#### Multi-scale techno-economic assessment of nitrogen recovery systems 1 for livestock operations 2

Edgar Martín-Hernández<sup>a,b,c,\*</sup>, Clara Montero-Rueda<sup>a</sup>, Gerardo J. Ruiz-Mercado<sup>d,e</sup>, Céline Vaneeckhaute<sup>b,c</sup>, Mariano Martín<sup>a</sup>

<sup>a</sup>Department of Chemical Engineering, University of Salamanca, Plza. Caídos 1-5, 37008 Salamanca, Spain <sup>b</sup>BioEngine - Research Team on Green Process Engineering and Biorefineries, Chemical Engineering Department, 6

Université Laval, 1065 Ave. de la Médecine, Québec, QC, G1V 0A6, Canada <sup>c</sup>CentrEau, Centre de recherche sur l'eau, Université Laval, 1065 Avenue de la Médecine, Québec, QC, G1V 0A6,

Canada

<sup>d</sup>Center for Environmental Solutions and Emergency Response (CESER), US Environmental Protection Agency, 10 26 West Martin Luther King Drive, Cincinnati, Ohio 45268, United States 11 12

<sup>e</sup>Chemical Engineering Graduate Program, Universidad del Atlántico, Puerto Colombia 080007, Colombia

#### Abstract 13

3

7

8

q

Intensive livestock farming generates vast amounts of organic materials, which are an important source of nitrogen releases. These anthropogenic nitrogen releases contribute to multiple environmental problems, including eutrophication of water systems, contamination of drinking water sources, and greenhouse gas emissions. Nitrogen recovery and recycling are technically feasible, and there exists a number of processes for nitrogen recovery from livestock waste in the form of different products. In this work, a multi-scale techno-economic assessment of techniques for nitrogen recovery and recycling is performed. The assessment performed included the material flow analysis of each process, from waste collection to final treatment, to determine the recovery efficiency and nitrogen losses of each system and nitrogen recovery cost, as well as an environmental cost-benefit analysis to compare the nitrogen recovery cost versus the economic losses derived from its uncontrolled release into the environment. The results show that transmembrane chemisorption process results in the lowest recovery cost, 3.4-10.4 USD per kilogram of nitrogen recovered in the range of scales studied. The recovery of nitrogen from livestock waste through three technologies, i.e., transmembrane chemisorption, MAPHEX, and stripping in packed bed, is revealed to be cost-effective. Since the economic losses due to the harmful effects of nitrogen into the environment are estimated at 32-35 USD per kilogram of nitrogen released, nitrogen recycling is an environmentally and economically

<sup>\*</sup>Corresponding author

Email address: edmah2@ulaval.ca (Edgar Martín-Hernández)

beneficial approach to reduce nutrient pollution caused by livestock operations.

- 14 Keywords: Organic Waste, Nitrogen Recovery, Nutrient Pollution, Livestock Industry,
- 15 Techno-economic assessment

# <sup>16</sup> 1. Introduction

The agricultural sector experimented with industrialization processes since the XIX century, 17 pursuing the intensification of food production, i.e., increasing the agricultural production per unit 18 of input resources, including land, labor, and feedstock among others (FAO, 2004). During the 19 last decades, the agricultural intensification is driven by a sustained increase in the population, the 20 average income growth in both developed and developing countries, and the trade liberalization and 21 logistics advancements leading the transnational trade of products (Baker and Da Silva, 2014). As 22 a result of this intensification process, the largest quantity and variety of agri-products in human 23 history is produced and distributed nowadays. However, multiple environmental challenges must 24 be faced because of the industrialization of agriculture and farming activities such as depletion of 25 nutrients and organic matter in soil, excessive and inefficient use of synthetic fertilizers to maintain 26 high crop yields, spatial concentration and inappropriate management of livestock manure, and 27 biodiversity loss, among others. 28

Within the context of nutrient management, livestock facilities have decoupled the previous 20 synergistic link where the organic waste from livestock activities were used as a nutrient and organic 30 matter supply for crop production (Bouwman et al., 2009). This decoupling created a dependency 31 on mineral and synthetic fertilizers for crop production, as well as serious problems concerning 32 the adequate management of animal manure intensive livestock operations, known as concentrated 33 animal feeding operations (CAFOs) (U.S. Department of Agriculture, 2011). Therefore, there exist 34 a disconnection between the areas with a large surplus of nutrients as a result of manure releases 35 from intensive livestock operations, and croplands demanding nitrogen and phosphorus relying on 36 synthetic fertilizers to keep high crop yield rates (Kahiluoto et al., 2021). It must be noted that 37 applying manure in croplands is a common practice, supplying nutrients for crops growing. However, 38 since manure is a bulky material expensive to transport, the application of manure is limited to 39

the fields in the vicinity of livestock operations, which are eventually overloaded with nutrients, 40 creating a nutrient legacy that is further transported to waterbodies by runoff. This process results 41 in the eutrophication of waterbodies, which can lead algal bloom episodes. In addition, a fraction of 42 the nitrogen released into the environment volatilizes in the form of ammonia, contributing to the 43 degradation of air quality and the formation of greenhouses formation (de Vries, 2021). Therefore, 44 the effective redistribution of nutrients is challenging and expensive due to the sparse location of 45 livestock operations and the high density of organic materials, including both manure and digestate 46 (Sampat et al., 2018a). 47

As a result, implementing technologies and processes for the recovery of nutrients in a form 48 suitable for easy transport and use in croplands is of utmost importance to restore the circularity 49 of agricultural nutrients usage disrupted by intensive industrial practices. However, the selection 50 of the most suitable technology for an individual livestock operation is not a trivial process, but 51 it is affected by multiple factors such as recovery efficiency, technology readiness, and the effect 52 of the scale on the economic performance of the process. In this regard, previous works assessed 53 and compared different technologies for the recovery of nitrogen from agricultural wastes, but they 54 do not capture the effect of the economies of scale (Munasinghe-Arachchige and Nirmalakhandan, 55 2020; De Vrieze et al., 2019; Bolzonella et al., 2018), or they are limited to a few technologies 56 (Kar et al., 2023). For example, Beckinghausen et al. (2020) reports a lack of techno-economic 57 analyses for nitrogen recovery techniques to identify the most suitable technology according to the 58 characteristics of each facility. Additionally, these studies do not capture the effects of integrating 59 nitrogen recovery technologies with additional processes for the recovery of other resources from 60 organic material such as anaerobic digestion. 61

This work aims to fill the gap in the literature providing a systematic techno-economic assessment of processes for nitrogen recovery from livestock waste for different scales of livestock facilities and different types of livestock waste, i.e., dairy, beef, swine, and poultry manure. Five processes representing the main technologies available for nitrogen recovery from livestock manure are assessed and compared, i.e., transmembrane chemisorption, ammonia evaporation and scrubbing, striping in packed tower, MAPHEX, and struvite precipitation. Each system is evaluated performing a

material flow analysis (MFA) (Brunner and Rechberger, 2016) of the whole process, from organic 68 feedstock collection to the final treatment, to determine the nitrogen recovery efficiency; and a 69 techno-economic analysis (TEA) (Burk, 2018) to estimate the recovery cost of nitrogen, including 70 a study of the effects of the economies of scale on the cost of nitrogen recovery. The assessment 71 of such technologies is performed through a flexible framework analyze multiple scales, livestock 72 wastes, and the potential integration of nitrogen recovery processes with anaerobic digestion. We 73 note that the scope of the work is limited to the techno-economic assessment of the nitrogen re-74 covery processes. As such, the nitrogen releases resulting from the fraction of nitrogen that the 75 recovery processes do not capture are the only emissions evaluated. 76

The objective of this work is to determine the economic dimension of nitrogen recovery from livestock waste attending to the economies of scale and the type of livestock. We compare the cost of nitrogen recovery with the cost caused by the environmental and social damages caused by the releases of nitrogen into the environment which are estimated by Sobota et al. (2015) and Compton et al. (2017) at 32.50 and 35.15 USD per kg of nitrogen released respectively, and contextualize the advantages of implementing nitrogen recovery techniques to to close the nutrients loop from livestock to crops.

# <sup>84</sup> 2. Methods

#### <sup>85</sup> 2.1. Livestock manure

Animals at different life stages generate different amounts of manure with different compositions. Data reported by the US Department of Agriculture (U.S. Department of Agriculture, 2009, 2000) and Pagliari and Laboski (2012) is used to determine the flow and composition of manure as a function of the number and type of animals in a facility, as listed in Table 1. AU denotes animal units, which is defined as 1000 pounds (453.6 kg) of live animal (U.S. Department of Agriculture, 2011).

We note that swine, dairy, beef, and poultry facilities are comprised of animals at different life stages, and thus the animal distribution shown in Table 2 for the techno-economic assessment (Statistics Canada – Statistique Canada, 2022).

Livestock type	Water (%)	Volatile solids (%)	Manure (kg/day/AU)	AU eq. (AU/animals)	${\scriptstyle \substack{\mathrm{N}_{\mathrm{total}}\\(\%)}}$	$\begin{array}{c} P_{total} \\ (\%) \end{array}$	$\begin{array}{c} \mathrm{N_{inorg}:N_{total}}\\ \mathrm{(ratio)} \end{array}$	$\begin{array}{c} P_{\rm inorg} : P_{\rm total} \\ (\rm ratio) \end{array}$
Dairy cow	87	10.98	37.88	0.74	0.59	0.08	0.33	0.54
Dairy heifer	83	13.04	29.95	0.94	0.48	0.09	0.33	0.54
Dairy calf	83	9.28	29.95	4.00	0.51	0.06	0.33	0.54
Beef cow	88	10.58	28.58	1.00	0.34	0.08	0.10	0.55
Beef calf	88	10.00	28.14	4.00	0.58	0.10	0.10	0.55
Sow	90	9.17	11.34	2.67	0.64	0.20	0.61	0.77
Boar	90	8.93	8.62	2.67	0.74	0.27	0.61	0.77
Piglets	90	9.99	39.92	9.09	1.05	0.17	0.61	0.77
Chickens layers	75	19.30	28.45	250	1.92	0.58	0.57	0.57
Chickens pullets	75	19.30	20.68	455	1.92	0.58	0.57	0.57
Chickens broilers	75	19.09	37.21	455	1.09	0.32	0.57	0.57

Table 1: Livestock manure properties. AU denotes animal units, while the *inorg* subscript denote the inorganic fraction of nutrients (U.S. Department of Agriculture, 2009, 2000; Pagliari and Laboski, 2012).

Table 2: Distribution of animals in swine, dairy, beef, and poultry facilities.

	Dairy		Beef		Swine		Poultry	
	$\begin{array}{c} \text{Heads} \\ (\%) \end{array}$	Animal units (%)	$\frac{\text{Heads}}{(\%)}$	Animal units (%)	$\frac{\text{Heads}}{(\%)}$	Animal units (%)	$\frac{\text{Heads}}{(\%)}$	Animal units (%)
Dairy cow	54	70	-	-	-	-	-	-
Dairy heifer	23	24	-	-	-	-	-	-
Dairy calf	23	6	-	-	-	-	-	-
Beef cow	-	-	66	89	-	-	-	-
Beef calf	-	-	34	11	-	-	-	-
Sow	-	-	-	-	7.6	22	-	-
Boar	-	-	-	-	0.4	1	-	-
Piglets	-	-	-	-	92	77	-	-
Chicken layers	-	-	-	-	-	-	21	33
Chicken pullets	-	-	-	-	-	-	8	7
Chicken broilers	-	-	-	-	-	-	71	60

# 95 2.2. Computational framework

The structure of the computational framework to connect the information relative to the size 96 of the livestock facilities and livestock waste composition with the MFA and TEA models is shown 97 in Figure 1. First, the data regarding the composition of the livestock waste to be processed 98 according to the type of animals of the livestock facility studied is imported into the MFA model 99 accordingly to the data collected in Tables 1 and 2 in order to determine the mass flows through the 100 different stages of the process, including the fractions of nitrogen that are recovered and released. 101 We consider two different scenarios as described in Section 2.3, i) the implementation of nitrogen 102 recovery processes standalone, and ii) the integration of anaerobic digestion and nitrogen recovery 103 systems. The information about the mass flows of the processes is fed into the techno-economic 104

model, performing a preliminary design and sizing of the process units to estimate the the capital
expenditures (CAPEX) and operating expenses (OPEX).

<sup>107</sup> These models are solved recursively for the different nitrogen recovery systems, livestock wastes,

<sup>108</sup> and sizes of livestock facilities to estimate the manure processing and nitrogen recovery costs for the different combinations of these parameters.



Figure 1: Structure of the computation framework for the multi-scale techno-economic assessment of nitrogen recovery systems for livestock operations. Boxes in blue and green denote the results obtained from the MFA and TEA models respectively.

109

# 110 2.3. Nitrogen recovery processes

The framework proposed comprises all livestock manure processing stages from manure collection to final nitrogen recovery, as shown in Figure 2. The main aspects of each processing stage are described in this section, while the comprehensive techno-economic modeling details can be found in the Supplementary Material.



Figure 2: Flowchart of the processes assessed for the processing of livestock manure. CHP unit stands for combined heat and power unit.

# 115 2.3.1. Anaerobic digestion

Livestock manure can be processed in an anaerobic digestion (AD) unit to produce biogas 116 and digestate. These materials can be further processed to recover valuable resources, such as 117 electricity and thermal energy generated from biogas. AD results in the partial mineralization of 118 the organic fraction of nitrogen and phosphorus, as shown in Table 3 (Fangueiro et al., 2020). In 119 addition, the total solids decreases as a consequence of transforming volatile solids into biogas. 120 AD process is typically carried out either at mesophilic (25 and 45 °C) or thermophilic (45 and 121 50 °C) temperatures and atmospheric pressure, with retention times between 30-40 and 15-20 days 122 respectively. Additionally, there exists low temperature digestion at psychrophilic conditions (< 25123 °C), although it involves longer retention times between 70 and 80 days (Al Saedi et al., 2008). A 124 digestion temperature of 40 °C and a retention time of 21 days are assumed in this work (Bolzonella 125 et al., 2018). 126

The composition of biogas produced is based on data reported by Ciborowski (2001). The energy requirements of the AD unit ( $Q_{digester}$ ), described in Eq 1, comprise the energy required for warming up the substrate from ambient temperature (assumed to be 12 °C) ( $Q_{feedstock}$ ) to the digestion temperature (40 °C), and the energy supplied to offset the digester heat losses ( $Q_{losses}$ ) (Penn State Extension, 2016). The heat generated due to microbial activity is considered negligible

	Volatile solids (%)	${\mathop{\rm N_{inorg}}\limits_{(\%)}}$	$\stackrel{\rm P_{inorg}}{(\%)}$	Biogas generation (m <sup>3</sup> / kg <sub>VS</sub> )
Variation (%)				
or	-52.5	+45	+16	0.57
generation rate $(m^3 / kg_{VS})$				

Table 3: Variation of organic material properties and biogas generation after the anaerobic digestion of livestock manure. VS subscript denotes volatile solids (Fangueiro et al., 2020).

<sup>132</sup> (Lübken et al., 2007). The details of the energy balance to the AD unit can be found in the <sup>133</sup> Supplementary Material, Eqs. 1S-8S. A maximum digester size  $(n_{AD, max})$  of 6000 m<sup>3</sup> is assumed <sup>134</sup> (Fachagentur Nachwachsende Rohstoffe, 2010).

$$Q_{\text{digester}} = Q_{\text{feedstock}} + Q_{\text{losses}} \tag{1}$$

<sup>135</sup> Correlations to estimate the CAPEX and OPEX have been developed based on data reported <sup>136</sup> by the USDA (Beddoes et al., 2007), as shown in Eqs. 9S-11S of the Supplementary Material.

# 137 2.3.2. Biogas conditioning

The raw biogas generated is conditioned to remove its impurities. Most of the moisture is 138 removed through condensation by compressing and cooling down the biogas stream. Hydrogen 139 sulfide  $(H_2S)$  is removed by using a fixed bed of  $Fe_2O_3$ , capturing the  $H_2S$  as  $Fe_2S_3$ . The bed 140 can be regenerated using the oxygen contained in air, leading to the formation of elementary 141 sulfur (Ryckebosch et al., 2011). Ammonia and the remaining moisture are removed through a 142 pressure swing adsorption (PSA) system. For both processes, two adsorption units are typically 143 installed in-parallel arrangement, so that one unit is in operation while the other bed is undergoing 144 regeneration. Removal yields of 100% have been assumed. More details can be found in Section 145 S1.2 of the Supplementary Material. 146

#### <sup>147</sup> 2.3.3. Combined heat and power generation

<sup>148</sup> Biogas is valorized using a combined heat and power (CHP) unit to produce electricity and <sup>149</sup> heat, which can be used to cover the thermal energy demand of the AD unit, and for the nitrogen <sup>150</sup> recovery processes if a source of heat is needed, e.g., in the ammonia evaporation process. The <sup>151</sup> energy recovered from biogas is estimated through its low heating value (LHV). LHV of biogas is a <sup>152</sup> function of methane content, and it can be estimated using Eq. 2 (Ludington, 2013), where  $x_{CH4}$ <sup>153</sup> refers to the methane mass fraction. Biogas combustion in the CHP unit is performed considering <sup>154</sup> a 20% air excess.

$$LHV_{\text{biogas}}\left(^{\text{J}/\text{m}^3}\right) = -46.26 \cdot 10^6 \cdot x_{\text{CH4}}^2 + 70.87 \cdot 10^6 \cdot x_{\text{CH4}} + 2.29 \cdot 10^6 \tag{2}$$

Based on data reported by CHP unit manufacturers, the electricity and thermal efficiencies 155 assumed are 40% and 50% respectively (Clarke Energy, 2013). The heat produced can be classified 156 in high-grade heat (HGH), which is recovered from the exhaust gases of combustion at 450 °C, and 157 low-grade heat (LGH) recovered from other points of the equipment at lower temperature. HGH 158 and LGH account for 62% and 38% of total heat energy respectively. LGH is used to cover the 159 energy demand of the AD units, while HGH is used for heat-intensive processes such as ammonia 160 evaporation. If the LGH from the CHP unit is not enough to cover the energy requirements of the 161 AD process, a fraction of HGH can be used to supplement the thermal energy supply. 162

# <sup>163</sup> 2.3.4. Digestate solid-liquid separation

Nutrients contained in manure or digestate can be found as part of organic and inorganic compounds. Organic nutrients are in the form of carbon-based solid compounds, and therefore they are mostly contained in the solid phase of the organic material. The nutrients bonded to organic compounds are not available for plants immediately, but they have to undergo a mineralization process to be transformed into inorganic nutrients (U.S. Department of Agriculture, 2009). On the other hand, the nitrogen contained in inorganic compounds can be used by plants and it is water soluble.

The liquid fraction of either manure or digestate, containing most of inorganic nutrients, is 171 recovered through a solid-liquid separation stage and further processed for nitrogen recovery. The 172 solid phase of the organic material can be composted, promoting the mineralization of a fraction of 173 the organic nutrients, and can be used as nutrient supplement for crop production. In addition, on-174 going studies employ these remaining organic materials as a growth medium for microorganisms of 175 industrial interest (Lamolinara et al., 2022). A screw press unit is considered for waste liquid-solid 176 phases separation (Møller et al., 2000). The partition coefficients for the different components, and 177 CAPEX and OPEX estimations considering the discretization of equipment size due to commercial 178 standard sizes are shown in the Section S1.3 of the Supplementary Material. We note the MAPHEX 179 process integrates the solid-liquid separation stage, and therefore no additional units are required 180 by this system. 181

#### 182 2.3.5. Nitrogen recovery systems

The technologies for nitrogen recovery assessed in this work, illustrated in Figure 3, are described 183 in this section. These processes target the recovery of inorganic nitrogen, since this is the water-184 soluble fraction of nitrogen contained in organic materials. In order to ensure the feasibility of the 185 nitrogen recovery processes assessed, only processes with a technology readiness level (TRL) equal 186 or above 6 (technology demonstrated in relevant environment) have been considered. The TRL of 187 each process is reported in Figure 3. It must be highlighted that the scope of this work focuses 188 on the recovery and recycling of nitrogen in order to achieve a circular economy of nutrients. As 189 such, processes for nitrogen removal resulting in products that cannot be recycled have not been 190 considered. 191

We note the OPEX reported include the expenditures of running each process, but do not include the amortization of the CAPEX. After estimating the CAPEX and OPEX of each process, CAPEX amortization and OPEX are combined to estimate the total cost of nitrogen recovery.

MAPHEX. MAPHEX is a nutrient recovery system based on physico-chemical separations de veloped by Penn State University and the U.S. Department of Agriculture (USDA), as shown in
 Fig. 3a. It is conceived as a mobile modular system which can be set in two interconnected truck



(d) Evaporation and wet scrubbing (TRL:8).



(e) Stripping and wet scrubbing (TRL: 8).

Figure 3: Schemes and technology readiness level (TRL) of the nitrogen recovery systems studied in the technoeconomic assessment.

trailers (Church et al., 2016). MAPHEX involves three stages: liquid-solid separation with a screw 198 press and a centrifuge, addition of iron sulfate to improve nutrients retention, and filtration with 199 diatomaceous earth as filter media. This combination of processes results in a liquid effluent mostly 200 composed of water with a low content of nutrients, while the nutrients are recovered as a solid 201 compound mainly composed of organic matter. The organic solid obtained contains the 93 % of 202 the total solids in the raw material, with a moisture content of 75%. 90% of both nitrogen and 203 phosphorus are recovered from this solid material (Church et al., 2018, 2016). This organic solid 204 has a lower density of nutrients (nitrogen and phosphorus) than other recovered products, such as 205 ammonium sulphate or struvite, resulting in a product with a lower market value. Moreover, the 206 low density of nutrients in the solid hinders the transportation and redistribution of the recovered 207 nitrogen to nutrient-deficient areas. As a result, no revenues from this product are considered. 208 Mass balances for MAPHEX, collected in Eqs. 18S-22S of the Supplementary Material, are based 209 on experimental data for full-scale modular units reported by Church et al. (2018). 210

Each MAPHEX unit is capable of processing up to 38 ton of manure per day with an associated operation cost of 0.054 USD per kilogram of manure processed. Capital cost of a MAPHEX unit is 291,000 USD (Church et al., 2018, 2016). Since MAPHEX is a unique-size system, the scaling study is based on the number of systems needed to process the livestock organic materials of each scenario considered.

Struvite precipitation. Struvite is a solid compound comprised by magnesium, ammonium, and 216 phosphate that can be formed through the chemical reaction shown in Eq. 3. Struvite precipitation 217 is a process usually aimed for phosphorus recovery, however struvite formation requires a significant 218 amount of nitrogen, and can therefore be intended for the simultaneous recovery of ammonia and 219 phosphorus. In this regard, recent studies have been published exploring the potential of struvite 220 production as a process for nitrogen recovery from livestock (Astals et al., 2021; Ha et al., 2023). 221 However, since phosphorus concentration in manure is lower than nitrogen concentration, phospho-222 rus acts as the limiting reactant for struvite formation, and in turn, for nitrogen recovery. It is 223 assumed that pH is adjusted to 9 for optimal struvite formation by using sodium hydroxide (Tao 224

<sup>225</sup> et al., 2016).

$$MgNH_4PO_4 \cdot 6H_2O \downarrow \rightleftharpoons Mg^{2+} + NH_4^+ + PO_4^{3-}$$
(3)

Ammonium, phosphate, and other relevant compounds for struvite formation, such as carbon-226 ates competing for phosphate ions to form calcium-based precipitates, are part of chemical systems 227 controlled by thermodynamic equilibrium. Therefore, a thermodynamic model for the formation of 228 struvite and calcium precipitates accounting the variability of elements concentration in manure was 229 developed in a previous work (Martín-Hernández et al., 2020). The chemical systems considered in 230 the model for estimating struvite formation are included in Tables 5S and 6S of the Supplementary 231 Material. Particularly, we note that calcium ions competing with magnesium ions for the phosphate 232 ions interfering in the formation of struvite. Consequently, a correlation to estimate the formation 233 of struvite as a function of calcium concentration in the organic feedstock processed has been used 234 in this work, Eq. 14S (Martín-Hernández et al., 2020). 235

Struvite precipitation is based on a single pass fluidized bed reactor (FBR), with no recirculation, 236 and conical design, as shown in Fig. 3b. The organic material is pumped to the bottom of the 237 reactor, as well as the magnesium supplement. The struvite particles grow, increasing their size, 238 until their mass overcomes the drag force of the uplift stream. The conical design of the reactor 239 keeps the small and lighter particles on the large diameter section at the top of the reactor, where 240 the superficial velocity is slower. As the particles increase their mass, they settle gradually to lower 241 levels of the reactor, where the diameter is smaller and the superficial velocity and drag force larger, 242 until they are finally settled on the bottom of the reactor. The liquid phase exits the reactor from 243 the top, where the cross-section is the widest, to ensure the retention of struvite fines. 244

For economic evaluation purposes, an unique-size system based on commercial struvite precipitation processes available is considered. This system has a capacity for processing up to 48,000 kg of digestate per day, with an associated CAPEX of 625,000 USD per each unit, plus 420,000 USD for the struvite dryer that serves all struvite precipitation units (AMPC, 2018). The OPEX of this system is 0.012 USD per kg of feedstock processed (AMPC, 2018). The scaling study is based on the number of reactors needed to process the livestock organic materials of each scenario considered.

Transmembrane chemisorption. Transmembrane chemisorption is a process based on the separation of gaseous species contained in a liquid stream by using a hydrophobic membrane. An additional liquid-solid separation stage is required to enhance the removal of solids and avoid clogging issues in the membrane. A centrifuge is considered for this stage, assuming the separation coefficients reported by Møller et al. (2000).

In the membrane unit, an acid stripping solution circulates on the lumen side of the membrane to capture the recovered gaseous components. For the case of ammonia recovery, a solution of sulfuric acid is commonly used as stripping fluid, resulting in the formation of ammonium sulfate, Eq 5. A 10% sulfuric acid solution is considered in this work (Darestani et al., 2017). Ammonia recovery efficiency is improved by displacing the ammonia-ammonium equilibrium, shown in Eq. 4, by raising the pH level up to 11 by adding sodium hydroxide.

$$\mathrm{NH}_3 + \mathrm{H}^+ \frac{K_b}{K_a} \mathrm{NH}_4^+ \tag{4}$$

$$2NH_3 + H_2SO_4 \to (NH_4)_2SO_4 \tag{5}$$

Membrane sizing is performed through the membrane mass balances proposed by Rongwong and 263 Sairiam (2020). These are based on the distribution of ammonia species within the digestate, mass 264 transfer on the digestate, wetted membrane, and non-wetted membrane phases, and the diffusion 265 of ammonia through the membrane. Ammonia diffusion through the membrane is driven by bulk 266 and Knudsen diffusivities, which consider the mean free path of molecules and the pore diameter 267 respectively. Mass transfer resistances on the permeate are considered negligible because ammonia 268 is rapidly converted into ammonium sulfate as a consequence of the excess concentration of sulfuric 269 acid in this stream. Therefore, the cross-sectional area of the lumen and shell sides, as well as the 270

<sup>271</sup> length of the membrane are estimated based on mass balances, as shown in Eqs. 23S-57S of the
<sup>272</sup> Supplementary Material respectively.

One or multiple membrane modules can be used to achieve the total membrane length (z)required to reach a certain efficiency, as shown in Eq. 6.  $n_{\text{series}}$  denotes the necessary number of membrane units of length  $L_{\text{module}}$  in-series arrangement to achieve the desired recovery efficiency. The length of the different membrane units is reported in Table 4.  $n_{\text{parallel}}$  refers to the number of membrane units in-parallel arrangement required to process the flow of organic material generated at the livestock facilities and processing capacities reported in Table 4.

Liquid-Cel<sup>T</sup> Extra Flow membranes (3M, 2021) have been considered for economic evaluation purposes since their use for ammonia recovery is widely reported in the literature (Darestani et al., 2017; Rongwong and Sairiam, 2020; Linstrom and Mallard, 2001). The characteristics of these membranes are collected in Table 7S of the Supplementary Material, while costs of membrane modules are shown in Table 4. If the capacity of the largest membrane module is not enough for the treatment of the organic feedstock, the installation of several parallel units is considered, as shown in Eq. 7.

$$n_{\rm series} = \left\lceil \frac{z}{L_{\rm module}} \right\rceil \tag{6}$$

$$n_{\text{parallel}} = \begin{cases} 1 & \text{if } \dot{V}_{\text{organic waste}} \leq 125 \frac{\text{m}^3}{\text{h}} \\ \left\lceil \frac{\dot{V}_{\text{organic waste}}\left(\frac{\text{m}^3}{h}\right)}{125} \right\rceil & \text{if } \dot{V}_{\text{organic waste}} > 125 \frac{\text{m}^3}{\text{h}} \end{cases}$$
(7)

286

The membrane module CAPEX is estimated through Eq. 60S, assuming a membrane lifetime ( $t_{\text{module}}$ ) of 10 years (Verrecht et al., 2010), and a plant lifetime ( $t_{\text{plant}}$ ) of 20 years. The use of sulfuric acid as stripping fluid and membrane cleaning is the main contributor to the membrane OPEX, as shown in Eq. 61S. The membrane cleaning cost ( $c_{\text{cleaning}}$ ) is reported to be between

Membrane module	Flow capacity (m3/h)	Diameter (m)	Length (m)	Cost (USD)
$2.5 \times 8$	0.1 - 0.7	0.067	0.200	5,000
$4 \times 13$	0.5 - 3.4	0.116	0.242	5,700
8x20	1 - 11	0.219	0.406	$14,\!150$
10x28	10 - 48	0.279	0.683	17,000
14x40	16 - 125	0.356	1.129	$24,\!300$

Table 4: Membrane modules size and cost (3M, 2021; SG Projects, 2021; DPC Water Solutions, 2021).

<sup>291</sup> 2% and 25% of the OPEX (Yu et al., 2020; Verrecht et al., 2010), for which we assume an average
<sup>292</sup> value of 13.5%. Additionally, the cost of the pumps needed for driving the digestate and stripping
<sup>293</sup> fluid streams through the membrane modules is considered. Cost estimation of pumps is collected
<sup>294</sup> in Eqs. 62S-66S of the Supplementary Material.

The sizing and cost estimation for the centrifuge unit are shown in Figure 4S of the Supplementary Material.

Total CAPEX and OPEX for the recovery of nitrogen from livestock digestate using a transmembrane chemisorption process result from the sum of membrane and pump costs are described in Eqs. 67S and 68S respectively.

*Evaporation and wet scrubbing.* Nitrogen can be recovered through ammonia evaporation, by 300 drying the digestate in a belt dryer unit. The operation of a belt dryer unit requires a minimum 301 concentration of solids of 10% - 12% (Bolzonella et al., 2018). Therefore, the liquid and solid outlet 302 streams from the solid-liquid separation stage are combined to obtain a stream with the desired 303 solids content. After a solids content adjustment, digestate is dried in the belt drier, as shown in 304 Fig 3d. This unit dries the digestate over the belt with a stream of hot air crossing the belt through 305 orifices on it. Heat is transferred from the hot air stream to the digestate on the belt to increase 306 temperature and evaporate ammonia and a fraction of the moisture. 307

The belt dryer model assumes the evaporation of two components (i.e., water and ammonia) in no equilibrium with a continuous extraction of the vapor phase (Treybal, 1980). Since the amount of ammonia in digestate is significantly lower than moisture content, air saturation with water vapor is considered the evaporation limit. It must be noted the moisture carrying capacity of air (i.e., the saturation point) is temperature dependent. Consequently, this requires to solve the mass and energy balances simultaneously, which are reported in the Supplementary Material Eqs. 69S-81S. An ammonia removal efficiency ( $\eta_{\text{belt dryer}}$ ) of 80% has been assumed for mass balance calculation (Awiszus et al., 2018a). Gaseous ammonia is further recovered through acidic scrubbing, as described below.

Capital expenses estimation for the belt dryer units are based on the energy required for am-317 monia evaporation, as shown in Eqs. 82S to 85S. A drying efficiency ( $\eta_{\text{belt dryer}}$ ) of 0.6 has been 318 assumed from the experimental work reported by Awiszus et al. (2018b). Belt dryer scale-up is 319 based on the correlation proposed by Towler and Sinnott (2012). The reference values and scale 320 factor used in this correlation are taken from costs and capacities reported by Turley et al. (2016), 321 as well as the maximum belt dryer capacity used to compute the number of dryer units needed 322  $(n_{\text{belt drver}})$ . These data are used to estimate the scale-up factor, which is estimated equal to 0.7. 323 Belt dryer operating costs are due to electrical consumption, which has been estimated in 0.099 kW 324 of electricity per kW of thermal energy used by the unit (Awiszus et al., 2018b). 325

The ammonia contained in the gaseous streams from ammonia evaporation can be recovered in an acidic scrubbing stage using a solution of sulfuric acid in water, as described in Fig. 3e. Ammonia is trapped in the liquid stream, reacting with the sulfuric acid to form ammonium sulfate. The mass balances for the scrubber unit are performed assuming the water transferred to the gas stream reach that saturation. A typical recovery efficiency for full-scale ammonia scrubbers of 96% is selected based on the work of Melse and Ogink (2005).

The water flow needed to perform the scrubbing operation is computed through the operating line of the unit (L/G). Following the general design rules for scrubbing units, the design operating line is assumed as twice the slope of the minimum operating line. The mass balances of the scrubbing columns are shown in Eqs. 113S-117S of the Supplementary Material. The amount of sulfuric acid supplied to make-up the sulfate used for ammonium sulfate formation is slightly larger than the stoichiometric amount of the precipitation reaction, 3.5 kg of H<sub>2</sub>SO<sub>4</sub> per kg of NH<sub>3</sub> recovered (Bolzonella et al., 2018).

<sup>339</sup> CAPEX of scrubber units is estimated through the column's volume by using the correlation

described in Eq. 106S. The estimation of the scrubber dimensions is collected in Eqs. 118S-120S 340 of the Supplementary Material. Scrubber diameter is based on the gas velocity in the equipment 341 (Melse and Ogink, 2005). The number of units is set by the maximum diameter of scrubbers, which 342 is assumed equal to 1.2 m accordingly to the general design rules for packed columns (Branan, 2005). 343 Similarly, to the case of stripping columns, the height of scrubbing towers is computed through the 344 transfer units method, as described by Couper et al. (2005). The cost of the compressor is estimated 345 through Eq. 127S. Operating expenses of scrubbing are mainly related to the use of sulfuric acid 346 and the compression cost. 347

Stripping and wet scrubbing. Nitrogen recovery by the stripping of the liquid phase of manure or digestate is a widely used technique based on the transfer of ammonia from liquid digestate to an air stream. An additional liquid-solid separation stage is required to enhance the removal of solids and avoid clogging issues in the stripping unit. A centrifuge is considered for this stage, assuming the separation coefficients reported by Møller et al. (2000).

A packed stripping tower is used for recovering the ammonia contained in the liquid stream, as illustrated in Figure 3e, and it is further recovered through acidic scrubbing, as described below. Two-inch (0.051 m) Intalox packing is considered (Strigle, 1994), which packing factor ( $F_P$ ) is assumed to be 18 ft<sup>-1</sup> (59 m<sup>-1</sup>) (Geankoplis, 2003). The packed stripping tower is modeled using the number of transfer units (NTU) method (Metcalf and Eddy, 2014). The pressure drop of the packed tower is estimated through the correlation proposed by Kister and Gill (1991), Eq. 87S.

The tower diameter is calculated through the tower flooding capacity. A tower flooding capacity 359 correlation considering the packing pressure drop was developed by using ALAMO (Wilson and 360 Sahinidis, 2017) based on the flooding curves developed by Strigle (1994), which is shown in Figure 361 4 and Eq. 8, where P is in inch H<sub>2</sub>O/ft,  $v_G$  denotes the superficial gas velocity in ft/s,  $\rho_G$  the gas 362 density in  $lb/tt^3$ ,  $\rho_L$  the liquid density in  $lb/tt^3$ ,  $\nu$  the kinematic viscosity in censtokes,  $G_G$  the gas 363 mass velocity in  $lb/ft^2 \cdot s$ , and  $G_L$  the liquid mass velocity in  $lb/ft^2 \cdot s$ . Two-inch (0.051 m) Intalox 364 packing is considered (Strigle, 1994), whose packing factor  $(F_P)$  is assumed to be 18 ft<sup>-1</sup> (59 m<sup>-1</sup>) 365 (Geankoplis, 2003). The operating line considered, defined as the ratio of gas and liquid volumetric 366

<sup>367</sup> flows, can be computed through Eqs. 88S to 91S.



Figure 4: Tower flooding capacity correlation considering the packing pressure drop. P is in  $inch H_2O/ft$ , Y and X axis are defined in Eqs. 9 and 10 respectively.

$$Y = -0.25 \cdot X + 0.22 \cdot \ln(P) - 0.78 \cdot 10^{-1} \cdot P^2 + 0.19 \cdot 10^{-1} \cdot X^3 - 0.39 \cdot X \cdot P$$
(8)

$$+ 0.49 \cdot 10^{-2} \cdot (X \cdot P)^3 + 0.89$$
$$Y = v_G \left(\frac{\rho_G}{\rho_L - \rho_G}\right)^{0.5} F_P^{0.5} \nu^{0.05}$$
(9)

$$X = \log_{10} \left( \frac{G_L}{G_G} \left( \frac{\rho_G}{\rho_L} \right)^{0.5} \right) \tag{10}$$

The liquid mass velocity is a known parameter since it corresponds to the digestate being processed. The gas velocity is estimated by combining the pressure drop and the flooding capacity of the packed tower, i.e., Eqs. 87S, 88S, and 91S. The design gas mass velocity considered is 0.7 times the theoretical gas mass velocity, Eq. 92S, while the liquid design mass velocity is computed by combining Eqs. 91S and 92S. Design restrictions reported by Branan (2005) are considered for the sizing of the packed tower. The tower height is estimated through the height and number of transfer units (Metcalf and Eddy, 2014), as described in the Supplementary Material, Eqs. 99S to
104S.

The number of stripping units needed  $(n_{\text{stripping tower}})$  is calculated as the product of the number of stripping units in-series arrangement to satisfy the packed towers height limit  $(n_{\text{stripping tower}}^{\text{series}})$ and the number of stripping units in-parallel arrangement to process the amount of waste generated in the livestock facility under evaluation  $(n_{\text{stripping tower}}^{\text{parallel}})$ . CAPEX of stripping packed towers is estimated based on the columns volume using a correlation based on data from CAPCOST (Turton, 2010), as shown Eq. 106S. Additionally, capital expenses of compressor units are estimated based on the correlation reported by Almena and Martín (2016), Eq. 108S.

The gaseous ammonia obtained from the stripping process is captured by wet scrubbing in a process similar to that described for the evaporation process. The sizing and cost estimation for the centrifuge unit are shown in Figure 4S of the Supplementary Material.

## 386 2.4. Economic assessment

The total costs for the treatment of livestock organic materials and for the recovery of nitrogen 387 are estimated for each nitrogen recovery system evaluated. These are defined in Eqs. 11 and 12 388 respectively for each evaluated process J, where k represents the possible products obtained, i 389 denotes the discount rate (assumed to be 7%), and  $n_{\text{plant}}$  represents the process lifetime, which is 390 assumed to be 20 years. We note that each nitrogen recovery process J is comprised by a set of j391 processing units, including the equipment from manure collection to the final recovery of nitrogen. 392 The estimation of the cost of nitrogen recovery includes OPEX and CAPEX amortization of all 393 equipment involved in the processing of livestock organic materials, as well as the potential incomes 394 from the sale of recovered products for those processes producing struvite or ammonium sulphate 395 (i.e, evaporation, stripping, and transmembrane chemisorption). The selling prices considered are 396 0.85 USD per kilogram of struvite (Molinos-Senante et al., 2011), and 0.12 USD per kilogram of 397 ammonium sulphate (Incro, 2021). Conversely, the liquid and organic solid effluents containing low 398 concentrations of nitrogen, such as the products obtained from the MAPHEX system, are considered 399 products with no market value. This assumption is based on the fact that, although they can be used 400

for nutrient supplementation in croplands, they are too bulky for being economically transported to
nutrient deficient areas. Therefore, similarly to manure, they can just be applied locally, hindering
their use as a bio-based substitute of synthetic nitrogen fertilizers.

$$Cost_{J}^{nitrogen}\left(\frac{\text{USD}}{\text{kg}_{N \text{ recovered}}}\right) = \sum_{j} \frac{OPEX_{j} + CAPEX_{j} \cdot \frac{i \cdot ((1+i)^{n}\text{plant})}{((1+i)^{n}\text{plant}-1)} - \sum_{k} \dot{m}_{j,k} \cdot Price_{k}}{\dot{m}_{N_{recovered}}}$$
(11)  
$$Cost_{J}^{processing}\left(\frac{\text{USD}}{\text{kg}_{Waste \text{ processed}}}\right) = \sum_{j} \frac{OPEX_{j} + CAPEX_{j} \cdot \frac{i \cdot ((1+i)^{n}\text{plant})}{((1+i)^{n}\text{plant}-1)} - \sum_{k} \dot{m}_{j,k} \cdot Price_{k}}{\dot{m}_{Waste_{processed}}}$$
(12)

# 404 3. Results and discussion

# 405 3.1. Nitrogen flows and recovery efficiency

Figure 5 shows the nitrogen flows of the evaluated systems considering the entire processes, including the pretreatment and final nitrogen recovery. These flows were analyzed to determine the fraction of nitrogen recovered as inorganic products, either in the form of ammonium sulphate solution or as struvite, the nitrogen contained in the side streams of the processes (usually comprised by organic solids), and the fraction of nitrogen not recovered and released into the environment.

MAPHEX is a manure processing system that integrates all the stages from the feed of raw 411 manure to the recovery of the final products combining different liquid-solid separation and chemical 412 coagulation stages (Church et al., 2018), and therefore no additional pretreatment stages are needed 413 for this system Figure 5a. 88.7% of the nitrogen is recovered within an organic solid material, 414 although with the market value of this product is limited due to the low concentration of nitrogen. 415 Struvite precipitation shows a low efficiency for nitrogen recovery as struvite (3.2%), Figure 5b, 416 since phosphate is the limiting compound for struvite precipitation. This is due to the fact that 417 this compound is in much lower concentrations than nitrogen in the organic materials released by 418 livestock, as shown in Table 1. As a result, a significant fraction of nitrogen is not recovered but 419 released in a liquid stream, similarly to ammonia evaporation process, as observed in 5b. 420

Nitrogen recovery by transmembrane chemisorpotion results in that 9.5% of nitrogen is removed
within the solid fraction obtained from the liquid-solid separation stage using a screw press unit. A
significant fraction of nitrogen, 58%, is recovered as a solution of ammonium sulphate, as illustrated
in Figure 5c.



(b) Struvite precipitation

Figure 5: Relative flows of inorganic nitrogen in the studied processes. Since a fraction of organic nitrogen in livestock manure is mineralized after the anaerobic digestion stage, the 100% refers to the inorganic nitrogen in digestate. The green color denotes the nitrogen recovered as part of an solid stream, purple color denotes the nitrogen recovered in the form of struvite, the blue color denotes the nitrogen recovered as ammonium sulphate, and the red color the non-recovered nitrogen.



(e) Stripping and wet scrubbing

Figure 5: Relative flows of inorganic nitrogen in the studied processes. Since a fraction of organic nitrogen in livestock manure is mineralized after the anaerobic digestion stage, the 100% refers to the inorganic nitrogen in digestate. The green color denotes the nitrogen recovered as part of an solid stream, purple color denotes the nitrogen recovered in the form of struvite, the blue color denotes the nitrogen recovered as ammonium sulphate, and the red color the non-recovered nitrogen (cont.).

The evaporation and scrubbing process, Figure 5d, requires an inlet stream with a certain solids content for the drying process in the belt dryer unit. Therefore, a solid adjustment must <sup>427</sup> be performed, discarding a large fraction of the liquid phase of digestate, which contains most of
<sup>428</sup> the inorganic nitrogen. As a result, a significant fraction of nitrogen, 79%, is released in a liquid
<sup>429</sup> stream.

The stripping and scrubbing process, shown in Figure 5e, requires of a liquid-solid separation pretreatment similar to the transmembrane chemisorption, resulting in a solid stream containing 9.5% of the nitrogen. 83.4% of nitrogen is recovered in the form of ammonium sulphate after the srubbing stage, while 7.1% of nitrogen is lost though the different processing stages.

As observed in Figure 5, nitrogen can be transferred into three streams, i.e., the nitrogen 434 recovered in the form of valuable products (struvite or ammonium sulphate), nitrogen removed 435 from the livestock waste through a solid stream in an analogous way to the nutrients removed from 436 the wastewater through sewage sludge, which can be used as soil amendment for agriculture, be 437 disposed, or further processes for resource and energy recovery, while another fraction of nitrogen 438 is not captured in any stage of the process and it is released into the environment through liquid 439 effluents or as gaseous ammonia. While the nitrogen recovered in the form of valuable products or 440 within an organic solid stream is captured and it can be managed, the nitrogen released into the 441 environment lead to harmful environmental effects (de Vries, 2021). 442

Attending to this taxonomy, struvite production result in the most limited nitrogen recovery 443 efficiency as valuable product, 3%, although the struvite obtained is a high value product since it 444 contains both phosphorus and nitrogen, and its potential as fertilizer has been extensively studied 445 (Vaneeckhaute et al., 2015). Transmembrane chemisorption and ammonia stripping result in the 446 best nitrogen recovery efficiencies as valuable product, 58% and 83% respectively, recovering the 447 nitrogen in the form of ammonium sulphate. MAPHEX transfers almost of 80% of nitrogen to an 448 organic solid stream, limiting the recycling of nitrogen to the use of this solid as soil amendment for 449 agriculture and hindering its transportation and redistribution, but limits the fraction of nitrogen 450 released to the environment to 11%. Finally, struvite production and ammonia evaporation are the 451 processes that result in the largest nitrogen releases into the environment, 93% and 79% respectively. 452

453 3.2. Economic assessment and scale-up

Nitrogen recovery and manure processing costs for each nitrogen management system evaluated 454 are estimated through Eqs. 11 and 12 for each evaluated technology J and each livestock waste, 455 considering that the livestock facilities of swine, dairy, beef, and poultry are comprised of animals 456 at different life stages, as described in Table 2. Two scenarios including or excluding the imple-457 mentation of AD have been studied, whose results are shown in Figures 6 and 7 respectively. The 458 estimation of the nitrogen recovery cost includes OPEX and CAPEX amortization of all equipment 459 involved in the processing of livestock organic materials, as well as incomes from the sales of stru-460 vite or ammonium sulphate. Conversely, we have assumed that that no incomes can be obtained 461 from the product generated from the MAPHEX system. In addition, generic correlations based on 462 inorganic nitrogen content in manure are developed to estimate the manure processing and nitro-463 gen recovery cost for any livestock waste that the flow of inorganic nitrogen contained in manure 464 is a known value are provided in Tables 5 and 6, which graphical representations can be found in 465 Figures 5S and 6S. 466

467 CAPEX and OPEX of the processes estimated in terms of inorganic nitrogen content in manure 468 for different scales are shown in the Supplementary Material, Figures 7S and 8S. The costs are 469 normalized by the amount of inorganic nitrogen contained in excreted manure. These can be 470 transformed into number of animals through the data reported in Table 1.





Figure 6: Processing and nitrogen recovery costs of the assessed nitrogen recovery technologies for different livestock facility sizes, including the cost of pretreatment and AD stages.

System	Correlation	$\begin{array}{c} \text{Manure processing cost} \\ \text{(USD/}_{kg_{manure \ processed}}) \end{array}$	Total nitrogen recovery cost $\binom{\text{USD}/\text{kg}_{N \text{ recovered}}}{}$		
		Parameters	Parameters		
MAPHEX	$C = a \cdot x^b$	a=0.163 b=-0.0372	a=20.439 b=-0.0372		
Struvite	$C = a \cdot x^b$	a=0.758	a=2942.411		
precipitation		b=-0.648	b=-0.648		
Transmembrane	$C = a \cdot x^b$	a=0.0971	a = 17.635		
chemisorption		b=-0.290	b = -0.284		
Evaporation &	$C = a \cdot x^b$	a=0.309	a=210.547		
scrubbing		b=-0.0234	b=-0.0234		
Stripping &	$C = a \cdot x^b$	a=0.253	a=33.814		
scrubbing		b=-0.0298	b=-0.0298		

Table 5: Correlations to estimate the processing cost of the evaluated technologies as a function of the nitrogen contained in as-excreted manure (x), including the cost of AD stage.

Considering the treatment cost per kilogram of livestock manure processed, we note the cost of 471 AD stage represents a large fraction of the CAPEX, as observed in Figures 7S and 8S. As a result, 472 lower treatment costs are achieved if AD is not considered, as observed by comparing Figures 473 6a and 7a. Focusing on the nitrogen recovery technologies, struvite precipitation and membrane 474 systems are the processes with the lowest processing cost, ranging between 0.15 to 0.02 USD per 475 kg of livestock manure processed. In addition, the influence of the economies of scale of these 476 two processes has a significant impact on the treatment cost of manure. The economies of scale 477 are particularly intense for small-scale facilities, up to 400 animal units. For larger facilities an 478 asymptotic behavior is observed, resulting in little improvements in terms of nitrogen recovery cost. 479 Conversely, the influence of the economies of scale is much lower for the other processes. 480

As a consequence of the effect of the economies of scale in the treatment cost of manure, nitrogen recovery is more economically feasible in the larger livestock operations, which represent the largest environmental threat, and it is hindered in the small-scale facilities, which are the predominant livestock operations in many regions (Lowder et al., 2016; FAO, 2013). Therefore, the development and implementation of modular and mobile nutrient recovery systems might play a crucial role
in regions where small-scale livestock operations are predominant for the treatment of manure at
affordable costs.





Figure 7: Processing and nitrogen recovery costs of the assessed nitrogen recovery technologies for different livestock facility sizes, excluding the cost of anaerobic digestion stage.

Although the treatment cost per kilogram of livestock manure treated is a common metric used 488 to measure and compare the processing costs of nitrogen recovery processes (De Vrieze et al., 2019; 489 Bolzonella et al., 2018) accounting for the operating expenses, amortized capital cost, and incomes 490 from the sales of recovered products, as shown in Eq. 12, it presents some deficiencies since it does 491 not include the nitrogen recovery efficiency of each process. This can lead to an inadequate compar-492 ison of processes since some of them might result in a low treatment cost but with a low nitrogen 493 recovery efficiency. Therefore, measuring the processing cost as a function of the recovered nitrogen, 494 as shown in Figures 6b and 7b, might be a more appropriate metric for comparing different systems. 495 Accordingly to this approach, the economic performance of struvite precipitation dramatically de-496 creases as a result of the low nitrogen recovery efficiency of this technology. Conversely, MAPHEX 497 is revealed as more competitive process when the nitrogen recovered is considered. Since MAPHEX 498 is a single size modular technology, its recovery cost shows a linear behavior, slightly affected by 499 adding extra in-parallel modules to process large amounts of organic material. Membrane system 500 is the process with the lowest nitrogen recovery cost, from 10.4 to 3.4 USD per kilogram of nitrogen 501 recovered, depending on the processing capacity of the system. In addition, it should be noted the 502 exclusion of the AD stage result in lower investment costs, but decrease the amount of inorganic 503 nitrogen available for recovery since the partial mineralization of the organic nitrogen contained in 504 manure achieved by AD does not occur in this scenario. As a consequence of the lower amount of 505 inorganic nitrogen available for recovery, the specific nitrogen recovery cost increases in the scenario 506 not including AD in spite of its lower equipment costs. 507

System	Correlation	$\frac{\text{Manure processing cost}}{(^{\text{USD}}/_{\text{kg}_{\text{manure processed}}})}$	$\frac{\text{Total nitrogen recovery cost}}{\binom{\text{USD}/\text{kg}_{\text{N recovered}}}{\text{Parameters}}}$
		Farameters	Farameters
Ammonia		a = 0.283	a = 278.944
evaporation		b = -0.00989	b = -0.00989
Struvite		a = 0.701	a = 3947.540
precipitation		b = -0.676	b = -0.676
MADUEY	$C = a m^b$	a = 0.163	a=29.634
MAFILLA	$C = u \cdot x$	b = -0.0372	b = -0.0372
Stripping in		a=0.226	a = 43.780
packed tower		b = -0.0128	b = -0.0128
Membrane		a = 0.0429	a = 11.427
		b = -0.185	b = -0.182

Table 6: Correlations to estimate the processing cost of the evaluated technologies as a function of the nitrogen contained in as-excreted manure (x), excluding the cost of anaerobic digestion stage.

Additionally, the environmental and social damage costs of releasing nitrogen to the environment 508 is illustrated in Figures 6b and 7b. The economic losses due to nitrogen releases have been estimated 509 based on the environmental and social cost of atmospheric NH<sub>3</sub> releases, and land, freshwater, and 510 groundwater nitrogen loading. The cost of nitrogen release considering these damages reported 511 by Sobota et al. (2015) and Compton et al. (2017) are 32.50 and 35.15 USD per kg of N released 512 respectively. Therefore, the recovery of nitrogen by using membrane systems and the MAPHEX 513 system result in economic savings with respect to nitrogen release to the environment. Although this 514 consideration might provide an economical support for nitrogen recovery from livestock manure, in 515 addition to the environmental arguments, the implementation of these systems result in additional 516 costs 0.016 to 0.32 USD per kg of manure processed, as shown in Figures 6 and 7. These costs 517 might make nitrogen recovery to be economically unfeasible for the operators of livestock facilities 518 and governmental economic support could be needed. 519

This information can be a driver for the deployment of livestock manure treatment processes for nitrogen recovery. However, a debate can be raised regarding what stakeholders should cover

the cost of nitrogen recovery from livestock industry. On the one hand, if nitrogen recovery is 522 not performed at livestock facilities, nitrogen releases result in environmental and social damage 523 costs. These costs are usually covered by national and regional governments, which are ultimately 524 funded by the taxpayers as a whole, including those individuals that are not are not stakeholders 525 in the livestock sector either as producers, distributors or consumers. On the other hand, the 526 implementation of nutrient recovery systems could impact the economy of livestock farms, which 527 in turn could result in the raise of livestock products cost, impacting the final consumers. This 528 approach might seem fairer since it only involves producers and consumers of livestock products. 529 However, it would lead to comparative disadvantages between different livestock farms as a result 530 of the savings in nitrogen recovery costs due to the economies of scale, as shown in Figures 6a 531 and 7a. Consequently, small facilities would be more affected by the costs nitrogen recovery than 532 large farms. Therefore, alternative economic schemes should be developed to mitigate the economic 533 impact of the implementation of nitrogen recovery systems at livestock facilities. In this regard, 534 previous efforts developed for phosphorus recovery at livestock facilities can be adapted for nitrogen 535 recovery. For instance, the development of a market for trading emissions allowances has been 536 proposed for phosphorus releases from livestock farms (Sampat et al., 2018b). This scheme might 537 be explored for nitrogen releases. Additionally, incentive policies for the implementation of nutrient 538 recovery systems in livestock facilities have been studied for the case of phosphorus, including the 539 fair allocation under limited incentive budgets scenarios (Martín-Hernández et al., 2022), which 540 could be adapted to the case of nitrogen recovery. 541

### 542 4. Conclusions

Intensive livestock operations generate vast amounts of organic materials in the form of manure, which is a source of nitrogen releases into the environment. Since these releases are significant contributors to the eutrophication of waterbodies, and they can result in harmful environmental impacts such as algal bloom episodes, the recovery of nitrogen at livestock facilities is a desirable measure to reduce the environmental footprint of the food production system.

548 Several processes have been proposed for nitrogen recovery from organic materials, and there-

fore the selection of the most suitable process has to be addressed. However, multiple dimensions 549 must be considered in the decision-making process, including the nitrogen recovery efficiency, the 550 capital and operating expenses, and the impact of the economies of scale in the final cost of nitrogen 551 recovery. A multi-scale techno-economic study has been performed to determine the most suitable 552 nitrogen recovery system based on the processing capacity. The mass flows throughout all stages 553 from manure collection to the final treatments have been analyzed to determine the nitrogen flows 554 throughout each studied system. Two metrics have been considered to measure the operating cost 555 of each technology, the manure treatment cost (USD/kg<sub>manure processed</sub>), that it is a metric widely 556 used in literature, and the nitrogen recovery cost (USD/kg<sub>N recovered</sub>). Since the first metric does not 557 account for the nitrogen recovery efficiency of each system, significant differences on the relative per-558 formance among the different technologies are found. This is because some technologies that result 559 in low manure treatment costs show low nitrogen recovery efficiencies, resulting in comparatively 560 large nitrogen recovery costs. However, transmembrane chemisorption is revealed as the most cost-561 effective nitrogen recovery technology, resulting in costs of 0.02-0.06 USD/kg<sub>manure processed</sub>, and 562  $3.4-10.4 \text{ USD/kg}_{N \text{ recovered}}$ . Moreover, comparing the negative economic impact of nitrogen releases 563 into the environment, estimated between 32.50 and  $35.15 \text{ USD/kg}_{N \text{ released}}$  with the cost of nitrogen 564 recovery, three technologies reveal to be economically advantageous, transmembrane chemisorption, 565 MAPHEX, and stripping in packed bed. 566

It should be noted that the performance of nitrogen recovery processes might be affected by the 567 content of total solids, carbonates, calcium, and other salts in raw and digested livestock waste. 568 The presence of these compounds might negatively affect different aspects of the performance 569 of the recovery processes, including the formation of undesired precipitates, excessive membrane 570 fouling, reduction of mass and heat transfer, among others. However, specific information for the 571 formation of these compounds from manure and digestate is limited, specially in comparison with 572 the information available for municipal wastewater treatment. Therefore, the study of the influence 573 of these parameters in the performance of nutrient recovery processes requires of further efforts to 574 achieve a better understanding of the recovery processes and the boundaries of their technical and 575 economic feasibility. 576

Future research is needed for the evaluation of all the emissions of the nitrogen recovery processes 577 through the life-cycle assessment framework in order to include the environmental dimension in 578 the decision-making process of selecting the most suitable nitrogen recovery system from a multi-579 criteria perspective. Additional lines of work include the discussion about what stakeholders in 580 the production and consumption cycle should assume the costs associated with nitrogen recovery. 581 Additionally, further studies have to be addressed to design and evaluate incentive policies for 582 the effective deployment of combined phosphorous-nitrogen recovery systems at intensive livestock 583 operations. 584

#### 585 5. Acknowledgments

E.M.H. and M.M. acknowledge funding from Junta de Castilla y León, Spain, under grant EDU/556/2019. C.M.R. acknowledge funding from the University of Salamanca TCUE Universidad-Empresa programme.

Disclaimer: The views expressed in this article are those of the authors and do not necessarily reflect the views or policies of the U.S. Environmental Protection Agency. Mention of trade names, products, or services does not convey, and should not be interpreted as conveying, official U.S. EPA approval, endorsement, or recommendation.

# 593 References

- <sup>594</sup> 3M, 2021. 3M<sup>™</sup>Liqui-Cel<sup>™</sup>Data Sheets. Technical Report. 3M.
- <sup>595</sup> Al Saedi, T., Rutz, D., Prassl, H., Köttner, M., Finsterwalder, T., Volk, S., Janssen, R., 2008.
   <sup>596</sup> Biogas Handbook. University of Southern Denmark, Esbjerg.
- <sup>597</sup> Almena, A., Martín, M., 2016. Technoeconomic analysis of the production of epichlorohydrin from
   <sup>598</sup> glycerol. Industrial & Engineering Chemistry Research 55, 3226–3238.
- <sup>599</sup> AMPC, 2018. Struvite or Traditional Chemical Phosphorus Precipitation What Option Rocks?
- https://www.ampc.com.au/uploads/cgblog/id408/2018-1026\_-\_Final\_Report.pdf. [On-
- line; accessed 20-March-2019].

- Astals, S., Martínez-Martorell, M., Huete-Hernández, S., Aguilar-Pozo, V., Dosta, J., Chimenos,
   J., 2021. Nitrogen recovery from pig slurry by struvite precipitation using a low-cost magnesium
   oxide. Science of The Total Environment 768, 144284.
- Awiszus, S., Meissner, K., Reyer, S., Müller, J., 2018a. Ammonia and methane emissions during
   drying of dewatered biogas digestate in a two-belt conveyor dryer. Bioresource Technology 247,
   419–425.
- Awiszus, S., Meissner, K., Reyer, S., Müller, J., 2018b. Utilization of digestate in a convective hot
   air dryer with integrated nitrogen recovery. Landtechnik 73, 106–114.
- Baker, D., Da Silva, C., 2014. Trends in agri-food systems: Drivers, changes, impacts and overall
   assessment. Technical Report. FAO Policy Learning Programme.
- Beckinghausen, A., Odlare, M., Thorin, E., Schwede, S., 2020. From removal to recovery: An
   evaluation of nitrogen recovery techniques from wastewater. Applied Energy 263, 114616.
- <sup>614</sup> Beddoes, J.C., Bracmort, K.S., Burns, R.T., Lazarus, W.F., 2007. An Analysis of Energy Produc-
- tion Costs from Anaerobic Digestion Systems on U.S. Livestock Production Facilities. Technical
- 616 Report. U.S. Department of Agriculture.
- 617 Bolzonella, D., Fatone, F., Gottardo, M., Frison, N., 2018. Nutrients recovery from anaerobic
- digestate of agro-waste: Techno-economic assessment of full scale applications. Journal of Environmental Management 216, 111–119.
- Bouwman, A., Beusen, A.H., Billen, G., 2009. Human alteration of the global nitrogen and phos phorus soil balances for the period 1970–2050. Global Biogeochemical Cycles 23.
- <sup>622</sup> Branan, C.R., 2005. Rules of Thumb for Chemical Engineers (Fourth Edition). Fourth edition ed.,
- Gulf Professional Publishing, Burlington. doi:https://doi.org/10.1016/B978-075067856-8/
- <sup>624</sup> 50000-1.
- <sup>625</sup> Brunner, P.H., Rechberger, H., 2016. Handbook of material flow analysis: For environmental, <sup>626</sup> resource, and waste engineers, Second Edition (2nd ed.). CRC press.

- Burk, C., 2018. Techno-economic modeling for new technology development. Chemical Engineering
   Progress 114, 43–52.
- <sup>629</sup> Church, C.D., Hristov, A.N., Bryant, R.B., Kleinman, P.J.A., Fishel, S.K., 2016. A Novel Treatment
   <sup>630</sup> System to Remove Phosphorus from Liquid Manure. Applied Engineering in Agriculture 32,
- <sup>631</sup> 103-112. URL: http://elibrary.asabe.org/abstract.asp?aid=46616&t=3&dabs=Y&redir=
- 632 &redirType=, doi:10.13031/aea.32.10999.
- 633 Church, C.D., Hristov, A.N., Kleinman, P.J., Fishel, S.K., Reiner, M.R., Bryant, R.B., 2018. Ver-
- satility of the MAnure PHosphorus EXtraction (MAPHEX) System in Removing Phosphorus,
- Odor, Microbes, and Alkalinity from Dairy Manures: A Four-Farm Case Study. Applied En-
- gineering in Agriculture 34, 567-572. URL: http://elibrary.asabe.org/abstract.asp?AID=
- <sup>637</sup> 48976&t=3&dabs=Y&redir=&redirType=, doi:10.13031/aea.12632.
- <sup>638</sup> Ciborowski, P., 2001. Anaerobic Digestion of Livestock Manure for Pollution Control and Energy
- <sup>639</sup> Production: A Feasibility Assessment. Technical Report. Minnesota Pollution Control Agency.
- <sup>640</sup> Clarke Energy, 2013. CHP efficiency for biogas. Technical Report. Clarke Energy.
- <sup>641</sup> Compton, J.E., Leach, A.M., Castner, E.A., Galloway, J.N., 2017. Assessing the social and en <sup>642</sup> vironmental costs of institution nitrogen footprints. Sustainability: The Journal of Record 10,
   <sup>643</sup> 114–122.
- <sup>644</sup> Couper, J.R., Penney, W.R., Fair, J.R., Walas, S.M., 2005. Chemical process equipment: selection
   <sup>645</sup> and design. Gulf Professional Publishing.
- Darestani, M., Haigh, V., Couperthwaite, S.J., Millar, G.J., Nghiem, L.D., 2017. Hollow fibre
   membrane contactors for ammonia recovery: Current status and future developments. Journal
   of Environmental Chemical Engineering 5, 1349–1359.
- <sup>649</sup> De Vrieze, J., Colica, G., Pintucci, C., Sarli, J., Pedizzi, C., Willeghems, G., Bral, A., Varga, S.,
- <sup>650</sup> Prat, D., Peng, L., et al., 2019. Resource recovery from pig manure via an integrated approach:

- A technical and economic assessment for full-scale applications. Bioresource Technology 272, 582–593.
- <sup>653</sup> DPC Water Solutions, 2021. Debubblers. Technical Report. DPC Water Solutions.
- Fachagentur Nachwachsende Rohstoffe, 2010. Biogas Guide. From production to use. Technical
   Report.
- Fangueiro, D., Snauwaert, E., Provolo, G., Hidalgo, D., Adani, F., Kabbe, C., Bonmati, A.,
  Brandsma, J., 2020. Mini-paper Available technologies for nutrients recovery from animal
  manure and digestates. Technical Report. EIP-AGRI Focus Group-Nutrient recycling.
- <sup>659</sup> FAO, 2004. The ethics of sustainable agricultural intensification. FAO, Rome.
- FAO, 2013. 2000 world census of agriculture. analysis and international comparison of the results
   (1996–2005).
- Geankoplis, C.J., 2003. Transport processes and separation process principles: (includes unit oper ations). Prentice Hall Professional Technical Reference.
- Ha, T.H., Mahasti, N.N., Lu, M.C., Huang, Y.H., 2023. Ammonium-nitrogen recovery as struvite
   from swine wastewater using various magnesium sources. Separation and Purification Technology
   308, 122870.
- <sup>667</sup> Incro, 2021. Ammonium sulphate price. Personal communication.
- Kahiluoto, H., Pickett, K.E., Steffen, W., 2021. Global nutrient equity for people and the planet.
   Nature Food 2, 857–861.
- Kar, S., Singh, R., Gurian, P.L., Hendricks, A., Kohl, P., McKelvey, S., Spatari, S., 2023. Life
- cycle assessment and techno-economic analysis of nitrogen recovery by ammonia air-stripping
- from wastewater treatment. Science of The Total Environment 857, 159499.
- <sup>673</sup> Kister, H., Gill, D., 1991. Predict flood point and pressure drop for modern random packings.
  <sup>674</sup> Chemical Engineering Progress 87, 32–42.

- 475 Lamolinara, B., Pérez-Martínez, A., Guardado-Yordi, E., Fiallos, C.G., Diéguez-Santana, K., Ruiz-
- Mercado, G.J., 2022. Anaerobic digestate management, environmental impacts, and techno economic challenges. Waste Management 140, 14–30.
- Linstrom, P.J., Mallard, W.G., 2001. The nist chemistry webbook: A chemical data resource on
   the internet. Journal of Chemical & Engineering Data 46, 1059–1063.
- Lowder, S.K., Skoet, J., Raney, T., 2016. The number, size, and distribution of farms, smallholder
   farms, and family farms worldwide. World Development 87, 16–29.
- Lübken, M., Wichern, M., Schlattmann, M., Gronauer, A., Horn, H., 2007. Modelling the energy
  balance of an anaerobic digester fed with cattle manure and renewable energy crops. Water
  research 41, 4085–4096.
- Ludington, D., 2013. Calculating the Heating Value of Biogas. Technical Report. DLtech, Inc.
- Martín-Hernández, E., Hu, Y., Zavala, V.M., Martín, M., Ruiz-Mercado, G.J., 2022. Analysis of
- incentive policies for phosphorus recovery at livestock facilities in the great lakes area. Resources,
   Conservation and Recycling 177, 105973.
- Martín-Hernández, E., Ruiz-Mercado, G.J., Martín, M., 2020. Model-driven spatial evaluation
   of nutrient recovery from livestock leachate for struvite production. Journal of Environmental
   Management 271, 110967.
- Melse, R.W., Ogink, N., 2005. Air scrubbing techniques for ammonia and odor reduction at livestock
   operations: Review of on-farm research in the Netherlands. Transactions of the ASAE 48, 2303–
   2313.
- Metcalf, Eddy, 2014. Wastewater Engineering: Treatment and Resource Recovery. McGraw-Hill,
   New York.
- Molinos-Senante, M., Hernández-Sancho, F., Sala-Garrido, R., Garrido-Baserba, M., 2011. Eco nomic feasibility study for phosphorus recovery processes. Ambio 40, 408–416.

- Møller, H., Lund, I., Sommer, S., 2000. Solid-liquid separation of livestock slurry: Efficiency and
   cost. Bioresour. Technol. 74, 223–229.
- Munasinghe-Arachchige, S.P., Nirmalakhandan, N., 2020. Nitrogen-fertilizer recovery from the
   centrate of anaerobically digested sludge. Environmental Science & Technology Letters 7, 450–
   459.
- Pagliari, P.H., Laboski, C.A., 2012. Investigation of the inorganic and organic phosphorus forms in
   animal manure. Journal of Environmental Quality 41, 901–910.
- Penn State Extension, 2016. Agricultural Anaerobic Digesters: Design and Operation. Technical
   Report. Penn State University.
- Rongwong, W., Sairiam, S., 2020. A modeling study on the effects of pH and partial wetting
  on the removal of ammonia nitrogen from wastewater by membrane contactors. Journal of
  Environmental Chemical Engineering 8, 104240.
- Ryckebosch, E., Drouillon, M., Vervaeren, H., 2011. Techniques for transformation of biogas to
   biomethane. Biomass and Bioenergy 35, 1633–1645.
- Sampat, A.M., Martin-Hernandez, E., Martín, M., Zavala, V.M., 2018a. Technologies and logistics
  for phosphorus recovery from livestock waste. Clean Technologies and Environmental Policy 20,
  1563–1579.
- Sampat, A.M., Ruiz-Mercado, G.J., Zavala, V.M., 2018b. Economic and environmental analysis for
  advancing sustainable management of livestock waste: A Wisconsin Case Study. ACS Sustainable
  Chemistry & Engineering 6, 6018–6031.
- <sup>719</sup> SG Projects, 2021. Gas Transfer Membranes. Technical Report. SG Projects.
- <sup>720</sup> Sobota, D.J., Compton, J.E., McCrackin, M.L., Singh, S., 2015. Cost of reactive nitrogen release
- from human activities to the environment in the United States. Environmental Research Letters
  10, 025006.

- Statistics Canada Statistique Canada, 2022. Census of Agriculture. https://www.statcan.gc.
   ca/en/census-agriculture. [Online; accessed 03-Ocotober-2022].
- Strigle, R.F., 1994. Packed tower design and applications: random and structured packings. Gulf
  Pub Co.
- <sup>727</sup> Tao, W., Fattah, K., Huchzermeier, M., 2016. Struvite recovery from anaerobically digested dairy
- manure: A review of application potential and hindrances. Journal of Environmental Management
   169, 46–57.
- Towler, G., Sinnott, R., 2012. Chemical engineering design: principles, practice and economics of
   plant and process design. Elsevier.
- <sup>732</sup> Treybal, R.E., 1980. Mass transfer operations. New York 466.
- Turley, D., Hopwood, L., Burns, C., Di Maio, D., 2016. Assessment of Digestate Drying as an
  Eligible Heat Use in the Renewable Heat Incentive. Technical Report. NNFCC.
- Turton, R., 2010. CAPCOST software to accompany: Analysis, synthesis, and design of chemical
   processes. Technical Report. Upper Saddle River, N.J: Prentice Hall PTR.
- <sup>737</sup> U.S. Department of Agriculture, 2000. Manure Nutrients Relative to the Capacity of Cropland and

Pastureland to Assimilate Nutrients. Technical Report. United States Department of Agriculture.

- <sup>739</sup> U.S. Department of Agriculture, 2009. Waste Management Field Handbook. Technical Report.
- <sup>740</sup> U.S. Department of Agriculture, 2011. Animal Feeding Operations (AFO) and Concentrated
- Animal Feeding Operations (CAFO). https://www.nrcs.usda.gov/wps/portal/nrcs/main/
- national/plantsanimals/livestock/afo/. [Online; accessed 10-August-2020].
- Vaneeckhaute, C., Janda, J., Meers, E., Tack, F., 2015. Efficiency of soil and fertilizer phosphorus
- <sup>744</sup> use in time: A comparison between recovered struvite, fepo 4-sludge, digestate, animal manure,
- <sup>745</sup> and synthetic fertilizer. Nutrient use efficiency: from basics to advances, 73–85.

- <sup>746</sup> Verrecht, B., Maere, T., Nopens, I., Brepols, C., Judd, S., 2010. The cost of a large-scale hollow
  <sup>747</sup> fibre MBR. Water Research 44, 5274–5283.
- <sup>748</sup> de Vries, W., 2021. Impacts of nitrogen emissions on ecosystems and human health: A mini review.
  <sup>749</sup> Current Opinion in Environmental Science & Health 21, 100249.
- Wilson, Z.T., Sahinidis, N.V., 2017. The alamo approach to machine learning. Computers &
   Chemical Engineering 106, 785–795.
- Yu, H., Li, X., Chang, H., Zhou, Z., Zhang, T., Yang, Y., Li, G., Ji, H., Cai, C., Liang, H., 2020.
  Performance of hollow fiber ultrafiltration membrane in a full-scale drinking water treatment
  plant in china: a systematic evaluation during 7-year operation. Journal of Membrane Science
  613, 118469.