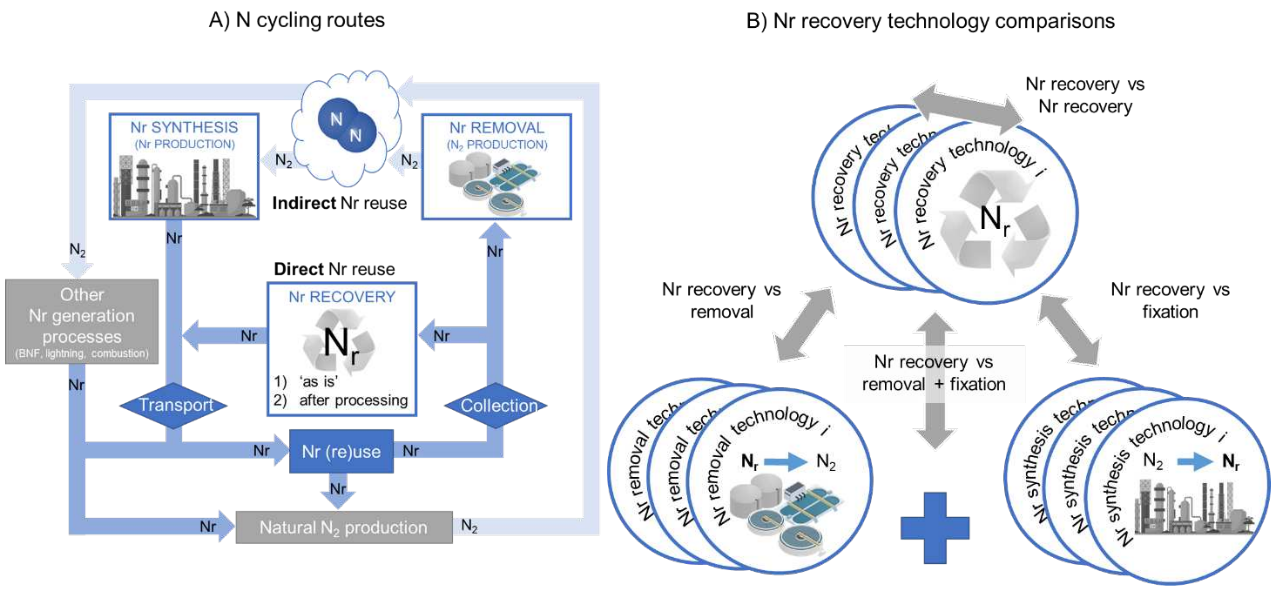

Figure 1: A) Indirect and direct reuse routes for reactive nitrogen (Nr) species in waste streams. B) Possible Nr technology comparisons used in sustainability analyses.

To reduce Nr pollution, different nitrogen management approaches are proposed. A preventive approach seeks to minimize Nr losses by increasing nitrogen use efficiency in the agri-food chain, and as such reduce diffuse Nr emissions (Kanter et al., 2020). A complementary curative approach proposes to recover Nr from point sources, such as sewered wastewater, and reuse it in the agri-food chain, thereby reducing overall Nr production and its associated impacts (Arashiro et al., 2018). For such collectable point source waste streams, three main N cycling routes can be distinguished: two for 'direct reuse', and one for 'indirect reuse'[2] with N passing over the atmosphere (Figure 1a). The first direct reuse route uses the waste stream 'as is' or after minimal processing (e.g., solid/liquid separation), typically in the form of ammoniacal and/or organic nitrogen, an approach often practiced for animal manure and livestock slurries. A second and more demanding direct reuse approach entails concentrating Nr in products after processing or refinement (i.e., removal of other elements, pollutants and/or organic matter). Direct Nr reuse 'as is' and the reuse through refinement is commonly considered as Nr 'recovery', and is adopted as a terminology throughout the document. Indirect Nr reuse is the nitrogen cycling method going over atmospheric N_2. The first step is usually termed Nr 'removal' and is based on the biological conversion of Nr to N_2. The second step is the (re)synthesis of Nr in centralized HB plants, or through biological nitrogen

[2] The terminology is derived from the concept of direct and indirect potable water reuse. In which indirect reuse refers to a passage of the water through the natural environment.

fixation. Through these steps, N molecules are indirectly reused by a removal-fixation-usage sequence across the atmospheric N_2 cycle. The atmospheric N_2 stock is amongst others maintained by nitrification/denitrification (N/DN) and anaerobic ammonium oxidation (anammox) (Burgin and Hamilton, 2007), both of which processes are applied in wastewater treatment for the removal of Nr.

Over the past decade, sustainability concerns have boosted the development of Nr recovery technologies for concentrated and refined products (see supplementary material (SM) section 1 for a literature search). The underlying concept of this Nr recovery paradigm is that it can provide a win-win solution as it can reduce the economic and environmental costs of Nr removal and Nr synthesis. However, the sustainability of Nr recovery and nutrient recovery in general is by no means self-evident. Maurer et al. (2003) used operational energy requirements to evaluate scenarios of source-separated urine treatment, concluding for instance that the route of Nr removal via partial nitritation/anammox (PN/A) and HB Nr synthesis required less energy than Nr recovery with air stripping and absorption in H_2SO_4. Similarly, a study conducted for a sewage treatment plant in Amsterdam suggested that Nr recovery results in only a limited improvement in sustainability and that a range of recovery technologies have higher N_2O emissions and energy demand than the combination of PN/A and HB (van der Hoek et al., 2018). The authors concluded that radical changes, such as separate collection and treatment of urine, and application of several Nr recovery methods in parallel would be required to substantially improve the sustainability of the biogeochemical N cycle. In a recent review of life cycle assessment (LCA) studies on nutrient recovery from wastewater, Lam et al. (2020) demonstrated that for sludge recycling and recovered products environmental benefits only marginally outweigh environmental impacts. However, Lam et al. (2020) only compared different Nr recovery options for direct Nr reuse with each other and did not systematically explore differences between Nr recovery and Nr removal, or Nr recovery compared to the indirect route via the combination of Nr removal and synthesis (Figure 1B). Neither did they detail the technologies for the recovery technologies investigated.

The study of Lam et al. (2020) also highlights the difficulties in comparing outcomes of LCA and sustainability analysis studies, because the diversity of methodological choices and assumptions (e.g., system boundaries, functional units, indicator selection) does not enable the identification for the reasons underlying the observed differences in the results. Finally, any assessment of technologies should account for the multi-dimensional nature of sustainability often summarized in the triple bottom line of profit, planet, people. That is, it should be assessed whether technologies are cost effective (profit), reduce environmental burdens (planet), while also being socially acceptable (people).

The aim of this study is therefore to critically and systematically analyze how the environmental and economic sustainability of Nr recovery technologies is evaluated in literature. Specifically, it is the aim to judge whether the direct or the indirect Nr reuse routes are preferable from a sustainability perspective. It is further the aim to identify methodological challenges that affect outcomes of sustainability assessment for Nr recovery technologies. Attending to this aim will provide insights as to whether the new paradigm of Nr recovery is environmentally and economically sustainable. The research will further provide methodological recommendations for future sustainability assessment studies. For methodological reasons, the social sustainability of Nr recovery technologies is not addressed in the systematic review but is part of the discussion (section 5.4).

The next section briefly describes the state-of-the-art of technologies related to direct and indirect Nr reuse routes, thereby providing important background information for the reader to contextualize the discussion. Thereafter, the methodology section is outlining the research approach taken with more information available in the SM. In sections 4.1-4.3, the studies included in the review are characterized including the technologies used, the methods applied and whether indirect and direct Nr reuse routes are investigated. This analysis is then used to answer the central research question as to whether the Nr recovery paradigm (i.e. direct reuse) is more sustainable than indirect reuse (section 4.4). In the discussion section, the findings are integrated and a critical evaluation of Nr reuse sustainability, methodologies, and a perspective on the role of Nr recovery in a new Nr economy is provided. The article ends with a set of conclusions and recommendations for future research.

2 Technologies for N cycling – An overview

2.1 Technologies for indirect Nr reuse: Cycling over the atmospheric N pool

2.1.1 N_2 production: current state and recent advances

In most liquid waste streams, reduced Nr is present, either as ammoniacal nitrogen and/or as organic nitrogenous compounds. Biological nitrogen removal based on autotrophic nitrification, and subsequent heterotrophic denitrification of the formed nitrate to N_2 is a process globally applied in wastewater treatment plants (WWTP) since the 1970s (Focht and Verstraete, 1977). Alternatives for this N/DN include the autotrophic/heterotrophic nitritation/denitritation (Nit/DNit) and the fully autotrophic partial nitritation/anammox (PN/A). Both Nit/DNit and PN/A are termed shortcut Nr removal processes, as they do not rely on the formation of nitrate but terminate the oxidation of ammoniacal nitrogen at nitrite, thereby reducing costs because of savings in aeration energy and chemical oxygen demand (Table 1).

Shortcut Nr removal has mainly been applied to treat the warmer (>25°C), higher strength (0.5-1.5 g NH_4^+-N/L) liquid streams after anaerobic digestion with low COD concentrations (<1g COD/g N) – e.g., sludge reject water in the side stream of municipal wastewater treatment plants. Industrial applications of Nit/DNit are limited thus far. For PN/A, in 2014 100 full-scale plants were commissioned, and the number has further increased since then (Lackner et al., 2014). Concerning municipal wastewater, the application of PN/A to the lower temperatures and Nr concentrations in the water line or mainstream is of great research interest as it may result in energy positive WWTPs. This has for instance been demonstrated in two installations (Strass, Austria and Changi, Singapore) (Wett et al., 2015; Winkler and Straka, 2019). It is expected that CO_2-eq. emissions from operation can be reduced from 7.0 kg CO_2-eq/kg Nr for a N/DN process to 4.5 kg CO_2-eq/kg Nr for a mainstream PN/A at similar N_2O emission (Table 1, for calculations SM section 3 Table S1). Meanwhile, costs may nearly halve to about 2.5 euro/kg Nr removed, due to savings in energy demand and carbon source consumption (Fux and Siegrist, 2004).

Table 1: Simplified comparison of three Nr removal pathways and the Haber-Bosch (HB) process. The O_2 and COD requirements for three Nr removal pathways are based on the stoichiometries including anabolism. a (Maurer et al., 2003), b (World Bank, 2021), c (Smith et al., 2020), d (Wang et al., 2021) e (Brentrup et al., 2016), f(Fux and Siegrist, 2004), g own calculations – using CO_2-eq emissions of 0.276 kg CO_2-eq./kWh, h own calculations based on (Maurer et al., 2003) and stoichiometric reduction of aeration energy demand.

	Unit	N/DN	Nit/DNit	PN/A	HB fertilizer
Oxygen demand	[g $O_{2\ needed}$/g $Nr_{oxidized}$]	4.2	3.2	2.0	NA
COD demand	[g $COD_{required}$/ g $Nr_{removed}$]	4.3	2.6	0.0	NA
Energy demand	[$kWh_{primary}$/kg $Nr_{fixed\ or\ removed}$]	12.5[a]	9.3[h]	6[h]	9.5[c] ($N\text{-}NH_3$) – best available technology
Costs / price	[EUR/kg Nr] (values recalculated to 2021 factor 1.27 for f)	5.2[f] (with external C source)		3.17[f] (for sludge reject water).	0.4-0.5 (2020) 0.6 – 2 (2021) (urea)[b]
Greenhouse gas emissions	[kg $CO_{2\text{-}eq}$/kg Nr]	5.0-7.0[g] (without-with methanol; 1% N emitted as N_2O) [g]	4.8-6.0[g] (without-with methanol; 1% N emitted as N_2O)[g]	4.5[g] (1% N emitted as N_2O) [g]	2[c] - 3.5[e] (urea) 2-3 (NH_3)[c, d]

2.1.2 Haber-Bosch Nr synthesis

The HB process converts atmospheric N_2 and hydrogen gas to ammonia in a reaction that is driven by high temperatures (~650-750 K), pressure (~100-200 bar) and a metal catalyst (Cherkasov et al., 2015).

Hydrogen is obtained from steam reforming with major feedstock being natural gas and coal, accounting for about 66% and 30% of the global consumption (International Fertilizer Organisation, 2014). The most efficient HB installations operate at 9.5 $kWh_{primary}$/kg $N\text{-}NH_3$, which is close to the theoretical limit of the process at 7.5 $kWh_{primary}$/kg $N\text{-}NH_3$ (80% for methane as feedstock and 20% to drive the process) (Cherkasov et al., 2015; Smith et al., 2020). Therefore, it has been argued that the process is highly efficient and restricted to further energetic process improvements. Along with the increase in energy efficiency, CO_2-eq. emissions decreased to 2-2.1 kg CO_2-eq./kg $N\text{-}NH_3$ (Smith et al., 2020). Factors contributing to this decrease are the shift to natural gas as feedstock instead of coal (International Fertilizer Organisation, 2014) and the reuse of about 50% of the CO_2 released in the process for urea production (Dawson and Hilton, 2011). The efficiency and large scale (2000-3000 tons NH_3/day) of HB installations result in a cost of 0.4-2 euro/kg N-urea (2020-2021). Only due to dramatic increases in energy prices in 2021 costs for urea-N now approach the order of magnitude of Nr removal technologies (World Bank, 2021) (Table 1).

2.2 Recovery technologies for direct Nr reuse

Nitrogen concentrations in wastewater are low. Even concentrated streams like undiluted urine (~4-14 g Nr/L, (Larsen et al., 2021) and manure slurry (max. ~3.5 g Nr/L, (Baldi et al., 2018; Cai et al., 2013)) contain maximally 1.4% Nr or less, compared to solid synthetic inorganic fertilizers such as ammonium nitrate and urea containing around 35-47% Nr. Recovery approaches from liquid waste streams (incl. the solids transported) are based on four groups of processes (Figure 2; SM Table S2): (i) capture, partition or 'accumulate' Nr from the liquid by assimilation in microbial biomass; (ii) biological Nr species conversion by either mineralization, releasing NH_4^+ from organic matter, or nitrification, converting NH_4^+ to NO_3^-; (iii) refinement of Nr through extraction, and (iv) the concentration of Nr through water removal in order to increase N concentrations, stabilize the product (i.e., drying) and to reduce transportation costs. The recovery pathways used are diverse and depend on the total nitrogen concentration, the speciation of Nr as total ammoniacal nitrogen or organically bound nitrogen and the level of contamination of amongst others: suspended solids, organics, heavy metals and pathogens. Some Nr recovery methods are feasible directly on water with low Nr concentrations, after removal of organics and suspended material. For sewage, microalgae production is such an example as well as ion exchange (Arashiro et al., 2019; Huang et al., 2020). Streams with higher Nr concentrations in the order of g Nr/L and few interfering compounds, can directly feed into the recovery process, as is typically considered for urine. Manure slurries contain relatively high Nr levels, but also suspended solids, and may undergo anaerobic treatment and/or solid/liquid separation prior to Nr recovery (Baldi et al., 2018), but have also been used as a substrate for stripping/absorption either directly on the manure or after solid/liquid separation (De Vrieze et al., 2019). Agri-food industry, including potato, brewery, dairy, and vegetable processing effluents contain relatively high COD level and intermediate N concentrations. Therefore, they commonly undergo anaerobic digestion (AD) followed by Nr recovery (Ghyselbrecht et al., 2018). However, as fecal contamination can easily be avoided in these wastewaters, it is possible to microbially assimilate N, e.g. with high-rate activate sludge, microalgae or purple non-sulfur bacteria, and to subsequently use the biomass as microbial or single-cell protein as feed ingredient or as organic fertilizer (Muys et al., 2020; Spiller et al., 2020). A similar approach is the biofloc technology commercially applied for aquaculture effluents, where an in-situ produced biomass consortium of bacteria and microalgae is grown on the effluent of aquaculture, subsequently harvested and provided as proteinaceous feed within the production system (Crab et al., 2012).

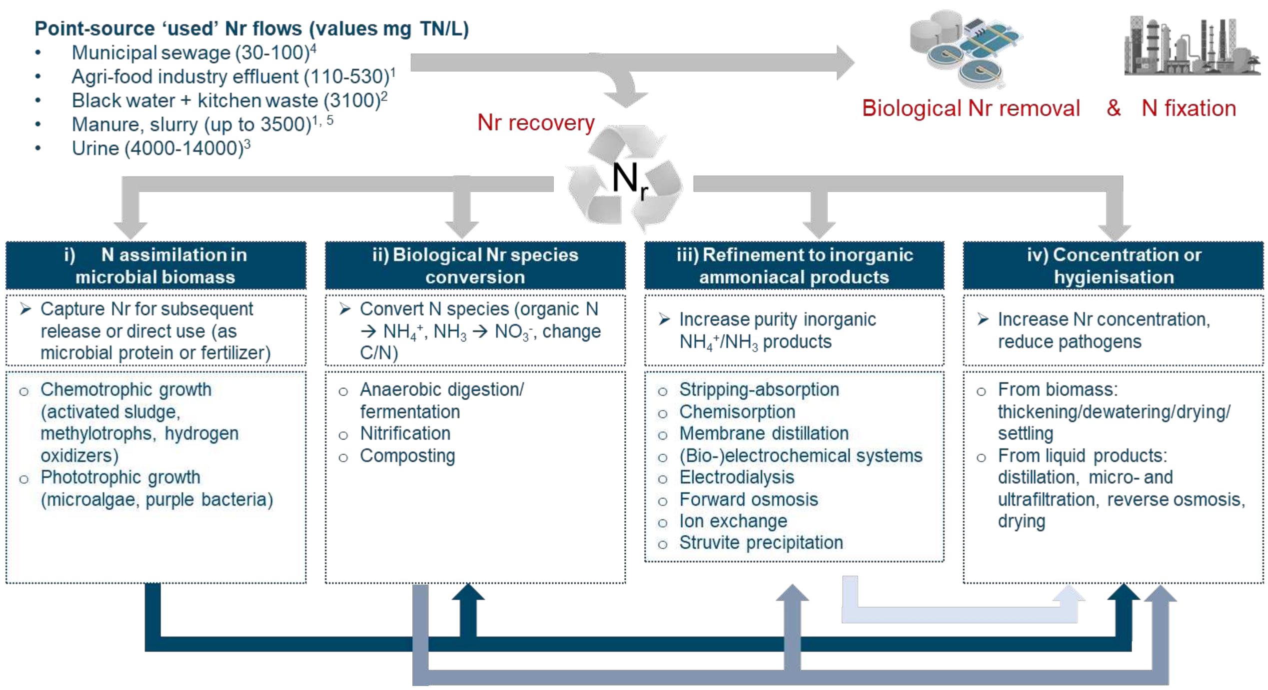

Figure 2: Overview of Nr recovery technology routes for several waste streams. For a summary of all technologies and references to each see Table S2 in the SM. TN = total nitrogen. References: 1 (Cai et al., 2013), 2 (Kujawa-Roeleveld and Zeeman, 2006), 3 (Larsen et al., 2021), 4 (Henze et al., 2008), 5 (Baldi et al., 2018)

3 Methodological approach

This review applied a structured analysis of bibliometric records for the period from 2000-2020. A two phased approach was adopted to the literature search, progressing from an explorative inductive enquiry to a deductive enquiry (Corbin and Strauss, 1990). In the explorative phase of the search, it was the objective to gain deeper insights into the papers that associated sustainability to nitrogen recovery in general. Therefore, a simple search was used with combinations of words related to sustainability assessment methods including the economic and environmental dimensions as well as words pertaining to recovery, reuse or similar (SM section 4). The initial search resulted in >900 publications found in Scopus and web of science. In an iterative process, the abstracts and body of these papers were screened for the following criteria.

- The study investigates Nr recovery processes that result in the accumulation of Nr in a final product (to be used in the agri-food value chain either as a fertilizer or feed).
- The study carries out an evidence-based evaluation with the aim to derive conclusions about the environmental or economic superiority of processes or end products
- The study benchmarks/ compares against a reference technology/product to determine superior performance of an Nr recovery technology (i.e., description of a process is insufficient).
- The study is not a review.

If studies did not comply with these criteria, they were not carried forward for further reading and coding.

Through several iterations of this process the number of papers was reduced to 38. While iteratively reading the papers a coding hierarchy emerged (SM section 5) (Corbin and Strauss, 1990). Based on the emergent coding structure, categories related to end products, substrates and technologies were implemented into the Boolean search strings that were applied in the second-round search. The combination of the search strings was carefully designed across more than 50 iterations in which the impact of the changes in each iteration was monitored. Finally, a search was carried out (SM section 4) that contained 407 papers and screened along the established criteria. Of these, 63 studies were retained for final analysis and coded along the developed coding framework (i.e., axial coding) (Corbin and Strauss, 1990). Coding was carried out using the NVivo software (QSR International Pty Ltd., 2018). The coding framework and the coding criteria can be accessed in SM, Table S3.

4 Results

4.1 Waste stream types, technologies and recovered end products

This section provides an overview of the technologies and substrates investigated by the studies included in this review, informing further analysis (4.4) and discussion (section 5.1). Of the 63 studies that met the screening criteria, nine studies investigated more than one substrate resulting in a total of 72 instances[3]. The most considered waste stream for implementation of Nr recovery technologies is municipal wastewater accounting for 29 studies (Table 2). Other frequently studied substrates are manure slurries (16) and urine (14). The sustainability of Nr recovery from source separated black water and kitchen waste is investigated in five studies and a variety of industrial wastewater types is investigated by eight studies (potato wastewater (Sigurnjak et al., 2016), agri-industry wastewater (Spanoghe et al., 2020; Vulsteke et al., 2017), aquaculture effluent (Vulsteke et al., 2017), coal wastewater (Bokun et al., 2019), air scrubber liquid (Sigurnjak et al., 2016; Vaneeckhaute et al., 2013), municipal solid waste leachate (Gu et al., 2019), not specified (Bratina et al., 2016)).

[3] Because if studies apply more than one substrate, they are counted separately.

Table 2: Technologies and substrates investigated by the reviewed studies. Total number of studies as well as distribution of technologies between the studied substrates and main technologies investigated. The column 'total studies' indicates the number of studies that apply a certain technology. This does not match the sum of the rows as a study may investigate more than one substrate. Similarly, the sum of the total of studies exceeds n=63 as studies investigate multiple technologies. To better distinguish between activated sludge system and high rate activated sludge a distinction has been made in this Table (i.e. both chemoheterotrophic metabolism). References to the studies can be found in the SM section 6.2. KW = kitchen waste, GW = grey water, BW = black water, S/L = solid liquid separation, # of studies = the number of studies investigating a specific substrate type.

			Substrates					
Category	**Technology**	**TRL**	**Sewage**	**Urine**	**Manure slurry**	**BW+GW +KW**	**other**	**Total # studies**
i. Biomass assimil.	Activated sludge	9	26	2	1	2	3	29
	Biomass Chemoorganoheterotrophi	9	2	0	1	0	1	3
	Biomass Photoheretotrophic	6-7	1	0	0	0	1	2
	Biomass Photolithoautotrophic	9	4	0	0	0	3	7
	Biomass Chemoautotrophic	6-7	1	0	2	0	0	2
ii. N convers.	Anaerobic Digestion	9	24	4	14	5	3	43
	Composting	9	3	0	2	3	0	8
	Fermentataion	9	2	0	1	0	1	3
	Nitrification for N recovery	9	0	0	0	0	0	0
iii. Refinement	Struvite precipitation	9	8	9	3	2	1	21
	Gas stripping - absorption	9	6	4	6	2	3	18
	Cation Exchange	8-9	3	1	0	0	0	4
	BES 'MFC-MEC'	4-5	1	4	0	0	0	4
	Membrane distillation	4-5	1	1	2	0	0	3
	Forward Osmosis	6-7	0	1	0	0	0	1
	Transmembrane Chemisorption	6-7	0	0	0	0	0	0
	Electrodialysis	6-7	0	0	0	0	0	0
iv. Concentration	Dewatering; S/L	9	15	2	10	4	4	30
	Drying	9	8	1	2	0	3	12
	Reverse Osmosis	9	1	2	1	2	0	4
	Evaporation	9	1	2	0	0	1	3
	Ultra Filtration	9	0	0	3	0	0	3
	Storage urine	9	1	1	0	1	0	1
	Hygienisation biosolids	9	0	0	1	0	0	1
	# technologies (Total is 24)		18	13	14	8	11	
	# studies investigating per substrate		29	14	16	5	8	72

Within the four classes of technology routes for Nr recovery introduced (Figure 2), it becomes evident that activated sludge (AS) (29) for assimilation of biomass (i), AD (43) for biological N species conversion (ii) and dewatering or solid/liquid (S/L) separation (30) for the concentration step (iv) are the most frequently used technologies (Table 2). Refinement technologies (iii) on the other hand are dominated by struvite precipitation (21) and gas stripping and absorption (18). The relevance of AD for the recovery of Nr from liquid streams has been described in literature before as a precursor for the application of refinement technologies or as a process that is followed by the treatment of the solids contained in the digestate (Acosta and De Vrieze, 2018). Both routes are also prevalent in this review as AD is 24 times the precursor for a refinement step or digestate treatment through composting of dewatered sludge (often with other substrates) (8) (Bratina et al., 2016; Johansson et al., 2008; Prado et al., 2020), drying (7) (Bolzonella et al., 2018), thickening/dewatering (8) and hygienisation (1) (Sigurnjak et al., 2017). The importance of the AS process is mainly a result of its dominance in municipal sewage treatment (26/29) as a pretreatment process to AD, and often followed by sludge treatment through dewatering and reuse. The Nr contained in the AS effluent may also undergo recovery for example through autotrophic algae-biomass production (Fang et al., 2016). Only three exceptions to the AS route for sewage are Pretel et al. (2015) and Xu et al. (2020) who applied an anaerobic membrane bioreactor after pre-treatment of municipal wastewater and Arashiro et al. (2018) who applied a high-rate algae pond on settled wastewater.

For this review, the category refinement to inorganic ammoniacal products is of key interest as here mineral fertilizer like products are generated (Table 2). Struvite precipitation (21) is combined with AD, except for the cases that treat urine, including Volpin et al. (2019) who precipitated struvite in the retentate of a FO urine treatment system and Igos et al. (2017) who combined struvite precipitation with microbial electrocatalysis (others de Faria et al., 2015; Ishii and Boyer, 2015; Landry and Boyer, 2016; Maurer et al., 2003). The gas stripping and absorption (18) technology is always applied after AD except when applied to urine or a few specific substrates and cases outlined below (e.g., coal gasification wastewater, regeneration of zeolite, sludge drying). The final product of gas stripping and absorption is ammonium sulfate (18), but in three cases ammonium sulfate is also obtained from membrane distillation (Dube et al., 2016; He et al., 2020; van der Hoek et al., 2018), in two other cases from bio-electrochemical systems (Igos et al., 2017; van der Hoek et al., 2018)[4] and in one case from the regeneration of an ion exchange resin with sulfuric acid (Kavvada et al., 2017). Four alternatives to ammonium sulfate are investigated: (i) ammonium acetate and (ii) ammonium citrate in a study by Jamaludin et al. (2018) that explores alternative scrubbing agents, (iii) aqueous ammonia from coal gasification wastewater in the study of Bokun et al. (2019), (iv) ammonium chloride from zeolite regeneration liquids (Lin et al., 2016), ammonia absorption in boric acid (Kuntke et al., 2012), or in an experimental setup on sewage drying off gases with phosphoric acid (Deviatkin et al., 2019). The remaining refinement technologies, including ion exchange, membrane distillation, bio-electrochemical systems and forward osmosis are of similar recurrence of between 1 to 4 studies (Table 2).

[4] Rodrigues 2015 makes use of bio-electrochemical system but does not define a product.

In addition to AS, several processes for Nr recovery through biomass assimilation (i) have been investigated. In nine cases, autotrophic biomass (seven microalgae -Photolithoautotrophic-; two methylotrophs or hydrogen oxidizing bacteria) production has been applied. The sustainability of heterotrophic biomass production, through high-rate AS processes, has been assessed three times (Table 2). Of these, biomass production for use as a fertilizer was studied in 6 cases. Specifically, Spanoghe et al. (2020) investigated the production of three microbial fertilizers (photolithoautotrophic, aerobic-heterotrophic and anaerobic photoheterotrophic biomass). Similarly, de Souza et al. (2019) applied an LCA to the production of settled algae biomass as a fertilizer on primary (i.e., screening and settling) municipal effluent, while Fang et al. (2016) produced algae on AS wastewater treatment plant effluent for fertigation. In one case, algae produced on sewage treatment plant effluent was anaerobically digested and digestate used as a fertilizer (Munasinghe-Arachchige et al., 2020). The concept of recovering Nr in biomass and use as an animal feed was studied in four instances, by Matassa et al. (2020) and Verbeeck et al. (2020) who investigated the production of biomass on methane, hydrogen, carbon monoxide and syngas (Matassa only); by Alloul et al. (2018) who proposed the valorization of chemical oxygen demand and nutrients contained in the wastewater as a feed protein source; and similarly, by Vulsteke et al. (2017) who suggested utilizing a consortium of microalgae flocs grown on aquaculture water as a shrimp feed.

Finally, it can be observed that the majority of technologies investigated is of a technology readiness level (TRL) that can be associated to full scale production (TRL 8-9). However, the estimation of the TRL per unit process (SM Table S2) does not always adequately reflect the state of the art of the technology for Nr recovery. Studies mainly explore novel combinations of existing technologies or their application in a novel context. For example, Volpin et al. (2019) proposed a novel combination of struvite precipitation with forward osmosis, achieved through a reverse flux of Mg^{2+} from the draw solution. Kjerstadius et al. (2015) investigated a wide range of source separated sanitation concepts all of which use TRL 9 technologies. However, source separation is only implemented in a hand full of site in the EU therefore leaving further options for performance improvements and hence reduction of environmental impacts and costs (Bisschops et al., 2019). Similarly, a recent survey shows that the frequently studied ammonia stripping and absorption is EU wide only implemented 8 times at commercial or pilot scale for sewage, manure and urine treatment (STOWA, 2021). In section 5.1, the implications of the difference between technology readiness will be discussed in the context of technology learning.

4.2 Benchmarking approaches

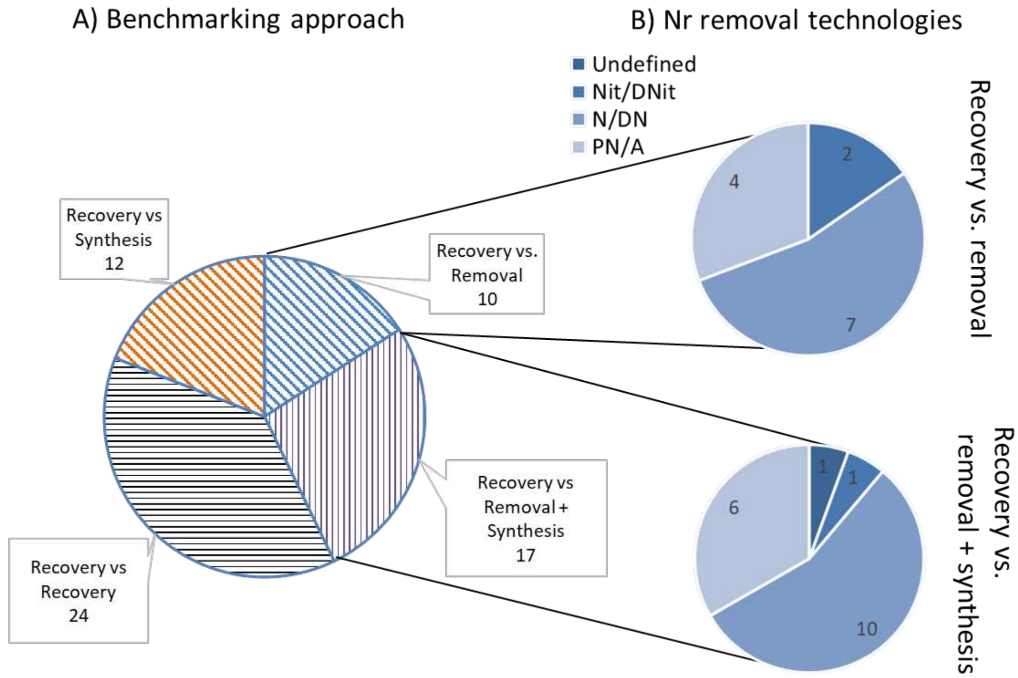

Figure 3: A. Frequency distribution of benchmarking approaches. Numbers show the number of studies using this benchmarking approach. B. Nr removal technologies for either recovery vs. removal + synthesis or recovery vs. removal. Numbers show the number of studies using a certain Nr removal technology. Numbers exceed those of figure a. since double counting is possible. Nit/DNit = Nitritation/Denitritation, N/DN = Nitrification/Denitrification, PN/A = Partial Nitritation/Anammox

To understand how reviewed articles investigated Nr recovery compared to other Nr recovery technologies, or Nr removal and/or Nr synthesis they were classified into (a method of classification is provided in SM Table S4): Nr recovery vs. Nr recovery, Nr recovery vs. Nr synthesis, Nr recovery vs. Nr removal, and Nr recovery vs. Nr removal + Nr synthesis (Figure 1B). The most frequently compared scenario is Nr recovery vs. Nr recovery with 24 of 63 studies (Figure 3). These are studies that make comparisons to evaluate the performance of recovery technologies between one another and therefore do not consider the alternative of Nr removal and Nr synthesis (i.e. indirect reuse). However, ten of these 24 studies account for the impact of Nr synthesis production only, mainly through the application of the LCA system expansion methodology. This is a method in which the impact of Nr synthesized via HB is subtracted from the recovered product (also referred to as ‘substitution’ or ‘off-set’), as the recovery is avoiding the production of HB Nr[5]. Therefore, it can be argued that these studies only partially compare the direct Nr reuse with the indirect reuse pathway.

Another 12 of 63 studies benchmarked Nr recovery vs. Nr synthesis. In these studies, a clear distinction between recovered Nr and synthesized Nr is made, instead of using the system expansion approach (de Souza et al., 2019; Dube et al., 2016). In total, 27 of 63 studies investigated Nr recovery vs. Nr removal (10) or Nr recovery vs. Nr removal + Nr synthesis (17 – of which 11 studies used the system expansion approach see SM section 6.1). Therefore, it can be argued that only 17 of the 63 studies carried out a comparison of the full set of options for Nr reuse as sketched in Figure 1.

Among the 27 studies (30 instances) that are including Nr removal in their comparison, 17 studies investigate Nr removal using N/DN, 10 studies PN/A and only three Nit/DNit (Figure 3B). Of the PN/A technologies, all considered scenarios focused on concentrated Nr streams such as side streams of WWTP (e.g., dewatering liquor) (Lin et al., 2016; van der Hoek et al., 2018), manure (De Vrieze et al., 2019; De Vrieze et al., 2016; Menkveld and Broeders, 2018) or urine (de Faria et al., 2015; Maurer et al., 2003). It can further be observed that Nr removal is in relative terms more frequently investigated for urine treatment (11/14), than for sewage (14/30) and especially for manure treatment (8/16) (SM Table S6). The dominant Nr removal technology used as a benchmark is therefore the established N/DN, while novel technologies such as PN/A and Nit/DNit are less frequently used.

4.3 System boundaries, zero-burden approach and functional unit

The system boundaries employed by sustainability assessment studies provide an indication whether the assessment is systemic (i.e. cradle-to-use and application) or more narrowly focused on products without the use phase. In the reviewed studies, cradle-to-gate evaluations dominate, with 42 of the 63 studies (or 47 of the 72 studied substrates - Figure 4) using this system boundary. Another 17 studies (21 of the studied substrates) perform cradle-to-use assessments, which is extending the system boundaries to field application or other uses of the recovered Nr products. Gate-to-gate boundaries are applied in four studies. Therefore, an approach that includes the reuse of recovered material is considered in less than half of the cases. However, in relative terms, especially the studies focused on manure (6/16 studies) and blackwater+ grey water + kitchen waste (3/5 studies) adopt system boundaries that extend to the use of fertilizer or other products (Figure 4). In the case of manure, this could be explained by the fact that animal

[5] One study (Hellström et al. (2008)) applied a compensation approach in which Nr required by agriculture that is not supplied through recovery is added to the impacts.

husbandry and crop production operate in the agricultural socio-technical systems and hence research with this focus may be more inclined to extend boundaries to application and use of recovered Nr. Whatever the reasons, ignoring the agricultural phase leads to the omission of the economic costs and environmental impacts related to post-treatment, storage, transport, and field application (Ishii and Boyer, 2015).

Of the 63 studies, 34 used the LCA methodology to quantify the potential environmental impacts of different Nr recovery technologies. All these LCA studies applied a zero-burden approach or did not define otherwise. When applying a zero-burden approach, the impact of upstream processes for the generation of the waste streams is not accounted for (Sfez et al., 2019). This implies that the impact of the waste stream production, from which Nr is recovered, is excluded from the analysis. Therefore, the application of the zero-burden approach reduces environmental impacts when compared to Nr products generated from primary inputs such as HB (section 5.3.2 for further discussion).

The reviewed studies applied a large variety of functional units (considering LCA studies only as they define functional units), including: m^3 of wastewater treated, m^3 of urine treated, treatment of 1 ton sludge, ton of manure processed, etc. Of these, most studies (28) take an input perspective by defining a reference flow, as also indicated by Lam et al. (2020). The output or product perspective is less frequently adopted (8). Consequently, Nr recovery is mainly considered in the context of waste treatment rather than Nr production. An assessment on how these differences affect the results is not possible with the present data.

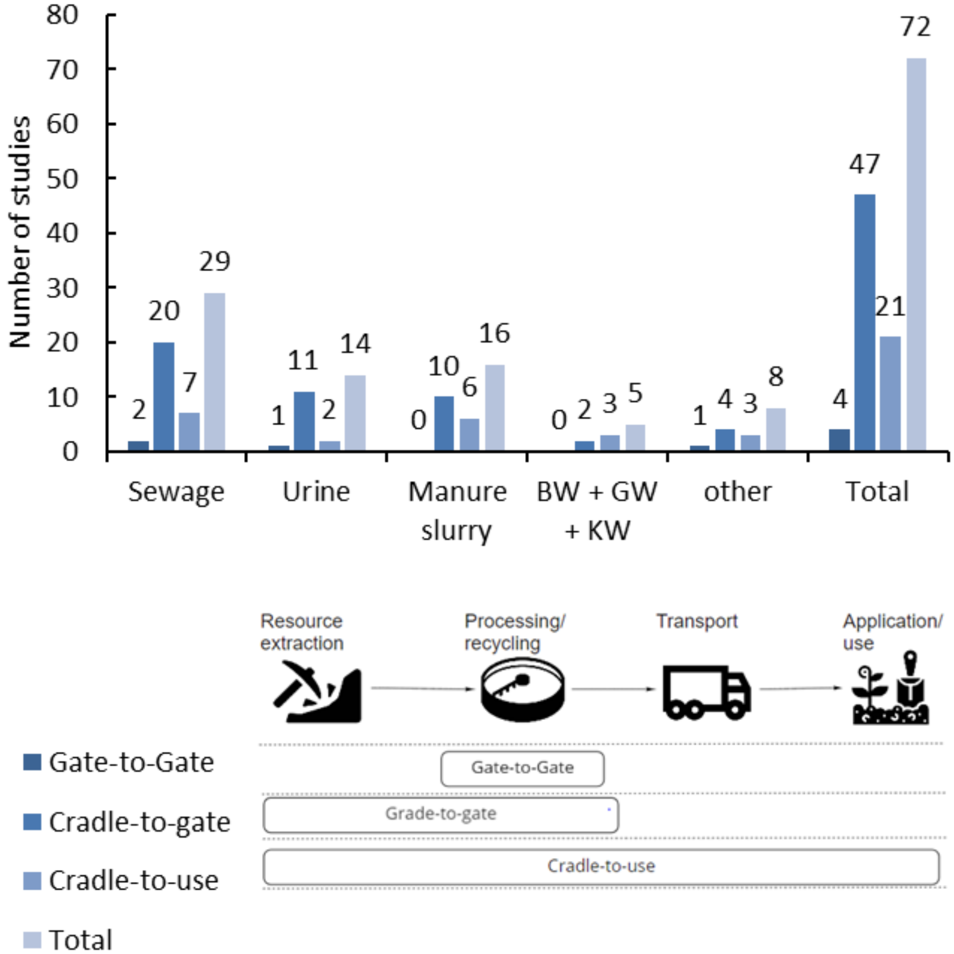

Figure 4: Frequency distribution of system boundaries applied in the studies. The number per substrate indicate individual studies that applied a specific system boundary. As there are studies investigating more than one substrate the sum of studies in the category 'Total' is exceeding n=63, totaling 72 – see also Table 2.

4.4 Conclusions of the studies regarding the sustainability of Nr recovery

Considering only the studies that compare Nr recovery vs. Nr removal, Nr recovery vs. synthesis or Nr recovery vs. Nr removal + Nr synthesis (n=39), it can be found that there is evidence for the sustainability of Nr recovery, because only 4 studies indicated a worse performance. All four studies arrived at this conclusion based on economic indicators (Figure 5). Bridle and Skrypski-Mantele (2000) assessed sludge reuse options and concluded that land application of digestate is a cheaper alternative than Nr recovery through thermal drying of raw or digested sludge. De Vrieze et al. (2019) found that all Nr and combined Nr with phosphorus recovery systems have higher costs than revenues, and especially that refinement technologies have higher operational and capital expenditure that cannot be compensated for by increased revenues. However, they also conclude that the economic viability of refinement options depends on the desired quality of end products. Similarly, Hermassi et al. (2018) suggested that the recovery of Nr using natural zeolites derived from fly ash is more costly than the indirect Nr reuse via the combination of HB and Nr removal via PN/A. Finally, Tao et al. (2019) came to the partial conclusion that at current market value, struvite production is not cost effective; though they also concluded that Nr recovery through stripping/absorption is cost effective in their case.

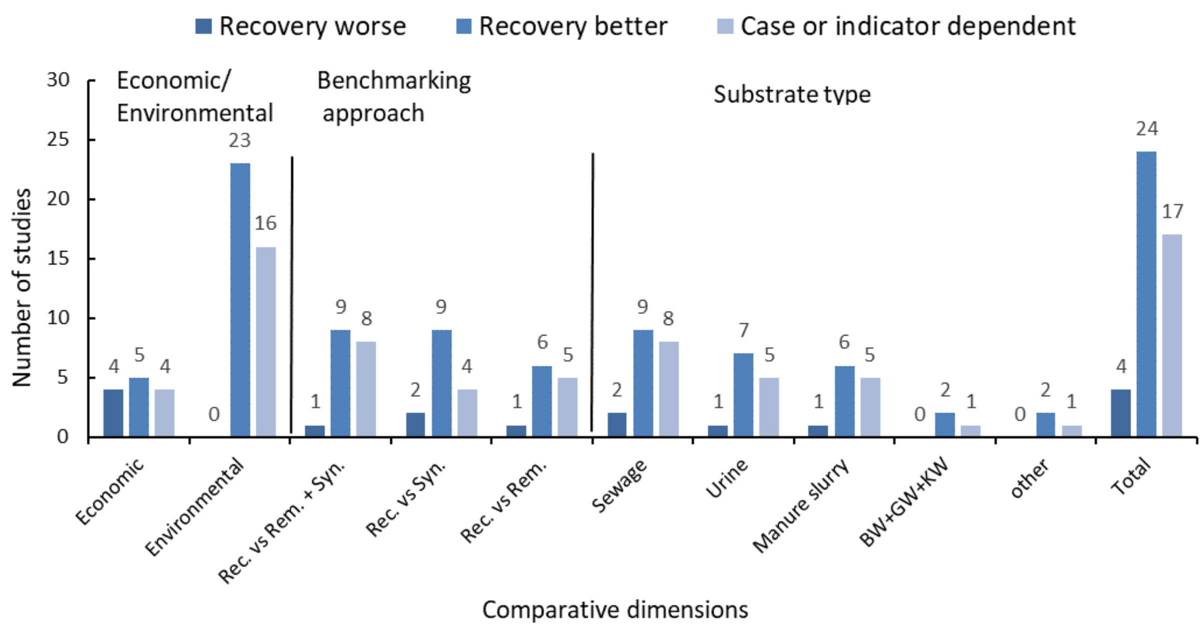

Figure 5: Conclusions of studies distinguished by the economic/ environmental, benchmarking approach and substrate type. Total number of studies deviates from figure Table 2 as, firstly the basis for this analysis excludes studies that only investigated recovery vs. recovery (n=39) and because studies are double counted when they have different conclusions for sustainability dimensions or when they have different substrates (references in SM Table S7-9). Abbreviations: Rec = recovery, Rem = removal, Syn = synthesis, BW = black water, GW = grey water, KW = kitchen waste

However, the conclusion that Nr recovery is more sustainable compared to Nr removal or Nr removal + synthesis is not straightforward. Although most studies concluded that Nr recovery is a sustainable alternative (24/39 – in total, 5 for economic and 23 for environmental aspect - Figure 5), drawing a conclusion on the sustainability of Nr recovery is challenging due to the multi-dimensionality of sustainability (i.e., different sustainability domains, different indicators, and different TRL). A substantial number of studies (17/39 in total) also reported case specific conclusions, including the fact that technologies performed better or worse for certain impact dimensions (Igos et al., 2017; Lin et al., 2016), that with further process optimization conclusions may change (de Souza et al., 2019; He et al., 2020), or that conclusions are dependent on the scale of the installation (De Vrieze et al., 2016). For illustration purposes, de Souza et al. (2019) showed that algae based organic fertilizer has an inferior environmental performance for three (climate change, particulate matter formation, freshwater eutrophication) out of the five environmental impact categories studied (terrestrial acidification, freshwater ecotoxicity). However, they also demonstrated that when other energy sources are used or other influents are selected, algae production and use as a fertilizer can become the preferable choice. Another example comes from Kavvada et al. (2017), who compared urine Nr recovery using ion exchange to Nr removal via N/DN in municipal sewage. Initially, their findings suggest superiority of Nr recovery from urine, but they also find that further development of the PN/A processes can reduce CO_2-eq. emissions to levels as found in their urine source separation and treatment system (~5-7 kg CO_2-eq./m^3 urine).

Furthermore, the outcomes of the reviewed studies do not indicate a systematic relationship between the economic and/or environmental performances, benchmarking approaches, substrate types, recovery technologies, or the system boundaries applied (Figure 5, and SM section 6.2). The difficulty of eliciting causal relationships between conclusion of reviewed studies was also reported by the review of Lam et al. (2020); who observed that most studies suggested positive environmental outcomes from wastewater-based nutrient recycling for agricultural land application, especially when chemical inputs are minimized and source separation of human excreta is applied.

5 Discussion

5.1 Sustainability of Nr recovery in the context of TRL

This review indicates that there is a case for the sustainability of Nr recovery from liquid waste streams because most of the analyzed studies indicate a superior performance of Nr recovery (24) or not an outright worse performance (17). However, 46 of the 63 studies do not account for the combination of Nr recovery, Nr synthesis, and Nr removal. This implies that these studies lack the systems perspective on Nr recovery to conclude about its sustainability, because they are not considering the possibility of Nr recycling via the indirect reuse route (Figure 1A).

Another observation is that in many cases concrete conclusions about sustainability are not easy to make as sustainability is a multi-dimensional concept. Therefore, it is difficult to balance different environmental indicators or the environmental and economic dimensions to arrive at a better or worse conclusion. What is also noteworthy is that, because scientific studies focus on low TRL technologies or novel combinations of established technologies, the use of different prospective scenarios is applied (section 4.1 & 4.4). In several cases, the assumptions for these scenarios e.g. related to the energy sources, type of waste stream, market prices, are modified resulting in changes to the initial findings (de Souza et al., 2019; Deviatkin et al., 2019). The aim of these scenario analyses is to estimate the performance of the novel technologies at full scale, when further optimized or when the operational context has changed (e.g., new energy sources). In literature, approaches that aim to estimate technological performance in the future can be classified into technological development and technological learning (Buyle et al., 2019). Technological development seeks to estimate the environmental and economic performance improvements of low TRL technologies to market readiness. Market ready technologies further evolve through technological learning, or a process of optimization (Thomassen et al., 2020). Methods to estimate these changes in technology include scaling factors, proxy technologies, learning curves and participatory methods amongst other (Buyle et al., 2019). In the present assessment of 63 studies, no study explicitly referred to the use of technological development assessment or technological learning or to any of the methods commonly used in prospective assessments. The use of expert judgement and literature values was the dominant mechanism for the development of evaluation scenarios and hence future technology performance. Therefore, it is recommended to explore how to integrate technology learning and technological development in environmental and economic assessment using the methodologies and recommendations suggested by Buyle et al. (2019) and Thomassen et al. (2020).

Another challenge for environmental and economic assessment will be to provide an on-par comparison between technologies for Nr recovery, Nr removal and Nr synthesis. Specifically, 27 studies compare Nr recovery with Nr removal technologies, but the majority (n= 17) chose to compare these technologies with conventional Nr removal using N/DN (section 4.2), thereby not considering the potential gains from innovations in removal technologies, such as PN/A (10 studies) or Nit/DNit (3 studies). For Nr removal technologies, it can therefore be suggested that future studies should pay more attention to match the TRL and learning stage of the Nr recovery technologies with that of the Nr removal technology. When studying emerging Nr recovery technologies, it can further be vital to account for advances in Nr synthesis (next section). Of course, accounting for emerging technologies for Nr removal and synthesis makes studies more challenging to conduct and implement due to the low availability of data about these technologies.

5.2 Nr recovery in the new Nr economy

It has been suggested that about 48% of the world’s population since 1908, when the HB process was patented, depended on the input of synthetic Nr fertilizers (Erisman et al., 2008). About 153 Tg Nr (62% derived from HB) are used in the agricultural system globally (Scheer et al., 2020). Of this, an estimated

43-48% is not accessible for recovery as it is ‘lost’ in diffuse gaseous emissions and runoff from agriculture (Matassa et al., 2015; Sutton et al., 2013). While exact numbers are subject to uncertainty, it is undebatable that HB Nr synthesis is essential to enable adequate nutrition for a global community, which is reflected in estimates by the UN that, at best, Nr production will be stagnant until the year 2050 (Sutton et al., 2013). The question is therefore not if industrial Nr synthesis is necessary, but how anthropogenic Nr production will be adapted to meet the sustainability challenges of the 21st century, and what the role of Nr recovery will be within such a new ‘Nr economy’.

Modern HB processes emit 2-3 kg CO_2-eq./ton N-NH_3. By producing so called blue ammonia through capturing and storing of CO_2, emissions can be reduced to < 0.6-0.73 kg CO_2-eq./ton N-NH_3 (Wang et al., 2021). However, current HB production remains dependent on fossil resources as an energy and H_2 source. Therefore, interest in renewable energy based or green ammonia is increasing. This is particularly interesting as a the share of renewables in the energy mix will increase in the future, raising concerns about intermitted utilization of excess energy (Macfarlane et al., 2020). Ammonia has been proposed as suitable energy carrier in this context, as it is a fuel with a higher volumetric energy density than H_2 and a high liquefaction temperature enabling easier storage (NH_3 lower heating value 11.2 MJ/L, H_2 2.46 MJ/L, boiling point at atmospheric conditions NH_3 = 33.3°C, H_2 = 252.9°C (Aziz et al., 2020; IEA, 2021). Furthermore, it is a chemical of versatile use in a range of products including pharmaceuticals and cooling systems (Smith et al., 2020; Zamfirescu and Dincer, 2008). Wang et al. (2021) demonstrated that electrolysis of water and the direct electrical synthesis of NH_3 can realize similar CO_2 emissions as the HB combined with carbon capture and storage at electricity generation emissions of around 50 gCO_2.-eq./kWh$_{el}$; or, when compared to current HB installations without carbon capture, at < 180 gCO_2.-eq./kWh$_{el}$ (for comparison: France 2021 = 81 gCO_2.-eq./kWh$_{el}$, wind power = 16 g CO_2-eq/ kWh$_{el}$ source (ecoinvent, 2021)). Wang et al. (2021) further calculated that this technology reaches cost parity at an electricity price of 0.02 €/kWh. Bicer et al. (2016) evaluated the use of renewable energy for H_2 feedstock production via electrolysis and provision of process energy demand in combination with HB. They show that CO_2 emission can be reduced by ~75% (down to 0.46 kg CO_2-eq./kg N) when using hydropower. Similarly, Matzen et al. (2015) concludes that NH_3 production from wind energy via water electrolysis and HB can be attractive from the economic and environmental perspectives. Further processing of NH_3 to urea, the world’s most used N fertilizers (~50% market share (Fertilizer Europe, 2021)), has been shown to be technically feasible at small scale and low CO_2-eq. emissions (Driver et al., 2019). Technology scenarios by the IEA (2021) suggest that CO_2 emissions of Nr synthesis may become less relevant in the next decades, when accounting for technological advances in N fixation, carbon capture and storge in combination with a transition to renewable energy. This finding is of relevance for Nr recovery as it may make the indirect Nr reuse route (Figure 1) more viable and potentially changing the conclusions arrived at by several of the reviewed studies.

Furthermore, green ammonia facilities are likely to be of a scale closer to that of Nr recovery technologies. To date, HB plants have capacities of about 2000-3000 ton NH_3/day (Wang et al., 2021). Production of NH_3 from renewable energy sources is suggested to fall within the range of 10-100s ton NH_3/day (Wang et al., 2021). Estimates for Nr recovery technologies are in the range of 0.1 – 10 ton NH_3/day[6]. Therefore, a situation may emerge where regionally directly reused Nr (i.e. recovered) and indirectly reused Nr (i.e., fixed form N_2) are available. Indeed, technologies such as 'N_2-applied' (https://n2applied.com/), where manure is enriched with atmospheric Nr through plasma technology demonstrate that the boundaries between the direct and indirect Nr reuse begin to blur (Figure 1). Scenarios for the future ammonia economy of the IEA (2021) suggest that Nr recovery and Nr synthesis must be complements, because utilization of 'waste nitrogen' may increase the nitrogen use efficiency. However, beyond this, the IEA (2021) roadmap does not detail the role of Nr reuse in the new Nr economy. It could be plausible that the new Nr economy comprises a variety of fit-for-purpose technologies at different scale for 'direct' and 'indirect' Nr reuse. This would resemble the vision for a renewable energy society that integrates a variety of energy technologies at different scales. Given the importance of this technological progress, future sustainability assessments should take a prospective approach for assessing environmental impacts or at least they should aim for comparison of technologies at similar TRL.

5.3 Methodological considerations

5.3.1 Multi-output systems

In section 4.2, the application of the system expansion was introduced, as a method where the impact of Nr synthesized via HB is subtracted from the recovered product. The same methodology is applied for other by-products of Nr recovery technologies (SM section 6.1). Examples of this are the co-production of heat, electricity, as well as P and K fertilizers (Ishii and Boyer, 2015). Furthermore, also avoided impacts of aeration electricity demand for Nr removal in municipal wastewater treatment are accounted for (Igos et al., 2017). Similarly, it has been suggested that studies focused on Nr recovery in feed products should

[6] Assumptions: 12 g Nr/person/day, large scale wastewater treatment plant 3 million PE. Equals 36t Nr/day at 100% recovery which is not realistic. Assumption: Manure digester – 3500 gNr/ manure, daily capacity 197t/day (large scale digester in NL) 0.69 t Nr/day.

account for aspects such as protein quality, polyunsaturated fatty acids, carotenoids, and vitamins (Spiller et al., 2020). These examples highlight that Nr recovery is often paired with the generation of other useful products in multi-output systems. Therefore, when applying the system expansion method, the by-products reduce environmental impacts as avoided products or improve the economic balance through the generation of additional income. This highlights that for Nr recovery, taking a single nutrient (i.e., Nr) perspective is too narrow, but that the additional benefits should, and must, be evaluated for a comprehensive assessment of Nr recovery sustainability. Interestingly, when comparing the contribution of avoided HB Nr fertilizer on the example of CO_2-eq. emissions, it does appear that it, with exceptions, plays a minor role (e.g. Arashiro et al., 2018; de Faria et al., 2015; de Souza et al., 2019; Fang et al., 2016; Igos et al., 2017; Ishii and Boyer, 2015; Johansson et al., 2008). Contrary to this, the generation of energy or production of energy carriers (e.g. biogas) (Arashiro et al., 2018) or the reduction of energy use (Igos et al., 2017; Ishii and Boyer, 2015) may have a larger effect on the environmental impact evaluation. Furthermore, as highlighted by several researchers, it appears opportune to aim for the recovery of products that are of high value instead of simply seeking to recover Nr. For example, Alloul et al. (2018); Matassa et al. (2015) suggest utilizing the COD and the nutrients contained in liquid waste streams to produce complex outputs including microbial protein and polyhydroxyalkanoates.

5.3.2 Zero burden assumption

As observed several years ago by Pradel et al. (2016), in the present research all studies employ the “burden-free” or “zero-burden” assumption (section 4.3). Implying that they do not account for the impact of upstream processes that are responsible for the generation of the waste streams Nr is derived from. Sfez et al. (2019) argue that in a circular economy the zero-burden assumption cannot hold, as ‘wastes’ are by-products of production processes which constitute raw materials for recovery. Accordingly, Sfez et al. (2019) are proposing two methodologies to avoid the zero-burden assumption by allocating a part of the production of goods (e.g., food) to the waste stream as well as allocating a part of the burden of treatment/recovery to the production of primary goods (e.g., food). Sfez et al. (2019) applied this to phosphorus recovery from wastewater treatment showing that this will increase the environmental impact between 27-80% for their case study. Therefore, environmental impact evaluations more in line with the circular economy paradigm (i.e. no zero burden) are likely to increase the environmental impact of Nr recovery technologies as they are derived from waste streams. Processes such as the HB or Nr removal are not affected by this since they are mostly based on primary raw material inputs. This implies that conclusions of comparative studies may change in favor of indirect Nr reuse if the zero burden is not applied.

5.4 Barriers to the implementation of resource recovery

In addition to environmental and economic aspects, a number of societal variables determine whether Nr recovery can be successfully implemented. Tur-Cardona et al. (2018) found that farmers in seven EU countries prefer fertilizers in the solid form due to easier storage and lower transport and application costs compared with liquid fertilizers. It is therefore noteworthy that a large share of the reviewed studies investigate the production of liquid fertilizers (e.g. 18 gas stripping/absorption, 4 cation exchange, 3 membrane distillation, 4 reverse osmosis – Figure 1). Nutrient concentration and absence of pollutants are also identified as decisive traits for farmers' willingness to replace synthetic with biobased fertilizers (Tur-Cardona et al., 2018). Recovered ammonium sulphate (up to 9%) and ammonium nitrate (up to 20%) in liquid form have been shown to realise similar Nr concentrations as their synthetic counterparts (Sigurnjak et al., 2019). However, for other recovered fertilizers, Nr concentrations are well below those of HB fertilizers (e.g., 46% N urea), with for example struvite up to 5.6% (Muys et al., 2021), dried microbial biomass up to 8.5% (Spiller et al., 2020); or 10-fold concentrated urine up to 10% (Maurer et al., 2003). Furthermore, concerns about the variability in nutrient content and product contamination of recovered products (e.g. heavy metals, micro-pollutants) have been shown to be barriers for the adoption of recovered Nr as a replacement of synthetic Nr (Case et al., 2017).

From an economic perspective, capital investments and payback periods are perceived by farmers as the main barriers to the implementation of manure processing technologies (Hou et al., 2018). Contrary to this, Lienert and Larsen (2010) reported the acceptance of urine source separation systems and urine reuse as fertilizers to be high among the general public, whereas liability claims, lower fertilizer quality as well as reduced acceptance by consumers limit farmers' acceptance. However, the recent revision of the EU fertilizer directive may help in stimulating Nr recovery and acceptance by farmers as it sets clear guidelines for the Nr content of recovered materials (EC, 2019). To qualify as a straight mineral Nr fertilizer, concentrations of 5% and 10% Nr by mass for liquid and solid fertilizer must be achieved. For mineral compound fertilizers, this level is reduced to 1.5% (liquid) and 3% N (solid) by mass under the condition of the presence of other components e.g., 3% P_2O_5 by mass (solid). However, Article 2.g. of the Nitrates Directive (EC, 1991) still poses barriers for the utilization of directly reused Nr as it defines 'livestock manure' as waste products excreted by livestock or a mixture of litter and waste products excreted by livestock, even in processed form. This implies that in nitrate vulnerable zones not more than

170 kg Nr/ha originating from livestock can be applied. Similarly, several EU countries (e.g. BE, NL, DE) only permit the application of human excreta-based fertilizer with specific derogations, as implemented for struvite (Muys et al., 2021).

6 Conclusions and recommendations

This review investigated whether Nr recovery is economically and ecologically sustainable. The analysis shows that Nr recovery from liquid waste streams can be considered an environmentally and often also an economically sustainable alternative to Nr removal and HB Nr synthesis. Therefore, it can be concluded that Nr recovery from liquid waste streams is a sustainable paradigm. However, it is also evident that due to the multidimensional character of sustainability straightforward conclusions on the sustainability performance of Nr recovery cannot always be drawn. Further conclusions and recommendations are:

- Of the 407 articles that resulted from the literature search, 63 evaluated Nr recovery sustainability in a comparative manner, yet only 17 studies included the comparison of Nr recovery with the combination of Nr removal and Nr synthesis. This low proportion suggests that the scientific community should focus on including the Nr system perspectives (i.e. incl. of Nr removal + Nr synthesis) in their studies (section 4.2).
- Concerns about climate change will drive changes in Nr synthesis technology, resulting in a new Nr economy that uses renewable energy in ‘decentralized’ installations. Future research should aim to benchmark Nr recovery against these novel technologies as this will provide valuable insights into whether direct Nr reuse can be more sustainable compared to indirect Nr reuse (section 5.2).
- Future studies should strive for an on-par comparison of Nr recovery technologies with Nr removal and Nr synthesis technologies. Specifically, innovations and associated low TRL technologies should be compared like for like across the domains of Nr recovery, Nr removal and Nr synthesis (section 4.1 & 5.1).
- Many studies make use of prospective or forward-looking technology scenarios in their evaluations. Studies pursuing such an approach should make better use of methodologies available in technology learning and technology development literature to estimate future performance (section 5.1).
- A minority of reviewed studies investigated the full value chain of recovered products (i.e., cradle-to-use/application). Therefore, researchers should strive to adopt the most comprehensive

system boundaries, thereby avoiding the omission of potentially relevant environmental impacts and costs (section 4.3).

- The production of by-products such as energy, P and/or K fertilizer, or even more complex products such as proteins, is of importance in making Nr recovery a viable alternative to Nr removal and Nr synthesis (section 4.2 & 5.3).
- Legal aspects, recovered Nr quality, and end user acceptance still pose barriers for implementation of Nr recovery (section 5.4).

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8 CRediT authorship contribution statement

Marc Spiller: Conceptualization, Methodology, Formal analysis, Writing - original draft, Data curation, Visualization. **Michele Moretti:** Methodology, Formal analysis, Writing - original draft, Data curation. **Jolien De Paepe**: Writing - review & editing, Visualization. **Siegfried Vlaeminck**: Conceptualization, Visualization, Writing - review & editing

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