

This item is the archived peer-reviewed author-version of:

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

Reference:

Spiller Marc, Moretti Michele, De Paepe Jolien, Vlaeminck Siegfried.- Environmental and economic sustainability of the nitrogen recovery paradigm : evidence from a structured literature review
Resources, conservation and recycling - ISSN 1879-0658 - 184(2022), 106406
Full text (Publisher's DOI): <https://doi.org/10.1016/J.RESCONREC.2022.106406>
To cite this reference: <https://hdl.handle.net/10067/1888730151162165141>

1 Environmental and economic sustainability of the nitrogen recovery 2 paradigm: Evidence from a structured literature review.

3

4 Marc Spiller^{1,2}, Michele Moretti^{3,4}, Jolien De Paepe^{1,2}, Siegfried E. Vlaeminck^{1,2}

5 ¹ Research Group of Sustainable Air, Energy and Water Technology, University of Antwerp,
6 Groenenborgerlaan 171, 2020 Antwerpen, Belgium

7 ² Centre for Advanced Process Technology for Urban Resource Recovery (CAPTURE), Frieda Saeysstraat 1,
8 9052 Gent, Belgium

9 ³ Research Group of Environmental Economics, University of Antwerp, Prinsstraat 13, 2000 Antwerpen,
10 Belgium

11 ⁴ Department of Agriculture, Food and Environment, University of Pisa, Via del Borghetto 80, 50124 Pisa,
12 Italy

13 Abstract

14 Our economy drives on reactive nitrogen (Nr); while Nr emissions to the environment surpass the
15 planetary boundary. Increasingly, it is advocated to recover Nr contained in waste streams and to reuse it
16 'directly' in the agri-food chain. Alternatively, Nr in waste streams may be removed as N₂ and refixed via
17 the Haber-Bosch process in an 'indirect' reuse loop. As a systematic sustainability analysis of 'direct' Nr
18 reuse and its comparison to the 'indirect' reuse loop is lacking, this structured review aimed to analyze
19 literature determining the environmental and economic sustainability of Nr recovery technologies.
20 Bibliometric records were queried from 2000-2020 using Boolean search strings, and manual text coding.
21 In total, 63 studies were selected for the review. Results suggest that 'direct' Nr reuse using Nr recovery
22 technologies is the preferred paradigm as the majority of studies concluded that it is sustainable or that
23 it can be sustainable depending on technological assumptions and other scenario variables. Only 17
24 studies compared the 'direct' with the 'indirect' Nr reuse route, therefore a system perspective in Nr
25 recovery sustainability assessments should be more widely adopted. Furthermore, Nr reuse should also
26 be analyzed in the context of a 'new Nr economy' that relies on decentralized Nr production from
27 renewable energy. It is also recommended that on-par technology readiness level comparisons should be
28 carried out, making use of technology development and technology learning methodologies. Finally, by-
29 products of Nr recovery are important to be accounted for as they are reducing the environmental
30 burdens through avoided impacts.

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

33

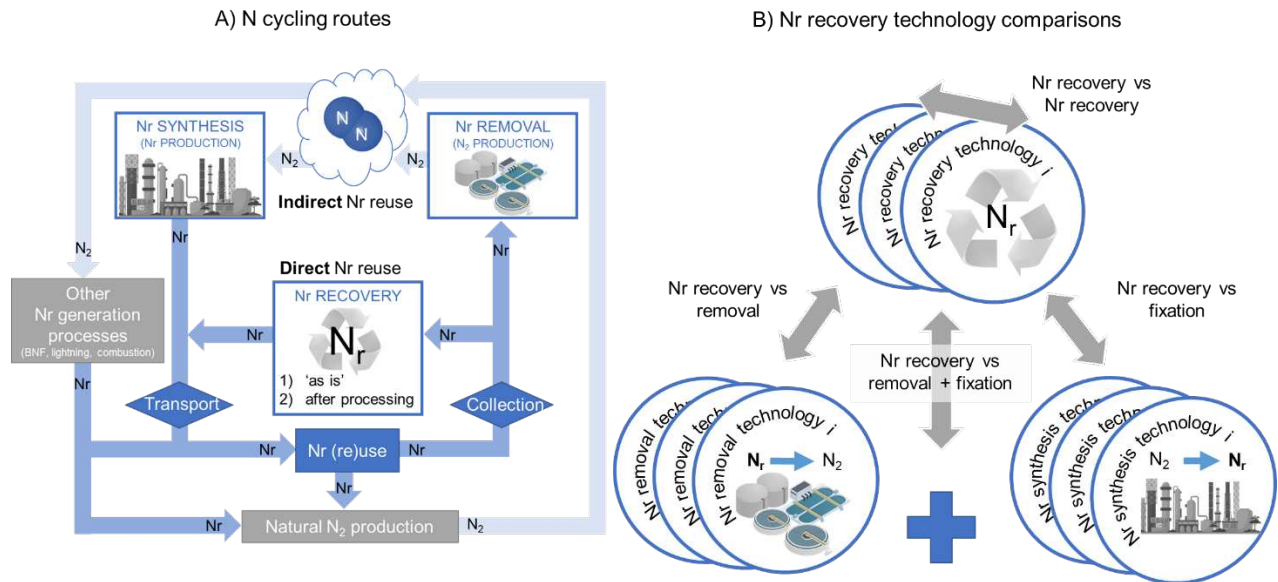
34 List of acronyms

| | | |
|----|----------------------|-------------------------------|
| 35 | AD | Anaerobic Digestion |
| 36 | AS | Activated Sludge |
| 37 | | |
| 38 | CO ₂ -eq. | CO ₂ equivalent |
| 39 | HB | Haber-Bosch |
| 40 | K | Potassium |
| 41 | LCA | Life Cycle Assessment |
| 42 | N/DN | Nitrification/Denitrification |
| 43 | Nit/DNit | Nitritation/Denitritation |
| 44 | Nr | Reactive Nitrogen |
| 45 | P | Phosphorus |
| 46 | PN/A | Partial Nitritation/Anammox |
| 47 | SM | Supplementary Material |
| 48 | TRL | Technology Readiness Level |
| 49 | Tg | Teragram |
| 50 | | |

51 1 Introduction: Nr in the Anthropocene

52 Nitrogen is an essential element for all living organisms. The biggest pool of nitrogen on our planet is the
53 atmosphere. By volume, dry air consists for 78% of dinitrogen (N₂), or an estimated 3,878,000,000
54 Teragram (Tg, million ton N)¹. Since the early 20th century, about 48% of the global population have been
55 depending on this atmospheric pool by industrially converting N₂ to ammonia (NH₃) and derived products
56 like urea and nitrate through the Haber-Bosch (HB) process (Erismann et al., 2008). On a global scale, this
57 process uses large amounts of fossil energy (1% of global energy and 2% of the global natural gas use)
58 (Cherkasov et al., 2015) and is responsible for significant CO₂-equivalent (CO₂-eq.) emissions (1.2%) (Smith
59 et al., 2020). Haber-Bosch further accounts for roughly one third (100-165 Tg N/year) of the total global
60 reactive nitrogen (Nr) generation, with the remained being derived from agricultural biological nitrogen
61 fixation (50-70 Tg N/year), fossil fuel combustions (27-33 Tg N/year) and natural biological nitrogen
62 fixation (58-128 Tg N/year) (Galloway et al., 2004; Scheer et al., 2020). The massive increase in
63 anthropogenic Nr flows and its associated emissions to the environment are profoundly altering the global
64 biogeochemical N cycle. The landmark publications of Rockström et al. (2009) and Steffen et al. (2015)
65 show that among all global environmental impacts, Nr is most severely exceeding the carrying capacity of
66 our planet (about 3 times), with agricultural fertilizer use, livestock wastes and urban wastewater having
67 major responsibility for this (Campbell et al., 2017; Fowler et al., 2013). Nr production is therefore one of
68 the most important elements through which humans are altering the global geochemical balance and
69 leaving their footprint on what can informally be referred to as the Anthropocene (National Geographic,
70 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



72

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

75 To reduce Nr pollution, different nitrogen management approaches are proposed. A preventive approach
 76 seeks to minimize Nr losses by increasing nitrogen use efficiency in the agri-food chain, and as such reduce
 77 diffuse Nr emissions (Kanter et al., 2020). A complementary curative approach proposes to recover Nr
 78 from point sources, such as seweraged wastewater, and reuse it in the agri-food chain, thereby reducing
 79 overall Nr production and its associated impacts (Arashiro et al., 2018). For such collectable point source
 80 waste streams, three main N cycling routes can be distinguished: two for 'direct reuse', and one for
 81 'indirect reuse'² with N passing over the atmosphere (Figure 1a). The first direct reuse route uses the
 82 waste stream 'as is' or after minimal processing (e.g., solid/liquid separation), typically in the form of
 83 ammoniacal and/or organic nitrogen, an approach often practiced for animal manure and livestock
 84 slurries. A second and more demanding direct reuse approach entails concentrating Nr in products after
 85 processing or refinement (i.e., removal of other elements, pollutants and/or organic matter). Direct Nr
 86 reuse 'as is' and the reuse through refinement is commonly considered as Nr 'recovery', and is adopted
 87 as a terminology throughout the document. Indirect Nr reuse is the nitrogen cycling method going over
 88 atmospheric N₂. The first step is usually termed Nr 'removal' and is based on the biological conversion of
 89 Nr to N₂. 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.

90 fixation. Through these steps, N molecules are indirectly reused by a removal-fixation-usage sequence
91 across the atmospheric N₂ cycle. The atmospheric N₂ stock is amongst others maintained by
92 nitrification/denitrification (N/DN) and anaerobic ammonium oxidation (anammox) (Burgin and Hamilton,
93 2007), both of which processes are applied in wastewater treatment for the removal of Nr.

94 Over the past decade, sustainability concerns have boosted the development of Nr recovery technologies
95 for concentrated and refined products (see supplementary material (SM) section 1 for a literature search).
96 The underlying concept of this Nr recovery paradigm is that it can provide a win-win solution as it can
97 reduce the economic and environmental costs of Nr removal and Nr synthesis. However, the sustainability
98 of Nr recovery and nutrient recovery in general is by no means self-evident. Maurer et al. (2003) used
99 operational energy requirements to evaluate scenarios of source-separated urine treatment, concluding
100 for instance that the route of Nr removal via partial nitritation/anammox (PN/A) and HB Nr synthesis
101 required less energy than Nr recovery with air stripping and absorption in H₂SO₄. Similarly, a study
102 conducted for a sewage treatment plant in Amsterdam suggested that Nr recovery results in only a limited
103 improvement in sustainability and that a range of recovery technologies have higher N₂O emissions and
104 energy demand than the combination of PN/A and HB (van der Hoek et al., 2018). The authors concluded
105 that radical changes, such as separate collection and treatment of urine, and application of several Nr
106 recovery methods in parallel would be required to substantially improve the sustainability of the
107 biogeochemical N cycle. In a recent review of life cycle assessment (LCA) studies on nutrient recovery from
108 wastewater, Lam et al. (2020) demonstrated that for sludge recycling and recovered products
109 environmental benefits only marginally outweigh environmental impacts. However, Lam et al. (2020) only
110 compared different Nr recovery options for direct Nr reuse with each other and did not systematically
111 explore differences between Nr recovery and Nr removal, or Nr recovery compared to the indirect route
112 via the combination of Nr removal and synthesis (Figure 1B). Neither did they detail the technologies for
113 the recovery technologies investigated.

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

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

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

140 2 Technologies for N cycling – An overview

141 2.1 Technologies for indirect Nr reuse: Cycling over the atmospheric N pool

142 2.1.1 N₂ production: current state and recent advances

143 In most liquid waste streams, reduced Nr is present, either as ammoniacal nitrogen and/or as organic
144 nitrogenous compounds. Biological nitrogen removal based on autotrophic nitrification, and subsequent
145 heterotrophic denitrification of the formed nitrate to N₂ is a process globally applied in wastewater
146 treatment plants (WWTP) since the 1970s (Focht and Verstraete, 1977). Alternatives for this N/DN include
147 the autotrophic/heterotrophic nitritation/denitritation (Nit/DNit) and the fully autotrophic partial
148 nitritation/anammox (PN/A). Both Nit/DNit and PN/A are termed shortcut Nr removal processes, as they
149 do not rely on the formation of nitrate but terminate the oxidation of ammoniacal nitrogen at nitrite,
150 thereby reducing costs because of savings in aeration energy and chemical oxygen demand (Table 1).

151 Shortcut Nr removal has mainly been applied to treat the warmer (>25°C), higher strength (0.5-1.5 g NH₄⁺
 152 N/L) liquid streams after anaerobic digestion with low COD concentrations (<1g COD/g N) – e.g., sludge
 153 reject water in the side stream of municipal wastewater treatment plants. Industrial applications of
 154 Nit/DNit are limited thus far. For PN/A, in 2014 100 full-scale plants were commissioned, and the number
 155 has further increased since then (Lackner et al., 2014). Concerning municipal wastewater, the application
 156 of PN/A to the lower temperatures and Nr concentrations in the water line or mainstream is of great
 157 research interest as it may result in energy positive WWTPs. This has for instance been demonstrated in
 158 two installations (Strass, Austria and Changi, Singapore) (Wett et al., 2015; Winkler and Straka, 2019). It
 159 is expected that CO₂-eq. emissions from operation can be reduced from 7.0 kg CO₂-eq/kg Nr for a N/DN
 160 process to 4.5 kg CO₂-eq/kg Nr for a mainstream PN/A at similar N₂O emission (Table 1, for calculations
 161 SM section 3 Table S1). Meanwhile, costs may nearly halve to about 2.5 euro/kg Nr removed, due to
 162 savings in energy demand and carbon source consumption (Fux and Siegrist, 2004).

163 *Table 1: Simplified comparison of three Nr removal pathways and the Haber-Bosch (HB) process. The O₂ and COD requirements*
 164 *for three Nr removal pathways are based on the stoichiometries including anabolism. a (Maurer et al., 2003), b (World Bank,*
 165 *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*
 166 *CO₂-eq emissions of 0.276 kg CO₂-eq./kWh, h own calculations based on (Maurer et al., 2003) and stoichiometric reduction of*
 167 *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-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 -eq/kg Nr] | 5.0-7.0 ^g (without-with methanol; 1% N emitted as N ₂ O) ^g | 4.8-6.0 ^g (without-with methanol; 1% N emitted as N ₂ O) ^g | 4.5 ^g (1% N emitted as N ₂ O) ^g | 2 ^c - 3.5 ^e (urea) 2-3 (NH ₃) ^{c, d} |

168

169 2.1.2 Haber-Bosch Nr synthesis

170 The HB process converts atmospheric N₂ and hydrogen gas to ammonia in a reaction that is driven by high
 171 temperatures (~650-750 K), pressure (~100-200 bar) and a metal catalyst (Cherkasov et al., 2015).

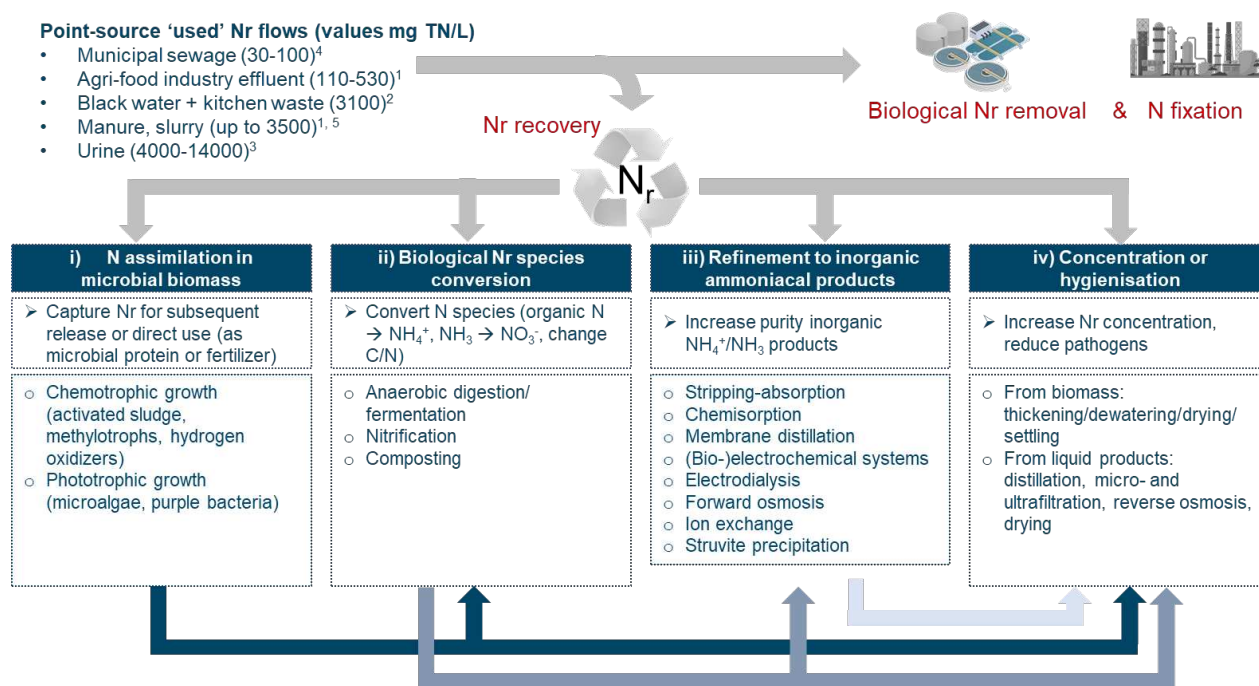
172 Hydrogen is obtained from steam reforming with major feedstock being natural gas and coal, accounting
173 for about 66% and 30% of the global consumption (International Fertilizer Organisation, 2014). The most
174 efficient HB installations operate at 9.5 kWh_{primary}/kg N-NH₃, which is close to the theoretical limit of the
175 process at 7.5 kWh_{primary}/kg N-NH₃ (80% for methane as feedstock and 20% to drive the process)
176 (Cherkasov et al., 2015; Smith et al., 2020). Therefore, it has been argued that the process is highly
177 efficient and restricted to further energetic process improvements. Along with the increase in energy
178 efficiency, CO₂-eq. emissions decreased to 2-2.1 kg CO₂-eq./kg N-NH₃ (Smith et al., 2020). Factors
179 contributing to this decrease are the shift to natural gas as feedstock instead of coal (International
180 Fertilizer Organisation, 2014) and the reuse of about 50% of the CO₂ released in the process for urea
181 production (Dawson and Hilton, 2011). The efficiency and large scale (2000-3000 tons NH₃/day) of HB
182 installations result in a cost of 0.4-2 euro/kg N-urea (2020-2021). Only due to dramatic increases in energy
183 prices in 2021 costs for urea-N now approach the order of magnitude of Nr removal technologies (World
184 Bank, 2021) (Table 1).

185 **2.2 Recovery technologies for direct Nr reuse**

186 Nitrogen concentrations in wastewater are low. Even concentrated streams like undiluted urine (~4-14 g
187 Nr/L, (Larsen et al., 2021) and manure slurry (max. ~3.5 g Nr/L, (Baldi et al., 2018; Cai et al., 2013)) contain
188 maximally 1.4% Nr or less, compared to solid synthetic inorganic fertilizers such as ammonium nitrate and
189 urea containing around 35-47% Nr. Recovery approaches from liquid waste streams (incl. the solids
190 transported) are based on four groups of processes (Figure 2; SM Table S2): (i) capture, partition or
191 'accumulate' Nr from the liquid by assimilation in microbial biomass; (ii) biological Nr species conversion
192 by either mineralization, releasing NH₄⁺ from organic matter, or nitrification, converting NH₄⁺ to NO₃⁻; (iii)
193 refinement of Nr through extraction, and (iv) the concentration of Nr through water removal in order to
194 increase N concentrations, stabilize the product (i.e., drying) and to reduce transportation costs. The
195 recovery pathways used are diverse and depend on the total nitrogen concentration, the speciation of Nr
196 as total ammoniacal nitrogen or organically bound nitrogen and the level of contamination of amongst
197 others: suspended solids, organics, heavy metals and pathogens. Some Nr recovery methods are feasible
198 directly on water with low Nr concentrations, after removal of organics and suspended material. For
199 sewage, microalgae production is such an example as well as ion exchange (Arashiro et al., 2019; Huang
200 et al., 2020). Streams with higher Nr concentrations in the order of g Nr/L and few interfering compounds,
201 can directly feed into the recovery process, as is typically considered for urine. Manure slurries contain
202 relatively high Nr levels, but also suspended solids, and may undergo anaerobic treatment and/or

203 solid/liquid separation prior to Nr recovery (Baldi et al., 2018), but have also been used as a substrate for
 204 stripping/absorption either directly on the manure or after solid/liquid separation (De Vrieze et al., 2019).
 205 Agri-food industry, including potato, brewery, dairy, and vegetable processing effluents contain relatively
 206 high COD level and intermediate N concentrations. Therefore, they commonly undergo anaerobic
 207 digestion (AD) followed by Nr recovery (Ghyselbrecht et al., 2018). However, as fecal contamination can
 208 easily be avoided in these wastewaters, it is possible to microbially assimilate N, e.g. with high-rate
 209 activate sludge, microalgae or purple non-sulfur bacteria, and to subsequently use the biomass as
 210 microbial or single-cell protein as feed ingredient or as organic fertilizer (Muys et al., 2020; Spiller et al.,
 211 2020). A similar approach is the biofloc technology commercially applied for aquaculture effluents, where
 212 an in-situ produced biomass consortium of bacteria and microalgae is grown on the effluent of
 213 aquaculture, subsequently harvested and provided as proteinaceous feed within the production system
 214 (Crab et al., 2012).

215



216

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

220

221 3 Methodological approach

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

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

238 If studies did not comply with these criteria, they were not carried forward for further reading and coding.
239

240 Through several iterations of this process the number of papers was reduced to 38. While iteratively
241 reading the papers a coding hierarchy emerged (SM section 5) (Corbin and Strauss, 1990). Based on the
242 emergent coding structure, categories related to end products, substrates and technologies were
243 implemented into the Boolean search strings that were applied in the second-round search. The
244 combination of the search strings was carefully designed across more than 50 iterations in which the
245 impact of the changes in each iteration was monitored. Finally, a search was carried out (SM section 4)
246 that contained 407 papers and screened along the established criteria. Of these, 63 studies were retained
247 for final analysis and coded along the developed coding framework (i.e., axial coding) (Corbin and Strauss,
248 1990). Coding was carried out using the NVivo software (QSR International Pty Ltd., 2018). The coding
249 framework and the coding criteria can be accessed in SM, Table S3.

250 4 Results

251 4.1 Waste stream types, technologies and recovered end products

252 This section provides an overview of the technologies and substrates investigated by the studies included
253 in this review, informing further analysis (4.4) and discussion (section 5.1). Of the 63 studies that met the
254 screening criteria, nine studies investigated more than one substrate resulting in a total of 72 instances³.
255 The most considered waste stream for implementation of Nr recovery technologies is municipal
256 wastewater accounting for 29 studies (Table 2). Other frequently studied substrates are manure slurries
257 (16) and urine (14). The sustainability of Nr recovery from source separated black water and kitchen waste
258 is investigated in five studies and a variety of industrial wastewater types is investigated by eight studies
259 (potato wastewater (Sigurnjak et al., 2016), agri-industry wastewater (Spanoghe et al., 2020; Vulsteke et
260 al., 2017), aquaculture effluent (Vulsteke et al., 2017), coal wastewater (Bokun et al., 2019), air scrubber
261 liquid (Sigurnjak et al., 2016; Vaneeckhaute et al., 2013), municipal solid waste leachate (Gu et al., 2019),
262 not specified (Bratina et al., 2016)).

263

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

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

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

271
 272 Within the four classes of technology routes for Nr recovery introduced (Figure 2), it becomes evident
 273 that activated sludge (AS) (29) for assimilation of biomass (i), AD (43) for biological N species conversion
 274 (ii) and dewatering or solid/liquid (S/L) separation (30) for the concentration step (iv) are the most
 275 frequently used technologies (Table 2). Refinement technologies (iii) on the other hand are dominated by
 276 struvite precipitation (21) and gas stripping and absorption (18). The relevance of AD for the recovery of
 277 Nr from liquid streams has been described in literature before as a precursor for the application of
 278 refinement technologies or as a process that is followed by the treatment of the solids contained in the
 279 digestate (Acosta and De Vrieze, 2018). Both routes are also prevalent in this review as AD is 24 times the

280 precursor for a refinement step or digestate treatment through composting of dewatered sludge (often
281 with other substrates) (8) (Bratina et al., 2016; Johansson et al., 2008; Prado et al., 2020), drying (7)
282 (Bolzonella et al., 2018), thickening/dewatering (8) and hygienisation (1) (Sigurnjak et al., 2017). The
283 importance of the AS process is mainly a result of its dominance in municipal sewage treatment (26/29)
284 as a pretreatment process to AD, and often followed by sludge treatment through dewatering and reuse.
285 The Nr contained in the AS effluent may also undergo recovery for example through autotrophic algae-
286 biomass production (Fang et al., 2016). Only three exceptions to the AS route for sewage are Pretel et al.
287 (2015) and Xu et al. (2020) who applied an anaerobic membrane bioreactor after pre-treatment of
288 municipal wastewater and Arashiro et al. (2018) who applied a high-rate algae pond on settled
289 wastewater.

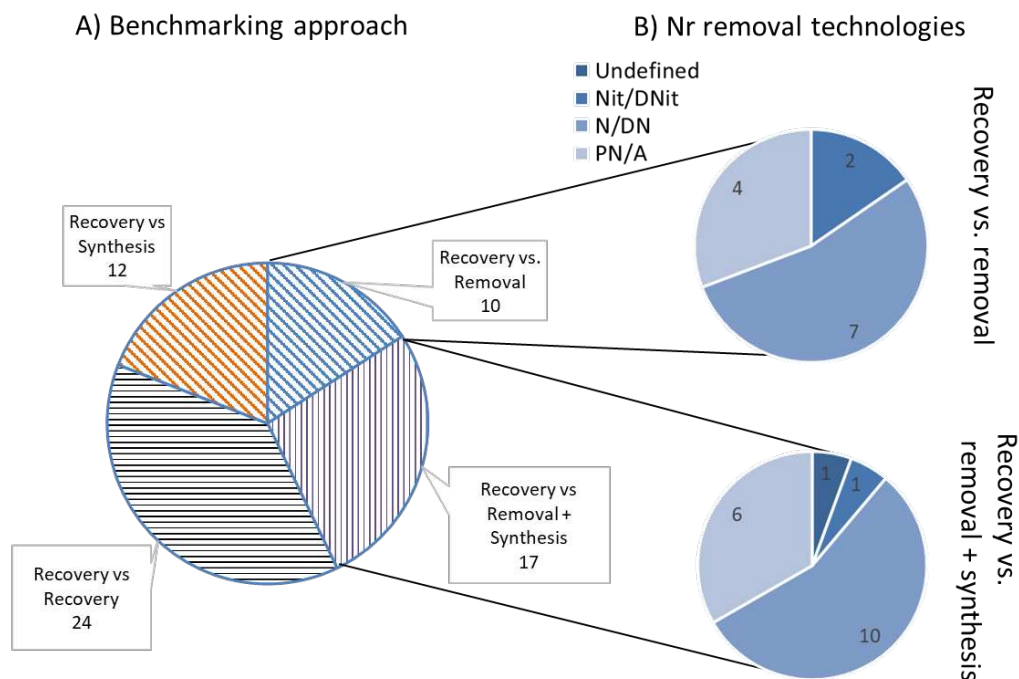
290 For this review, the category refinement to inorganic ammoniacal products is of key interest as here
291 mineral fertilizer like products are generated (Table 2). Struvite precipitation (21) is combined with AD,
292 except for the cases that treat urine, including Volpin et al. (2019) who precipitated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

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

309 In addition to AS, several processes for Nr recovery through biomass assimilation (i) have been
310 investigated. In nine cases, autotrophic biomass (seven microalgae -Photolithoautotrophic-; two
311 methylootrophs or hydrogen oxidizing bacteria) production has been applied. The sustainability of
312 heterotrophic biomass production, through high-rate AS processes, has been assessed three times (Table
313 2). Of these, biomass production for use as a fertilizer was studied in 6 cases. Specifically, Spanoghe et al.
314 (2020) investigated the production of three microbial fertilizers (photolithoautotrophic, aerobic-
315 heterotrophic and anaerobic photoheterotrophic biomass). Similarly, de Souza et al. (2019) applied an
316 LCA to the production of settled algae biomass as a fertilizer on primary (i.e., screening and settling)
317 municipal effluent, while Fang et al. (2016) produced algae on AS wastewater treatment plant effluent for
318 fertigation. In one case, algae produced on sewage treatment plant effluent was anaerobically digested
319 and digestate used as a fertilizer (Munasinghe-Arachchige et al., 2020). The concept of recovering Nr in
320 biomass and use as an animal feed was studied in four instances, by Matassa et al. (2020) and Verbeeck
321 et al. (2020) who investigated the production of biomass on methane, hydrogen, carbon monoxide and
322 syngas (Matassa only); by Alloul et al. (2018) who proposed the valorization of chemical oxygen demand
323 and nutrients contained in the wastewater as a feed protein source; and similarly, by Vulsteke et al. (2017)
324 who suggested utilizing a consortium of microalgae flocs grown on aquaculture water as a shrimp feed.

325 Finally, it can be observed that the majority of technologies investigated is of a technology readiness level
326 (TRL) that can be associated to full scale production (TRL 8-9). However, the estimation of the TRL per unit
327 process (SM Table S2) does not always adequately reflect the state of the art of the technology for Nr
328 recovery. Studies mainly explore novel combinations of existing technologies or their application in a
329 novel context. For example, Volpin et al. (2019) proposed a novel combination of struvite precipitation
330 with forward osmosis, achieved through a reverse flux of Mg^{2+} from the draw solution. Kjerstadius et al.
331 (2015) investigated a wide range of source separated sanitation concepts all of which use TRL 9
332 technologies. However, source separation is only implemented in a hand full of site in the EU therefore
333 leaving further options for performance improvements and hence reduction of environmental impacts
334 and costs (Bisschops et al., 2019). Similarly, a recent survey shows that the frequently studied ammonia
335 stripping and absorption is EU wide only implemented 8 times at commercial or pilot scale for sewage,
336 manure and urine treatment (STOWA, 2021). In section 5.1, the implications of the difference between
337 technology readiness will be discussed in the context of technology learning.

338 4.2 Benchmarking approaches



339
 340 *Figure 3: A. Frequency distribution of benchmarking approaches. Numbers show the number of studies using this benchmarking*
 341 *approach. B. Nr removal technologies for either recovery vs. removal + synthesis or recovery vs. removal. Numbers show the*
 342 *number of studies using a certain Nr removal technology. Numbers exceed those of figure a. since double counting is possible.*
 343 *Nit/DNit = Nitritation/Denitritation, N/DN = Nitrification/Denitrification, PN/A = Partial Nitritation/Anammox*

344 To understand how reviewed articles investigated Nr recovery compared to other Nr recovery
 345 technologies, or Nr removal and/or Nr synthesis they were classified into (a method of classification is
 346 provided in SM Table S4): Nr recovery vs. Nr recovery, Nr recovery vs. Nr synthesis, Nr recovery vs. Nr
 347 removal, and Nr recovery vs. Nr removal + Nr synthesis (Figure 1B). The most frequently compared
 348 scenario is Nr recovery vs. Nr recovery with 24 of 63 studies (Figure 3). These are studies that make
 349 comparisons to evaluate the performance of recovery technologies between one another and therefore
 350 do not consider the alternative of Nr removal and Nr synthesis (i.e. indirect reuse). However, ten of these
 351 24 studies account for the impact of Nr synthesis production only, mainly through the application of the
 352 LCA system expansion methodology. This is a method in which the impact of Nr synthesized via HB is
 353 subtracted from the recovered product (also referred to as ‘substitution’ or ‘off-set’), as the recovery is

354 avoiding the production of HB Nr⁵. Therefore, it can be argued that these studies only partially compare
355 the direct Nr reuse with the indirect reuse pathway.

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

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

371 4.3 System boundaries, zero-burden approach and functional unit

372 The system boundaries employed by sustainability assessment studies provide an indication whether the
373 assessment is systemic (i.e. cradle-to-use and application) or more narrowly focused on products without
374 the use phase. In the reviewed studies, cradle-to-gate evaluations dominate, with 42 of the 63 studies (or
375 47 of the 72 studied substrates - Figure 4) using this system boundary. Another 17 studies (21 of the
376 studied substrates) perform cradle-to-use assessments, which is extending the system boundaries to field
377 application or other uses of the recovered Nr products. Gate-to-gate boundaries are applied in four
378 studies. Therefore, an approach that includes the reuse of recovered material is considered in less than
379 half of the cases. However, in relative terms, especially the studies focused on manure (6/16 studies) and
380 blackwater+ grey water + kitchen waste (3/5 studies) adopt system boundaries that extend to the use of
381 fertilizer or other products (Figure 4). In the case of manure, this could be explained by the fact that animal

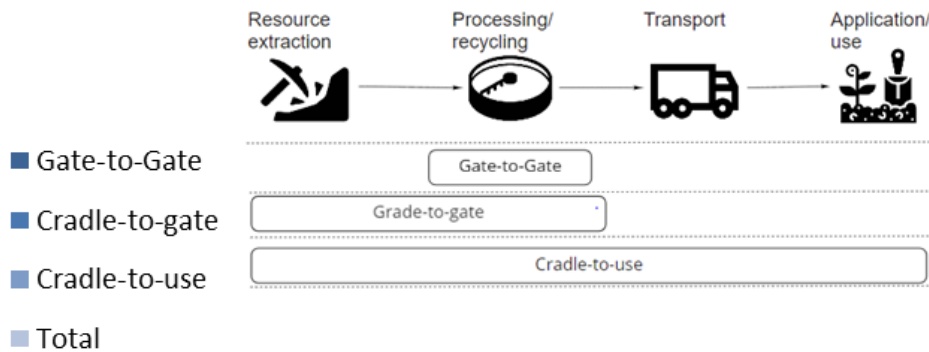
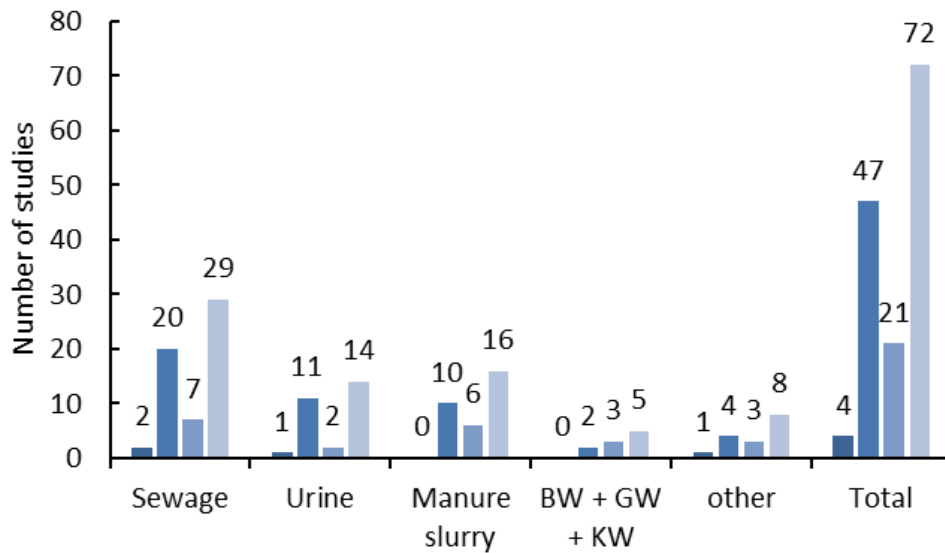
⁵ One study (Hellström et al. (2008)) applied a compensation approach in which Nr required by agriculture that is not supplied through recovery is added to the impacts.

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

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

394 The reviewed studies applied a large variety of functional units (considering LCA studies only as they
395 define functional units), including: m³ of wastewater treated, m³ of urine treated, treatment of 1 ton
396 sludge, ton of manure processed, etc. Of these, most studies (28) take an input perspective by defining a
397 reference flow, as also indicated by Lam et al. (2020). The output or product perspective is less frequently
398 adopted (8). Consequently, Nr recovery is mainly considered in the context of waste treatment rather
399 than Nr production. An assessment on how these differences affect the results is not possible with the
400 present data.

401



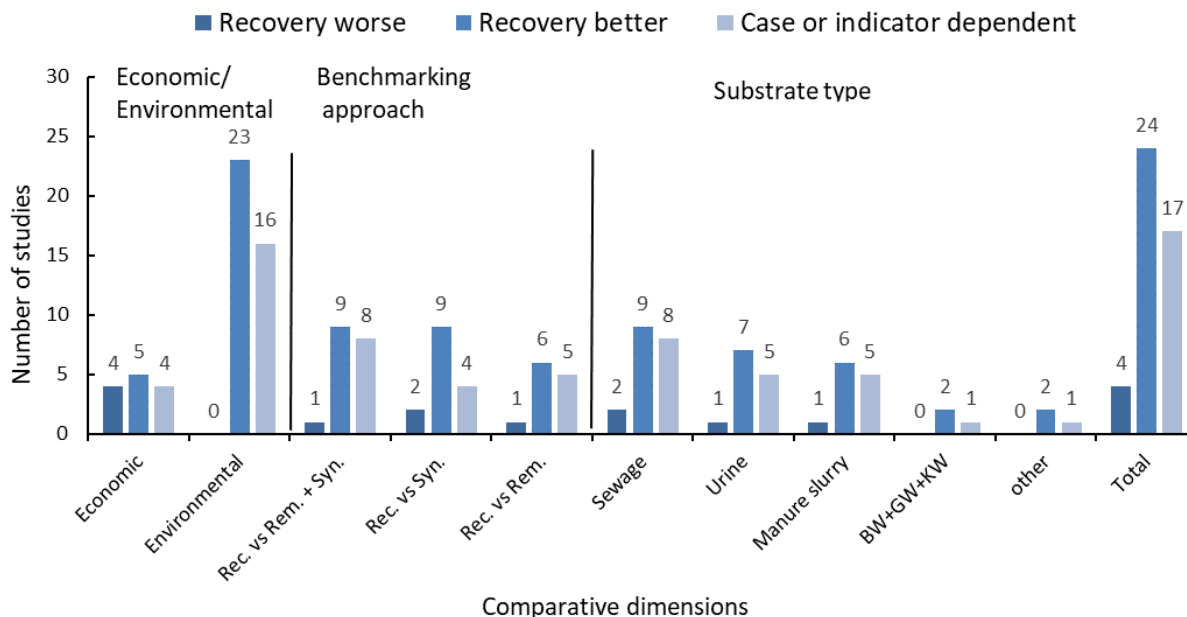
402

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

406 4.4 Conclusions of the studies regarding the sustainability of Nr recovery

407 Considering only the studies that compare Nr recovery vs. Nr removal, Nr recovery vs. synthesis or Nr
 408 recovery vs. Nr removal + Nr synthesis (n=39), it can be found that there is evidence for the sustainability
 409 of Nr recovery, because only 4 studies indicated a worse performance. All four studies arrived at this
 410 conclusion based on economic indicators (Figure 5). Bridle and Skrypski-Mantele (2000) assessed sludge
 411 reuse options and concluded that land application of digestate is a cheaper alternative than Nr recovery
 412 through thermal drying of raw or digested sludge. De Vrieze et al. (2019) found that all Nr and combined
 413 Nr with phosphorus recovery systems have higher costs than revenues, and especially that refinement
 414 technologies have higher operational and capital expenditure that cannot be compensated for by

415 increased revenues. However, they also conclude that the economic viability of refinement options
 416 depends on the desired quality of end products. Similarly, Hermassi et al. (2018) suggested that the
 417 recovery of Nr using natural zeolites derived from fly ash is more costly than the indirect Nr reuse via the
 418 combination of HB and Nr removal via PN/A. Finally, Tao et al. (2019) came to the partial conclusion that
 419 at current market value, struvite production is not cost effective; though they also concluded that Nr
 420 recovery through stripping/absorption is cost effective in their case.



421
 422 *Figure 5: Conclusions of studies distinguished by the economic/ environmental, benchmarking approach and substrate type. Total*
 423 *number of studies deviates from figure Table 2 as, firstly the basis for this analysis excludes studies that only investigated recovery*
 424 *vs. recovery (n=39) and because studies are double counted when they have different conclusions for sustainability dimensions or*
 425 *when they have different substrates (references in SM Table S7-9). Abbreviations: Rec = recovery, Rem = removal, Syn = synthesis,*
 426 *BW = black water, GW = grey water, KW = kitchen waste*

427 However, the conclusion that Nr recovery is more sustainable compared to Nr removal or Nr removal +
 428 synthesis is not straightforward. Although most studies concluded that Nr recovery is a sustainable
 429 alternative (24/39 – in total, 5 for economic and 23 for environmental aspect - Figure 5), drawing a
 430 conclusion on the sustainability of Nr recovery is challenging due to the multi-dimensionality of
 431 sustainability (i.e., different sustainability domains, different indicators, and different TRL). A substantial
 432 number of studies (17/39 in total) also reported case specific conclusions, including the fact that
 433 technologies performed better or worse for certain impact dimensions (Igos et al., 2017; Lin et al., 2016),
 434 that with further process optimization conclusions may change (de Souza et al., 2019; He et al., 2020), or
 435 that conclusions are dependent on the scale of the installation (De Vrieze et al., 2016). For illustration

436 purposes, de Souza et al. (2019) showed that algae based organic fertilizer has an inferior environmental
437 performance for three (climate change, particulate matter formation, freshwater eutrophication) out of
438 the five environmental impact categories studied (terrestrial acidification, freshwater ecotoxicity).
439 However, they also demonstrated that when other energy sources are used or other influents are
440 selected, algae production and use as a fertilizer can become the preferable choice. Another example
441 comes from Kavvada et al. (2017), who compared urine Nr recovery using ion exchange to Nr removal via
442 N/DN in municipal sewage. Initially, their findings suggest superiority of Nr recovery from urine, but they
443 also find that further development of the PN/A processes can reduce CO₂-eq. emissions to levels as found
444 in their urine source separation and treatment system (~5-7 kg CO₂-eq./m³ urine).

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

452 5 Discussion

453 5.1 Sustainability of Nr recovery in the context of TRL

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

460 Another observation is that in many cases concrete conclusions about sustainability are not easy to make
461 as sustainability is a multi-dimensional concept. Therefore, it is difficult to balance different
462 environmental indicators or the environmental and economic dimensions to arrive at a better or worse
463 conclusion. What is also noteworthy is that, because scientific studies focus on low TRL technologies or
464 novel combinations of established technologies, the use of different prospective scenarios is applied
465 (section 4.1 & 4.4). In several cases, the assumptions for these scenarios e.g. related to the energy sources,

466 type of waste stream, market prices, are modified resulting in changes to the initial findings (de Souza et
467 al., 2019; Deviatkin et al., 2019). The aim of these scenario analyses is to estimate the performance of the
468 novel technologies at full scale, when further optimized or when the operational context has changed
469 (e.g., new energy sources). In literature, approaches that aim to estimate technological performance in
470 the future can be classified into technological development and technological learning (Buyle et al., 2019).
471 Technological development seeks to estimate the environmental and economic performance
472 improvements of low TRL technologies to market readiness. Market ready technologies further evolve
473 through technological learning, or a process of optimization (Thomassen et al., 2020). Methods to
474 estimate these changes in technology include scaling factors, proxy technologies, learning curves and
475 participatory methods amongst other (Buyle et al., 2019). In the present assessment of 63 studies, no
476 study explicitly referred to the use of technological development assessment or technological learning or
477 to any of the methods commonly used in prospective assessments. The use of expert judgement and
478 literature values was the dominant mechanism for the development of evaluation scenarios and hence
479 future technology performance. Therefore, it is recommended to explore how to integrate technology
480 learning and technological development in environmental and economic assessment using the
481 methodologies and recommendations suggested by Buyle et al. (2019) and Thomassen et al. (2020).

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

493 5.2 Nr recovery in the new Nr economy

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

497 43-48% is not accessible for recovery as it is 'lost' in diffuse gaseous emissions and runoff from agriculture
498 (Matassa et al., 2015; Sutton et al., 2013). While exact numbers are subject to uncertainty, it is
499 undebatable that HB Nr synthesis is essential to enable adequate nutrition for a global community, which
500 is reflected in estimates by the UN that, at best, Nr production will be stagnant until the year 2050 (Sutton
501 et al., 2013). The question is therefore not if industrial Nr synthesis is necessary, but how anthropogenic
502 Nr production will be adapted to meet the sustainability challenges of the 21st century, and what the role
503 of Nr recovery will be within such a new 'Nr economy'.

504 Modern HB processes emit 2-3 kg CO₂-eq./ton N-NH₃. By producing so called blue ammonia through
505 capturing and storing of CO₂, emissions can be reduced to < 0.6-0.73 kg CO₂-eq./ton N-NH₃ (Wang et al.,
506 2021). However, current HB production remains dependent on fossil resources as an energy and H₂
507 source. Therefore, interest in renewable energy based or green ammonia is increasing. This is particularly
508 interesting as a the share of renewables in the energy mix will increase in the future, raising concerns
509 about intermitted utilization of excess energy (Macfarlane et al., 2020). Ammonia has been proposed as
510 suitable energy carrier in this context, as it is a fuel with a higher volumetric energy density than H₂ and a
511 high liquefaction temperature enabling easier storage (NH₃ lower heating value 11.2 MJ/L, H₂ 2.46 MJ/L,
512 boiling point at atmospheric conditions NH₃ = 33.3°C, H₂ = 252.9°C (Aziz et al., 2020; IEA, 2021).
513 Furthermore, it is a chemical of versatile use in a range of products including pharmaceuticals and cooling
514 systems (Smith et al., 2020; Zamfirescu and Dincer, 2008). Wang et al. (2021) demonstrated that
515 electrolysis of water and the direct electrical synthesis of NH₃ can realize similar CO₂ emissions as the HB
516 combined with carbon capture and storage at electricity generation emissions of around 50 gCO₂-
517 eq./kWh_{el}; or, when compared to current HB installations without carbon capture, at < 180 gCO₂-
518 eq./kWh_{el} (for comparison: France 2021 = 81 gCO₂-eq./kWh_{el}, wind power = 16 g CO₂-eq/ kWh_{el} source
519 (ecoinvent, 2021)). Wang et al. (2021) further calculated that this technology reaches cost parity at an
520 electricity price of 0.02 €/kWh. Bicer et al. (2016) evaluated the use of renewable energy for H₂ feedstock
521 production via electrolysis and provision of process energy demand in combination with HB. They show
522 that CO₂ emission can be reduced by ~75% (down to 0.46 kg CO₂-eq./kg N) when using hydropower.
523 Similarly, Matzen et al. (2015) concludes that NH₃ production from wind energy via water electrolysis and
524 HB can be attractive from the economic and environmental perspectives. Further processing of NH₃ to
525 urea, the world's most used N fertilizers (~50% market share (Fertilizer Europe, 2021)), has been shown
526 to be technically feasible at small scale and low CO₂-eq. emissions (Driver et al., 2019). Technology
527 scenarios by the IEA (2021) suggest that CO₂ emissions of Nr synthesis may become less relevant in the

528 next decades, when accounting for technological advances in N fixation, carbon capture and storage in
529 combination with a transition to renewable energy. This finding is of relevance for Nr recovery as it may
530 make the indirect Nr reuse route (Figure 1) more viable and potentially changing the conclusions arrived
531 at by several of the reviewed studies.

532 Furthermore, green ammonia facilities are likely to be of a scale closer to that of Nr recovery technologies.
533 To date, HB plants have capacities of about 2000-3000 ton NH₃/day (Wang et al., 2021). Production of NH₃
534 from renewable energy sources is suggested to fall within the range of 10-100s ton NH₃/day (Wang et al.,
535 2021). Estimates for Nr recovery technologies are in the range of 0.1 – 10 ton NH₃/day⁶. Therefore, a
536 situation may emerge where regionally directly reused Nr (i.e. recovered) and indirectly reused Nr (i.e.,
537 fixed form N₂) are available. Indeed, technologies such as ‘N₂-applied’ (<https://n2applied.com/>), where
538 manure is enriched with atmospheric Nr through plasma technology demonstrate that the boundaries
539 between the direct and indirect Nr reuse begin to blur (Figure 1). Scenarios for the future ammonia
540 economy of the IEA (2021) suggest that Nr recovery and Nr synthesis must be complements, because
541 utilization of ‘waste nitrogen’ may increase the nitrogen use efficiency. However, beyond this, the IEA
542 (2021) roadmap does not detail the role of Nr reuse in the new Nr economy. It could be plausible that the
543 new Nr economy comprises a variety of fit-for-purpose technologies at different scale for ‘direct’ and
544 ‘indirect’ Nr reuse. This would resemble the vision for a renewable energy society that integrates a variety
545 of energy technologies at different scales. Given the importance of this technological progress, future
546 sustainability assessments should take a prospective approach for assessing environmental impacts or at
547 least they should aim for comparison of technologies at similar TRL.

548 5.3 Methodological considerations

549 5.3.1 Multi-output systems

550 In section 4.2, the application of the system expansion was introduced, as a method where the impact of
551 Nr synthesized via HB is subtracted from the recovered product. The same methodology is applied for
552 other by-products of Nr recovery technologies (SM section 6.1). Examples of this are the co-production of
553 heat, electricity, as well as P and K fertilizers (Ishii and Boyer, 2015). Furthermore, also avoided impacts
554 of aeration electricity demand for Nr removal in municipal wastewater treatment are accounted for (Igos
555 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.

556 account for aspects such as protein quality, polyunsaturated fatty acids, carotenoids, and vitamins (Spiller
557 et al., 2020). These examples highlight that Nr recovery is often paired with the generation of other useful
558 products in multi-output systems. Therefore, when applying the system expansion method, the by-
559 products reduce environmental impacts as avoided products or improve the economic balance through
560 the generation of additional income. This highlights that for Nr recovery, taking a single nutrient (i.e., Nr)
561 perspective is too narrow, but that the additional benefits should, and must, be evaluated for a
562 comprehensive assessment of Nr recovery sustainability. Interestingly, when comparing the contribution
563 of avoided HB Nr fertilizer on the example of CO₂-eq. emissions, it does appear that it, with exceptions,
564 plays a minor role (e.g. Arashiro et al., 2018; de Faria et al., 2015; de Souza et al., 2019; Fang et al., 2016;
565 Igos et al., 2017; Ishii and Boyer, 2015; Johansson et al., 2008). Contrary to this, the generation of energy
566 or production of energy carriers (e.g. biogas) (Arashiro et al., 2018) or the reduction of energy use (Igos
567 et al., 2017; Ishii and Boyer, 2015) may have a larger effect on the environmental impact evaluation.
568 Furthermore, as highlighted by several researchers, it appears opportune to aim for the recovery of
569 products that are of high value instead of simply seeking to recover Nr. For example, Alloul et al. (2018);
570 Matassa et al. (2015) suggest utilizing the COD and the nutrients contained in liquid waste streams to
571 produce complex outputs including microbial protein and polyhydroxyalkanoates.

572 5.3.2 Zero burden assumption

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

586 inputs. This implies that conclusions of comparative studies may change in favor of indirect Nr reuse if the
587 zero burden is not applied.

588 5.4 Barriers to the implementation of resource recovery

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

604 From an economic perspective, capital investments and payback periods are perceived by farmers as the
605 main barriers to the implementation of manure processing technologies (Hou et al., 2018). Contrary to
606 this, Lienert and Larsen (2010) reported the acceptance of urine source separation systems and urine
607 reuse as fertilizers to be high among the general public, whereas liability claims, lower fertilizer quality as
608 well as reduced acceptance by consumers limit farmers' acceptance. However, the recent revision of the
609 EU fertilizer directive may help in stimulating Nr recovery and acceptance by farmers as it sets clear
610 guidelines for the Nr content of recovered materials (EC, 2019). To qualify as a straight mineral Nr
611 fertilizer, concentrations of 5% and 10% Nr by mass for liquid and solid fertilizer must be achieved. For
612 mineral compound fertilizers, this level is reduced to 1.5% (liquid) and 3% N (solid) by mass under the
613 condition of the presence of other components e.g., 3% P₂O₅ by mass (solid). However, Article 2.g. of the
614 Nitrates Directive (EC, 1991) still poses barriers for the utilization of directly reused Nr as it defines
615 'livestock manure' as waste products excreted by livestock or a mixture of litter and waste products
616 excreted by livestock, even in processed form. This implies that in nitrate vulnerable zones not more than

617 170 kg Nr/ha originating from livestock can be applied. Similarly, several EU countries (e.g. BE, NL, DE)
618 only permit the application of human excreta-based fertilizer with specific derogations, as implemented
619 for struvite (Muys et al., 2021).

620 6 Conclusions and recommendations

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

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

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

649 • The production of by-products such as energy, P and/or K fertilizer, or even more complex
650 products such as proteins, is of importance in making Nr recovery a viable alternative to Nr
651 removal and Nr synthesis (section 4.2 & 5.3).

652 • Legal aspects, recovered Nr quality, and end user acceptance still pose barriers for
653 implementation of Nr recovery (section 5.4).

654

655 7 Acknowledgements

656 The authors would like to thank the whole water team of the sustainable air and energy technology
657 research group for their valuable discussion and inputs to early drafts and presentations. Michele Moretti
658 would like to acknowledge the SUSFERT project funding from the Bio Based Industries Joint Undertaking
659 (BBI-JU) under the European Union's Horizon 2020 research and innovation program under grant
660 agreement No. 792021.

661 8 CRediT authorship contribution statement

662 **Marc Spiller:** Conceptualization, Methodology, Formal analysis, Writing - original draft, Data curation,
663 Visualization. **Michele Moretti:** Methodology, Formal analysis, Writing - original draft, Data curation.

664 **Jolien De Paepe:** Writing - review & editing, Visualization. **Siegfried Vlaeminck:** Conceptualization,
665 Visualization, Writing - review & editing

666

667 9 References

- 668 Acosta, N., De Vrieze, J., 2018. Anaerobic Digestion as Key Technology in the Bio-Based Economy, in:
669 Stams, A.J.M., Sousa, D. (Eds.), *Biogenesis of Hydrocarbons*. Springer International Publishing, Cham, pp.
670 1-19.
- 671 Alloul, A., Ganigué, R., Spiller, M., Meerburg, F., Cagnetta, C., Rabaey, K., Vlaeminck, S.E., 2018. Capture–
672 Ferment–Upgrade: A Three-Step Approach for the Valorization of Sewage Organics as Commodities.
673 *Environmental Science & Technology* 52(12), 6729-6742.
- 674 Arashiro, L.T., Ferrer, I., Rousseau, D.P.L., Van Hulle, S.W.H., Garfí, M., 2019. The effect of primary
675 treatment of wastewater in high rate algal pond systems: Biomass and bioenergy recovery. *Bioresource*
676 *Technology* 280, 27-36.
- 677 Arashiro, L.T., Montero, N., Ferrer, I., Acién, F.G., Gómez, C., Garfí, M., 2018. Life cycle assessment of high
678 rate algal ponds for wastewater treatment and resource recovery. *Science of The Total Environment* 622-
679 623, 1118-1130.
- 680 Aziz, M., Wijayanta, A.T., Nandiyanto, A.B.D., 2020. Ammonia as Effective Hydrogen Storage: A Review on
681 Production, Storage and Utilization. *Energies* 13(12), 3062.
- 682 Baldi, M., Collivignarelli, M., Abbà, A., Benigna, I., 2018. The Valorization of Ammonia in Manure Digestate
683 by Means of Alternative Stripping Reactors. *Sustainability* 10(9), 3073.
- 684 Bicer, Y., Dincer, I., Zamfirescu, C., Vezina, G., Raso, F., 2016. Comparative life cycle assessment of various
685 ammonia production methods. *Journal of Cleaner Production* 135, 1379-1395.
- 686 Bisschops, I., Kjerstadius, H., Meulman, B., Van Eekert, M., 2019. Integrated nutrient recovery from
687 source-separated domestic wastewaters for application as fertilisers. *Current Opinion in Environmental*
688 *Sustainability* 40, 7-13.
- 689 Bokun, C., Yu, Q., Siyu, Y., 2019. Integration of thermo-vapor compressors with phenol and ammonia
690 recovery process for coal gasification wastewater treatment system. *Energy* 166, 108-117.
- 691 Bolzonella, D., Fatone, F., Gottardo, M., Frison, N., 2018. Nutrients recovery from anaerobic digestate of
692 agro-waste: Techno-economic assessment of full scale applications. *Journal of environmental*
693 *management* 216, 111-119.
- 694 Bratina, B., Šorgo, A., Kramberger, J., Ajdnik, U., Zemljič, L.F., Ekart, J., Šafarič, R., 2016. From
695 municipal/industrial wastewater sludge and FOG to fertilizer: A proposal for economic sustainable sludge
696 management. *Journal of environmental management* 183, 1009-1025.
- 697 Brentrup, F., Lammel, J., Stephani, T., Christensen, B., 2016. Updated carbon footprint values for mineral
698 fertilizer from different world regions.
- 699 Bridle, T., Skrypski-Mantele, S., 2000. Assessment of sludge reuse options: a life-cycle approach. *Water*
700 *science and technology* 41(8), 131-135.
- 701 Burgin, A.J., Hamilton, S.K., 2007. Have we overemphasized the role of denitrification in aquatic
702 ecosystems? A review of nitrate removal pathways. *Frontiers in Ecology and the Environment* 5(2), 89-96.
- 703 Buyle, Audenaert, Billen, Boonen, Van, P., 2019. The Future of Ex-Ante LCA? Lessons Learned and Practical
704 Recommendations. *Sustainability* 11(19), 5456.
- 705 Cai, T., Park, S.Y., Li, Y., 2013. Nutrient recovery from wastewater streams by microalgae: Status and
706 prospects. *Renewable and Sustainable Energy Reviews* 19, 360-369.
- 707 Campbell, B.M., Beare, D.J., Bennett, E.M., Hall-Spencer, J.M., Ingram, J.S.I., Jaramillo, F., Ortiz, R.,
708 Ramankutty, N., Sayer, J.A., Shindell, D., 2017. Agriculture production as a major driver of the Earth system
709 exceeding planetary boundaries. *Ecology and Society* 22(4).
- 710 Case, S., Oelofse, M., Hou, Y., Oenema, O., Jensen, L.S., 2017. Farmer perceptions and use of organic waste
711 products as fertilisers—A survey study of potential benefits and barriers. *Agricultural systems* 151, 84-95.

712 Cherkasov, N., Ibhaddon, A.O., Fitzpatrick, P., 2015. A review of the existing and alternative methods for
713 greener nitrogen fixation. *Chemical Engineering and Processing: Process Intensification* 90, 24-33.

714 Corbin, J.M., Strauss, A., 1990. Grounded theory research: Procedures, canons, and evaluative criteria.
715 *Qualitative Sociology* 13(1), 3-21.

716 Crab, R., Defoirdt, T., Bossier, P., Verstraete, W., 2012. Biofloc technology in aquaculture: Beneficial effects
717 and future challenges. *Aquaculture* 356-357, 351-356.

718 Dawson, C.J., Hilton, J., 2011. Fertiliser availability in a resource-limited world: Production and recycling
719 of nitrogen and phosphorus. *Food Policy* 36, S14-S22.

720 de Faria, A.B., Spérandio, M., Ahmadi, A., Tiruta-Barna, L., 2015. Evaluation of new alternatives in
721 wastewater treatment plants based on dynamic modelling and life cycle assessment (DM-LCA). *Water*
722 *research* 84, 99-111.

723 de Souza, M.H.B., Calijuri, M.L., Assemany, P.P., de Siqueira Castro, J., de Oliveira, A.C.M., 2019. Soil
724 application of microalgae for nitrogen recovery: A life-cycle approach. *Journal of Cleaner Production* 211,
725 342-349.

726 De Vrieze, J., Colica, G., Pintucci, C., Sarli, J., Pedizzi, C., Willeghems, G., Bral, A., Varga, S., Prat, D., Peng,
727 L., Spiller, M., Buysse, J., Colsen, J., Benito, O., Carballa, M., Vlaeminck, S.E., 2019. Resource recovery from
728 pig manure via an integrated approach: A technical and economic assessment for full-scale applications.
729 *Bioresource Technology* 272, 582-593.

730 De Vrieze, J., Smet, D., Klok, J., Colsen, J., Angenent, L.T., Vlaeminck, S.E., 2016. Thermophilic sludge
731 digestion improves energy balance and nutrient recovery potential in full-scale municipal wastewater
732 treatment plants. *Bioresource Technology* 218, 1237-1245.

733 Deviatkin, I., Lyu, L., Chen, S., Havukainen, J., Wang, F., Horttanainen, M., Mänttari, M., 2019. Technical
734 implications and global warming potential of recovering nitrogen released during continuous thermal
735 drying of sewage sludge. *Waste Management* 90, 132-140.

736 Driver, J.G., Owen, R.E., Makanyire, T., Lake, J.A., McGregor, J., Styring, P., 2019. Blue Urea: Fertilizer With
737 Reduced Environmental Impact. *Frontiers in Energy Research* 7(88).

738 Dube, P., Vanotti, M., Szogi, A., García-González, M., 2016. Enhancing recovery of ammonia from swine
739 manure anaerobic digester effluent using gas-permeable membrane technology. *Waste management* 49,
740 372-377.

741 EC, 1991. Consolidated text: Council Directive of 12 December 1991 concerning the protection of waters
742 against pollution caused by nitrates from agricultural sources (91/676/EEC). [https://eur-](https://eur-lex.europa.eu/legal-content/EN/TXT/?qid=1561542776070&uri=CELEX:01991L0676-20081211)
743 [lex.europa.eu/legal-content/EN/TXT/?qid=1561542776070&uri=CELEX:01991L0676-20081211](https://eur-lex.europa.eu/legal-content/EN/TXT/?qid=1561542776070&uri=CELEX:01991L0676-20081211).
744 (02/12/2021).

745 EC, 2019. Regulation (EU) 2019/1009 of the European Parliament and of the council of 5 June 2019 laying
746 down rules on the making available on the market of EU fertilising products and amending regulations
747 (EC) no 1069/2009 and (EC) no 1107/2009 and repealing regulation (EC) no 2003/2003 (text with EEA
748 relevance). European Parliament. [https://eur-lex.europa.eu/legal-](https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32019R1009)
749 [content/EN/TXT/?uri=CELEX%3A32019R1009](https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32019R1009). (02/12/2021).

750ecoinvent, 2021. LCA database ecoinvent 3.8. <https://ecoinvent.org/>. (24/01/2022).

751 Erisman, J.W., Sutton, M.A., Galloway, J., Klimont, Z., Winiwarter, W., 2008. How a century of ammonia
752 synthesis changed the world. *Nature Geoscience* 1(10), 636-639.

753 Fang, L.L., Valverde-Pérez, B., Damgaard, A., Plósz, B.G., Rygaard, M., 2016. Life cycle assessment as
754 development and decision support tool for wastewater resource recovery technology. *Water research* 88,
755 538-549.

756 Fertilizer Europe, 2021. Industry facts and figures 2020. [https://www.fertilizerseurope.com/wp-](https://www.fertilizerseurope.com/wp-content/uploads/2020/07/Industry-Facts-and-Figures-2020-spreads.pdf)
757 [content/uploads/2020/07/Industry-Facts-and-Figures-2020-spreads.pdf](https://www.fertilizerseurope.com/wp-content/uploads/2020/07/Industry-Facts-and-Figures-2020-spreads.pdf). (24/12/2021).

758 Fowler, D., Coyle, M., Skiba, U., Sutton, M.A., Cape, J.N., Reis, S., Sheppard, L.J., Jenkins, A., Grizzetti, B.,
759 Galloway, J.N., Vitousek, P., Leach, A., Bouwman, A.F., Butterbach-Bahl, K., Dentener, F., Stevenson, D.,
760 Amann, M., Voss, M., 2013. The global nitrogen cycle in the twenty-first century. *Philosophical*
761 *Transactions of the Royal Society B: Biological Sciences* 368(1621), 20130164.

762 Fux, C., Siegrist, H., 2004. Nitrogen removal from sludge digester liquids by nitrification/denitrification or
763 partial nitritation/anammox: environmental and economical considerations. *Water Science and*
764 *Technology* 50(10), 19-26.

765 Galloway, J.N., Dentener, F.J., Capone, D.G., Boyer, E.W., Howarth, R.W., Seitzinger, S.P., Asner, G.P.,
766 Cleveland, C.C., Green, P.A., Holland, E.A., Karl, D.M., Michaels, A.F., Porter, J.H., Townsend, A.R.,
767 Vöosmarty, C.J., 2004. Nitrogen Cycles: Past, Present, and Future. *Biogeochemistry* 70(2), 153-226.

768 Ghyselbrecht, K., Monballiu, A., Somers, M.H., Sigurnjak, I., Meers, E., Appels, L., Meesschaert, B., 2018.
769 Stripping and scrubbing of ammonium using common fractionating columns to prove ammonium
770 inhibition during anaerobic digestion. *International Journal of Energy and Environmental Engineering* 9(4),
771 447-455.

772 Gu, N., Liu, J., Ye, J., Chang, N., Li, Y.-Y., 2019. Bioenergy, ammonia and humic substances recovery from
773 municipal solid waste leachate: A review and process integration. *Bioresource technology* 293, 122159.

774 He, Q., Xi, J., Shi, M., Feng, L., Yan, S., Deng, L., 2020. Developing a Vacuum-Assisted Gas-Permeable
775 Membrane Process for Rapid Ammonia Recovery and CO₂ Capture from Biogas Slurry. *ACS Sustainable*
776 *Chemistry & Engineering* 8(1), 154-162.

777 Henze, M., van Loosdrecht, M.C., Ekama, G.A., Brdjanovic, D., 2008. Biological wastewater treatment. IWA
778 publishing.

779 Hermassi, M., Dosta, J., Valderrama, C., Licon, E., Moreno, N., Querol, X., Batis, N., Cortina, J., 2018.
780 Simultaneous ammonium and phosphate recovery and stabilization from urban sewage sludge anaerobic
781 digestates using reactive sorbents. *Science of the total environment* 630, 781-789.

782 Hou, Y., Velthof, G., Case, S., Oelofse, M., Grignani, C., Balsari, P., Zavattaro, L., Gioelli, F., Bernal, M.,
783 Fanguero, D., 2018. Stakeholder perceptions of manure treatment technologies in Denmark, Italy, the
784 Netherlands and Spain. *Journal of Cleaner Production* 172, 1620-1630.

785 Huang, X., Guida, S., Jefferson, B., Soares, A., 2020. Economic evaluation of ion-exchange processes for
786 nutrient removal and recovery from municipal wastewater. *npj Clean Water* 3(1).

787 IEA, 2021. Ammonia Technology Roadmap. Towards more sustainable nitrogen fertiliser production.
788 [https://iea.blob.core.windows.net/assets/6ee41bb9-8e81-4b64-8701-](https://iea.blob.core.windows.net/assets/6ee41bb9-8e81-4b64-8701-2acc064ff6e4/AmmoniaTechnologyRoadmap.pdf)
789 [2acc064ff6e4/AmmoniaTechnologyRoadmap.pdf](https://iea.blob.core.windows.net/assets/6ee41bb9-8e81-4b64-8701-2acc064ff6e4/AmmoniaTechnologyRoadmap.pdf). (10/03/2022).

790 Igos, E., Besson, M., Navarrete Gutiérrez, T., Bisinella De Faria, A.B., Benetto, E., Barna, L., Ahmadi, A.,
791 Spérandio, M., 2017. Assessment of environmental impacts and operational costs of the implementation
792 of an innovative source-separated urine treatment. *Water Research* 126, 50-59.

793 International Fertilizer Organisation, 2014. IFA Stat.
794 [https://www.fertilizer.org/Public/Stewardship/Publication_Detail.aspx?SEQN=4886&PUBKEY=51F326FC-](https://www.fertilizer.org/Public/Stewardship/Publication_Detail.aspx?SEQN=4886&PUBKEY=51F326FC-E64F-4507-83A0-31B2AFAB8BB3)
795 [-E64F-4507-83A0-31B2AFAB8BB3](https://www.fertilizer.org/Public/Stewardship/Publication_Detail.aspx?SEQN=4886&PUBKEY=51F326FC-E64F-4507-83A0-31B2AFAB8BB3). (10/07/2020).

796 Ishii, S.K., Boyer, T.H., 2015. Life cycle comparison of centralized wastewater treatment and urine source
797 separation with struvite precipitation: Focus on urine nutrient management. *Water research* 79, 88-103.

798 Jamaludin, Z., Rollings-Scattergood, S., Lutes, K., Vaneckhaute, C., 2018. Evaluation of sustainable
799 scrubbing agents for ammonia recovery from anaerobic digestate. *Bioresource Technology* 270, 596-602.

800 Johansson, K., Perzon, M., Fröling, M., Mossakowska, A., Svanström, M., 2008. Sewage sludge handling
801 with phosphorus utilization—life cycle assessment of four alternatives. *Journal of Cleaner Production* 16(1),
802 135-151.

803 Kanter, D.R., Bartolini, F., Kugelberg, S., Leip, A., Oenema, O., Uwizeye, A., 2020. Nitrogen pollution policy
804 beyond the farm. *Nature Food* 1(1), 27-32.

805 Kavvada, O., Tarpeh, W.A., Horvath, A., Nelson, K.L., 2017. Life-Cycle Cost and Environmental Assessment
806 of Decentralized Nitrogen Recovery Using Ion Exchange from Source-Separated Urine through Spatial
807 Modeling. *Environmental Science & Technology* 51(21), 12061-12071.

808 Kjerstadius, H., Haghhighatafshar, S., Davidsson, Å., 2015. Potential for nutrient recovery and biogas
809 production from blackwater, food waste and greywater in urban source control systems. *Environmental*
810 *technology* 36(13), 1707-1720.

811 Kuntke, P., Śmiech, K.M., Bruning, H., Zeeman, G., Saakes, M., Sleutels, T.H.J.A., Hamelers, H.V.M.,
812 Buisman, C.J.N., 2012. Ammonium recovery and energy production from urine by a microbial fuel cell.
813 *Water Research* 46(8), 2627-2636.

814 Lackner, S., Gilbert, E.M., Vlaeminck, S.E., Joss, A., Horn, H., Van Loosdrecht, M.C.M., 2014. Full-scale
815 partial nitrification/anammox experiences – An application survey. *Water Research* 55, 292-303.

816 Lam, K.L., Zlatanović, L., Van Der Hoek, J.P., 2020. Life cycle assessment of nutrient recycling from
817 wastewater: A critical review. *Water Research* 173, 115519.

818 Landry, K.A., Boyer, T.H., 2016. Life cycle assessment and costing of urine source separation: Focus on
819 nonsteroidal anti-inflammatory drug removal. *Water research* 105, 487-495.

820 Larsen, T.A., Riechmann, M.E., Udert, K.M., 2021. State of the art of urine treatment technologies: A
821 critical review. *Water Research X*, 100114.

822 Lienert, J., Larsen, T.A., 2010. High acceptance of urine source separation in seven European countries: a
823 review. *Environmental science & technology* 44(2), 556-566.

824 Lin, Y., Guo, M., Shah, N., Stuckey, D.C., 2016. Economic and environmental evaluation of nitrogen
825 removal and recovery methods from wastewater. *Bioresource Technology* 215, 227-238.

826 Macfarlane, D.R., Cherepanov, P.V., Choi, J., Suryanto, B.H.R., Hodgetts, R.Y., Bakker, J.M., Ferrero Vallana,
827 F.M., Simonov, A.N., 2020. A Roadmap to the Ammonia Economy. *Joule* 4(6), 1186-1205.

828 Matassa, S., Batstone, D.J., Hülsen, T., Schnoor, J., Verstraete, W., 2015. Can Direct Conversion of Used
829 Nitrogen to New Feed and Protein Help Feed the World? *Environmental Science & Technology* 49(9),
830 5247-5254.

831 Matassa, S., Papirio, S., Pikaar, I., Hülsen, T., Leijenhof, E., Esposito, G., Pirozzi, F., Verstraete, W., 2020.
832 Upcycling of biowaste carbon and nutrients in line with consumer confidence: the “full gas” route to single
833 cell protein. *Green Chemistry* 22(15), 4912-4929.

834 Matzen, M.J., Alhajji, M.H., Demirel, Y., 2015. Technoeconomics and Sustainability of Renewable
835 Methanol and Ammonia Productions Using Wind Power-based Hydrogen. *Journal of Advanced Chemical*
836 *Engineering* 5(3).

837 Maurer, M., Schwegler, P., Larsen, T., 2003. Nutrients in urine: energetic aspects of removal and recovery.
838 *Water Science and technology* 48(1), 37-46.

839 Menkveld, H., Broeders, E., 2018. Recovery of ammonia from digestate as fertilizer. *Water Practice &*
840 *Technology* 13(2), 382-387.

841 Munasinghe-Arachchige, S.P., Abeywardana-Arachchige, I.S., Delanka-Pedige, H.M., Nirmalakhandan,
842 N., 2020. Sewage treatment process refinement and intensification using multi-criteria decision making
843 approach: a case study. *Journal of Water Process Engineering* 37, 101485.

844 Muys, M., Papini, G., Spiller, M., Sakarika, M., Schwaiger, B., Lesueur, C., Vermeir, P., Vlaeminck, S.E., 2020.
845 Dried aerobic heterotrophic bacteria from treatment of food and beverage effluents: Screening of
846 correlations between operation parameters and microbial protein quality. *Bioresource Technology* 307,
847 123242.

848 Muys, M., Phukan, R., Brader, G., Samad, A., Moretti, M., Haiden, B., Pluchon, S., Roest, K., Vlaeminck,
849 S.E., Spiller, M., 2021. A systematic comparison of commercially produced struvite: Quantities, qualities
850 and soil-maize phosphorus availability. *Science of The Total Environment* 756, 143726.

851 National Geographic, 2022. Anthropocene.
852 <https://www.nationalgeographic.org/encyclopedia/anthropocene/>. (15/03/2022).

853 Pradel, M., Aissani, L., Villot, J., Baudez, J.-C., Laforest, V., 2016. From waste to added value product:
854 towards a paradigm shift in life cycle assessment applied to wastewater sludge – a review. *Journal of*
855 *Cleaner Production* 131, 60-75.

856 Prado, L.O., Souza, H.H., Chiquito, G.M., Paulo, P.L., Boncz, M.A., 2020. A comparison of different scenarios
857 for on-site reuse of blackwater and kitchen waste using the life cycle assessment methodology.
858 *Environmental Impact Assessment Review* 82, 106362.

859 Pretel, R., Shoener, B., Ferrer, J., Guest, J., 2015. Navigating environmental, economic, and technological
860 trade-offs in the design and operation of submerged anaerobic membrane bioreactors (AnMBRs). *Water*
861 *research* 87, 531-541.

862 QSR International Pty Ltd., 2018. NVivo qualitative data analysis software,, Version 12 ed.

863 Rockström, J., Steffen, W., Noone, K., Persson, Å., Chapin, F.S., Lambin, E.F., Lenton, T.M., Scheffer, M.,
864 Folke, C., Schellnhuber, H.J., Nykvist, B., De Wit, C.A., Hughes, T., Van Der Leeuw, S., Rodhe, H., Sörlin, S.,
865 Snyder, P.K., Costanza, R., Svedin, U., Falkenmark, M., Karlberg, L., Corell, R.W., Fabry, V.J., Hansen, J.,
866 Walker, B., Liverman, D., Richardson, K., Crutzen, P., Foley, J.A., 2009. A safe operating space for humanity.
867 *Nature* 461(7263), 472-475.

868 Scheer, C., Fuchs, K., Pelster, D.E., Butterbach-Bahl, K., 2020. Estimating global terrestrial denitrification
869 from measured N₂O:(N₂O + N₂) product ratios. *Current Opinion in Environmental Sustainability* 47, 72-
870 80.

871 Sfez, S., De Meester, S., Vlaeminck, S.E., Dewulf, J., 2019. Improving the resource footprint evaluation of
872 products recovered from wastewater: A discussion on appropriate allocation in the context of circular
873 economy. *Resources, Conservation and Recycling* 148, 132-144.

874 Sigmundjak, I., Brienza, C., Snauwaert, E., De Dobbelaere, A., De Mey, J., Vaneekhaute, C., Michels, E.,
875 Schoumans, O., Adani, F., Meers, E., 2019. Production and performance of bio-based mineral fertilizers
876 from agricultural waste using ammonia (stripping-)scrubbing technology. *Waste Management* 89, 265-
877 274.

878 Sigmundjak, I., Michels, E., Crappé, S., Buysens, S., Tack, F.M.G., Meers, E., 2016. Utilization of derivatives
879 from nutrient recovery processes as alternatives for fossil-based mineral fertilizers in commercial
880 greenhouse production of *Lactuca sativa* L. *Scientia Horticulturae* 198, 267-276.

881 Sigmundjak, I., Vaneekhaute, C., Michels, E., Ryckaert, B., Ghekiere, G., Tack, F., Meers, E., 2017. Fertilizer
882 performance of liquid fraction of digestate as synthetic nitrogen substitute in silage maize cultivation for
883 three consecutive years. *Science of the Total Environment* 599, 1885-1894.

884 Smith, C., Hill, A.K., Torrente-Murciano, L., 2020. Current and future role of Haber–Bosch ammonia in a
885 carbon-free energy landscape. *Energy & Environmental Science* 13(2), 331-344.

886 Spanoghe, J., Grunert, O., Wambacq, E., Sakarika, M., Papini, G., Alloul, A., Spiller, M., Derycke, V., Stragier,
887 L., Verstraete, H., Fauconnier, K., Verstraete, W., Haesaert, G., Vlaeminck, S.E., 2020. Storage, fertilization
888 and cost properties highlight the potential of dried microbial biomass as organic fertilizer. *Microbial*
889 *Biotechnology* n/a(n/a).

890 Spiller, M., Muys, M., Papini, G., Sakarika, M., Buyle, M., Vlaeminck, S.E., 2020. Environmental impact of
891 microbial protein from potato wastewater as feed ingredient: Comparative consequential life cycle
892 assessment of three production systems and soybean meal. *Water Research* 171, 115406.

893 Steffen, W., Richardson, K., Rockstrom, J., Cornell, S.E., Fetzer, I., Bennett, E.M., Biggs, R., Carpenter, S.R.,
894 De Vries, W., De Wit, C.A., Folke, C., Gerten, D., Heinke, J., Mace, G.M., Persson, L.M., Ramanathan, V.,
895 Reyers, B., Sorlin, S., 2015. Planetary boundaries: Guiding human development on a changing planet.
896 *Science* 347(6223), 1259855-1259855.

897 STOWA, 2021. STIKSTOFTERUGWINNING UIT RIOOLWATER; VAN MARKTAMBITIE NAAR PRAKTIJK.

898 Sutton, M.A., Bleeker, A., Howard, C., Erisman, J., Abrol, Y., Bekunda, M., Datta, A., Davidson, E., De Vries,
899 W., Oenema, O., 2013. Our nutrient world. The challenge to produce more food & energy with less
900 pollution.

901 Tao, W., Bayrakdar, A., Wang, Y., Agyeman, F., 2019. Three-stage treatment for nitrogen and phosphorus
902 recovery from human urine: Hydrolysis, precipitation and vacuum stripping. *Journal of environmental*
903 *management* 249, 109435.

904 Thomassen, G., Van Passel, S., Dewulf, J., 2020. A review on learning effects in prospective technology
905 assessment. *Renewable and Sustainable Energy Reviews* 130, 109937.

906 Tur-Cardona, J., Bonnicksen, O., Speelman, S., Verspecht, A., Carpentier, L., Debruyne, L., Marchand, F.,
907 Jacobsen, B.H., Buysse, J., 2018. Farmers' reasons to accept bio-based fertilizers: A choice experiment in
908 seven different European countries. *Journal of Cleaner Production* 197, 406-416.

909 van der Hoek, J.P., Duijff, R., Reinstra, O., 2018. Nitrogen recovery from wastewater. *Sustainability*
910 *(Switzerland)* 10(12).

911 Vaneekhaute, C., Meers, E., Michels, E., Ghekiere, G., Accoe, F., Tack, F.M.G., 2013. Closing the nutrient
912 cycle by using bio-digestion waste derivatives as synthetic fertilizer substitutes: A field experiment.
913 *Biomass and Bioenergy* 55, 175-189.

914 Verbeeck, K., De Vrieze, J., Pikaar, I., Verstraete, W., Rabaey, K., 2020. Assessing the potential for up-
915 cycling recovered resources from anaerobic digestion through microbial protein production. *Microbial*
916 *Biotechnology*.

917 Volpin, F., Heo, H., Hasan Johir, M.A., Cho, J., Phuntsho, S., Shon, H.K., 2019. Techno-economic feasibility
918 of recovering phosphorus, nitrogen and water from dilute human urine via forward osmosis. *Water*
919 *Research* 150, 47-55.

920 Vulsteke, E., Van Den Hende, S., Bourez, L., Capoen, H., Rousseau, D.P.L., Albrecht, J., 2017. Economic
921 feasibility of microalgal bacterial floc production for wastewater treatment and biomass valorization: A
922 detailed up-to-date analysis of up-scaled pilot results. *Bioresource Technology* 224, 118-129.

923 Wang, M., Khan, M.A., Mohsin, I., Wicks, J., Ip, A.H., Sumon, K.Z., Dinh, C.-T., Sargent, E.H., Gates, I.D.,
924 Kibria, M.G., 2021. Can sustainable ammonia synthesis pathways compete with fossil-fuel based Haber–
925 Bosch processes? *Energy & Environmental Science* 14(5), 2535-2548.

926 Wett, B., Podmirseg, S.M., Gómez-Brandón, M., Hell, M., Nyhuis, G., Bott, C., Murthy, S., 2015. Expanding
927 DEMON Sidestream Deammonification Technology Towards Mainstream Application. 87(12), 2084-2089.

928 Winkler, M.K., Straka, L., 2019. New directions in biological nitrogen removal and recovery from
929 wastewater. *Current Opinion in Biotechnology* 57, 50-55.

930 World Bank, 2021. World Bank Commodities Price Data (The Pink Sheet).
931 <https://www.worldbank.org/en/research/commodity-markets>. (14/09/2020).

932 Xu, X., Dao, H., Bair, R., Uman, A.E., Yeh, D., Zhang, Q., 2020. Discharge or reuse? Comparative
933 sustainability assessment of anaerobic and aerobic membrane bioreactors. *Journal of Environmental*
934 *Quality* 49(3), 545-556.

935 Zamfirescu, C., Dincer, I., 2008. Using ammonia as a sustainable fuel. *Journal of Power Sources* 185(1),
936 459-465.

937