

 Open access • Journal Article • DOI:10.1007/S12649-016-9642-X

## **Nutrient recovery from digestate : systematic technology review and product classification** — [Source link](#)

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**Institutions:** Laval University, Ghent University

**Published on:** 13 Jun 2017 - Waste and Biomass Valorization (Springer Netherlands)

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2 **Systematic technology review and product classification**

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34

35 **Abstract**

36 Nutrient recovery from digested biodegradable waste as marketable products has become an  
37 important task for anaerobic digestion plants to meet both regulatory drivers and market demands,  
38 while producing an internal revenue source. As such, the present waste problem could be turned  
39 into an economic opportunity. The aim of this study was to provide a comprehensive overview and  
40 critical comparison of the available/emerging technologies for nutrient recovery from digestate, and  
41 a classification of the resulting end-products according to their fertilizer characteristics. Based on  
42 the stage of implementation, the technical performance, as well as financial aspects, struvite  
43 precipitation/crystallization, ammonia stripping and (subsequent) absorption using an acidic air  
44 scrubber were selected as best available technologies to be applied at full-scale for nutrient  
45 recovery as marketable fertilizer commodities. The resulting end-products can and should be  
46 classified as renewable N/P-precipitates and N/S-solutions, respectively, in fertilizer and  
47 environmental legislations. This would stimulate their use and foster nutrient recovery technology  
48 implementation.

49

50 **Key-words:** anaerobic digestion; bio-based fertilizers; residuals management; sustainable  
51 agriculture; sustainable resource management; waste valorization.

52

53 **1. Introduction**

54 In the transition from a fossil reserve-based to a bio-based economy, it has become a critical  
55 challenge to maximally close nutrient cycles and migrate to a more effective and sustainable  
56 resource management, both from an economic as an ecological perspective.<sup>1-5</sup> Medium (2020) and

57 long-term (2050) strategic environmental policy objectives are being or have been set across the  
58 world in order to support the growth of a more innovative, resource-efficient economy, based on  
59 the sustainable production of bio-based products (bio-energy and bio-materials) from renewable  
60 biomass sources.<sup>6-9</sup> In the framework of these objectives, the anaerobic (co-)digestion of sewage  
61 sludge, organic biological waste (crop residues and other food waste), and animal manure has  
62 been evaluated as one of the most energy-efficient and environmentally friendly technologies for  
63 bio-energy production, organic biodegradable waste valorization, and potential recovery of valuable  
64 nutrient resources, which are concentrated in the remaining (mineralized) digestate.<sup>9-11</sup> Despite its  
65 high potential, the further sustainable development of this technology is currently hindered,  
66 especially in high-nutrient regions, because these digestates can often not or only sparingly be  
67 returned to agricultural land in their crude unprocessed form. This technical barrier is mainly posed  
68 by legislative constraints (strict nitrogen (N) and phosphorus (P) fertilization levels in the framework  
69 of environmental legislations), as well as practical (large volumes) and economic (high  
70 transportation and storage costs) complications.<sup>12-13</sup> In many regions an important legislative  
71 bottleneck for the beneficial use of digestate derivatives is that, if the biodegradable material fed  
72 into the digester contains any waste, the digestate produced and its derivatives would normally be  
73 classified as waste and be subject to waste regulation controls. Moreover, in the European Union,  
74 all derivatives produced from animal manure, including digestates, are still categorized as animal  
75 manure in fertilizer legislation and treated accordingly. Hence, further processing of digestate is  
76 required in order to concentrate and recover the nutrients as high-quality end-products, thereby  
77 overcoming the obstacles related to the direct application of the product.

78         So far, the technical approach for digestate processing was similar to the approach for the  
79 treatment of manure and wastewater. This means that the focus was on little cost-effective,  
80 energy-intensive, and non-sustainable nutrient removal practices through destruction or emission,

81 e.g. biological nitrification/denitrification.<sup>13</sup> The challenge for anaerobic digestion plants now is to  
82 achieve optimal recovery and recycling of nutrients from the digestate in a sustainable way. As  
83 such, regulatory drivers can be met and an internal revenue source can be produced, i.e. the  
84 present 'waste' problem can be turned into an economic opportunity.

85 The selection of the nutrient recovery technology (NRT) depends on the input waste  
86 stream characteristics and has a strong influence on the composition and properties of the  
87 resulting fertilizer end-product and by-products. Understanding the fundamentals of the existing  
88 processes is thus of paramount importance to sustainably create new high-quality fertilizers.  
89 Contemporary knowledge on NRT's and product quality is spread over a handful of academic and  
90 industrial experts. Reviews on the potential of particular technologies have been published<sup>5,14-18</sup>,  
91 e.g. struvite crystallization<sup>14</sup>, microalgae production<sup>15</sup>, membranes<sup>16</sup>, and on P recovery only<sup>17-18</sup>,  
92 but a comprehensive overview is lacking. Moreover, a shortcoming of many research articles and  
93 reviews on nutrient recovery is the lack of attention given to the quality, value, and demand for the  
94 final nutrient product. Because of these flaws, the use of recovered bio-based fertilizers is currently  
95 not or not sufficiently encouraged in environmental legislations (mostly these products are still  
96 classified as waste, see above), although some of them have similar properties as conventional  
97 fossil reserve-based chemical fertilizers.<sup>19-22</sup> In turn, these legislative bottlenecks hinder the  
98 marketing and efficient use of bio-based products.

99 The aim of this review paper is to provide a systematic overview and a critical comparison  
100 of technologies for the recovery of macronutrients from digestate, as well as a classification of the  
101 resulting end-products based on their fertilizer characteristics. The focus is on the recovery of N, P,  
102 and potassium (K), but parallel attention is given to sulfur (S), calcium (Ca), and magnesium (Mg).  
103 The systematic review involves an exhaustively search of all the relevant peer-reviewed literature,

104 as well as grey literature and unpublished research findings. Moreover, an intensive discussion  
105 platform was established involving existing international academic experts, administrations, and  
106 companies active in the field of nutrient recovery. Included and excluded data were discussed with  
107 the expert panel and selected on the basis of predefined criteria. Financial and technical aspects  
108 for the most established technologies at full-scale were further investigated by requesting budget  
109 proposals using a predefined questionnaire to key technology suppliers in the field. The survey  
110 involved capital and operational costs, use of consumables, recovered product quality, and  
111 potential revenues, among other technical items.

112 Based on the findings, first the general composition of digestate is briefly discussed  
113 (Section 2). In the core of this paper, the technical and economic state-of-the-art of the existing  
114 technologies and those under development is explored (Section 3), and available information on  
115 product quality and value is compiled (Section 4.1). Market trends and outlook are also discussed  
116 (Section 4.2). As such, this review paper can provide the fundamental basis to classify and  
117 categorize recycled products in environmental and fertilizer legislations, thereby stimulating their  
118 economic valorization as marketable commodities. This, in turn, may foster the development and  
119 implementation of innovative technologies for nutrient recovery from digestate.

120

## 121 **2. Digestate characteristics**

122 Digestate is the remaining product after biogas production in an anaerobic digester. It contains the  
123 non-digested recalcitrant organic fraction, water, micro- and macronutrients.<sup>19,21-23</sup> The composition  
124 of digestate varies strongly according to the composition of the feedstock (biodegradable waste)  
125 that is digested. Hence, giving a standard composition of digestate is not possible. Because of this  
126 constraint, 213 digestates from different (co-)digestion plants in Flanders (Northern part of Belgium  
127 confronted with high nutrient pressure) were sampled and analyzed during four years (2008-

128 2011).<sup>24</sup> Product quality ranges are compiled in Supporting Information 1. Based on the results, a  
129 short overview of how physicochemical characteristics change during the digestion process and  
130 how the feedstock influences the digestate composition is given below.

131 During anaerobic digestion, easily degradable organic matter is converted into methane  
132 (CH<sub>4</sub>) and carbon dioxide (CO<sub>2</sub>), while complex organic matter, such as lignin, remains in the  
133 digestate, thereby increasing its amount of effective organic carbon (OC). This is the OC that  
134 remains in the soil after one year and thus contributes to the humus built-up (average: 33.7 kg ton<sup>-1</sup>  
135 in digestate vs. 20.0 kg ton<sup>-1</sup> in pig manure on fresh weight (FW)).<sup>24</sup> As such, the digestate contains  
136 important soil improving qualities.<sup>25</sup> The percentage of organic dry matter can vary between 30 and  
137 80 %, with lower values for increasing slurry fractions and higher values for increasing fractions of  
138 kitchen and garden waste (KGW). The average dry matter content of 100 % KGW was estimated  
139 at 21 %, <sup>25</sup> whereas the median dry matter content of the 213 studied digestates amounted to 8.7 %  
140 (Supporting Information 1).

141 Due to the degradation of more than 90 % of volatile fatty acids (VFA's), the pH is  
142 increased and odor emissions are significantly lower during the application of digestate on  
143 agricultural fields as compared to pig slurry.<sup>26</sup> The pH of slurry is on average 7.1<sup>25</sup>, whereas the  
144 median digestate pH amounts to 8.3 (Table 1). However, the higher pH causes an increased risk  
145 for NH<sub>3</sub> volatilisation. This is why injecting the digestate is strongly advised.<sup>25</sup>

146 Next, during the digestion process, organically bound N is released as ammonium (NH<sub>4</sub><sup>+</sup>),  
147 which is directly available for crop uptake. The higher the share of NH<sub>4</sub>-N, the higher the efficiency  
148 of the digestate as a N-fertilizer. An input stream with a high N-level is pig slurry (average: 6.78 kg  
149 N ton<sup>-1</sup> FW), in comparison to cattle slurry (3.75 kg N ton<sup>-1</sup> FW) and maize (4.00 kg N ton<sup>-1</sup> FW)<sup>25</sup>.  
150 The Vlaco (2012)<sup>24</sup> data showed a median total N-content of 4.2 kg N ton<sup>-1</sup> FW (Table 1). When  
151 digesting raw pig slurry, more than 80 % of the N becomes available as NH<sub>4</sub><sup>+</sup>. However, for

152 digestates produced from organic waste such as KGW, the share of  $\text{NH}_4^+$  is often not higher than  
153 44-47 %, which is even lower than the value for raw pig slurry ( $\pm 60$  %). Digestates with a low  $\text{NH}_4$ -  
154 N-content are mostly originating from organic food/industrial wastes, including KGW.<sup>25</sup>

155 Furthermore, the total P-content of the input streams is not changed during the digestion  
156 process, but the organically bound P becomes available for the plant during digestion. Pig slurry  
157 has a high  $\text{P}_2\text{O}_5$ -content of about  $5 \text{ kg ton}^{-1}$ . By adding co-products to pig slurry the  $\text{P}_2\text{O}_5$ -content of  
158 the digestate is somewhat lowered. The 213 studied digestates showed a median total  $\text{P}_2\text{O}_5$ -  
159 content of  $3.9 \text{ kg ton}^{-1}$  FW (Supporting Information 1).

160 Also the total contents of K, Ca, Mg, and heavy metals are not altered during anaerobic  
161 digestion. K, Ca, and Mg become soluble. Zinc (Zn) and copper (Cu) contents in the digestate can  
162 become critically high, especially during the digestion of 100 % pig slurry, as the dry matter content  
163 decreases. This can hinder the beneficial reuse of recovered products, although both elements are  
164 essential micronutrients for healthy plant growth.<sup>27</sup>

165 Finally, impurities such as weed seeds and pathogens can be killed off during the digestion  
166 process. The extent to which this inactivation is sufficient depends entirely on the temperature and  
167 residence time in the digester and on the type of organism.<sup>25</sup>

168

### 169 **3. Technology overview**

#### 170 **3.1 Digestate processing technologies**

171 In the past decade, a diverse range of technologies that can be applied for digestate processing  
172 has been developed. However, certainly not all of them can be considered as a nutrient recovery  
173 technology (NRT). To date, there is no straightforward definition of an NRT. In this review we  
174 consider an NRT as a process that: 1) creates an end-product with higher nutrient concentrations



175 than the crude digestate (= concentrated product that contains both minerals and organics), or 2)  
176 separates the envisaged nutrients from organic compounds, with the aim to produce an end-  
177 product that is fit for use in the chemical or fertilizer industry or as a chemical fertilizer substitute.  
178 The breakthrough of such technologies would make it possible to reuse reactive nutrients locally  
179 and close nutrient cycles in a cross-sectorial cradle-to-cradle approach (Supporting Information 2).  
180 An overview of existing digestate processing technologies is presented in Figure 1. The  
181 technologies delineated as NRT are indicated in shaded boxes. It must be noted that these  
182 technologies could also be applied on undigested manure, sludge, and wastewater. However, as  
183 mentioned above, implementing anaerobic digestion as an intermediate step can highly improve  
184 the overall process efficiency. In some cases opportunities for direct reuse, e.g. through fertigation  
185 or reuse of nutrients via irrigation with (waste)water containing nutrients, may also exist.<sup>28-29</sup>

186

187 **Figure 1:** Schematic overview of digestate processing technologies. Shaded boxes: nutrient  
188 recovery technology (NRT).

189

190 In general, digestate processing starts with a mechanical separation into a liquid fraction and solid  
191 fraction.<sup>30</sup> Most of the N and K ends up in the liquid fraction, while most of the recalcitrant organic  
192 matter, P, Ca, and Mg is recovered in the solid fraction.<sup>19</sup> From literature, the technologies for  
193 nutrient recovery from the liquid fraction available or under development today are: 1) chemical  
194 crystallization<sup>14,31</sup>, 2) gas stripping and absorption<sup>32-34</sup>, 3) acidic air scrubbing<sup>32,35</sup>, 4) membrane  
195 separation<sup>36-38</sup>, 5) ammonia sorption<sup>39-41</sup>, and 6) biomass production and harvest<sup>42,43</sup>. For the solid  
196 fraction, only P extraction from ashes produced by combustion or pyrolysis can be considered a  
197 potential NRT.<sup>44,45</sup> However, as it is critically questioned whether incineration is a sustainable

198 technology, the interest is growing to extract nutrients from the crude digestate, thereby eliminating  
199 the solid-liquid separation step or producing an organic, P-poor thick fraction.

200 For extractive nutrient recovery to become a viable option, the process must have an  
201 equivalent treatment efficiency as conventional treatment, the process must be cost-effective, the  
202 process must be simple to operate and maintain, and most importantly, there must be a market for  
203 the recovered nutrient products. The technical and economic state-of-the-art of the above  
204 mentioned NRT's is discussed below. Product quality and fertilizer markets are discussed in  
205 section 4. For convenience of discussion, all costs are expressed in euros (€), with the original  
206 currency unit in parenthesis.

207

## 208 **3.2 Phosphorus precipitation and crystallization (NRT 1)**

### 209 **3.2.1 Struvite recovery**

210 Nutrient recovery through P precipitation and crystallization is a mature technology, mostly  
211 involving the addition of Mg ( $MgO/MgCl_2$ ) to a solution containing soluble  $PO_4$ -P (ortho-P) and  
212 ammonium, thereby increasing the pH to 8.3-10 and inducing the precipitation of struvite,  
213  $MgNH_4PO_4 \cdot 6H_2O$ .<sup>14</sup> The process has been implemented at full-scale installations for wastewater,  
214 (digested) sludge, and manure treatment, as well as at pilot-scale for the treatment of crude  
215 digestate. The most established processes commercially available today are: i) *AIRPREX*, Berliner  
216 Wasserbetriebe (Germany, DE), , ii) *ANPHOS*, Colsen (the Netherlands, NL), iii) *CAFR*, NALVA  
217 (DE), iv) *Ceres*, Ceres Milieutechniek (Belgium, BE), v) *NuReSys*, Akwadok (BE)<sup>46</sup>, vi) *Nutritec*,  
218 Sustec (NL), vii) *Pearl*, Ostara (Canada, CA), viii) *Phosnix*, Unitika (Japan, JP), ix) *PHOSPAQ*,  
219 Paques (NL)<sup>47</sup>, and x) *PRISA*, Aachen University (DE)<sup>48</sup>. Moreover, in Gelderland (NL) four  
220 installations are available for the production of K-struvite ( $KMgPO_4 \cdot 6H_2O$ ) from calf manure.<sup>49</sup>

221 These processes have the ability to remove and recover over 80-90 % of the soluble P in the  
222 waste(water) flow, yet only 10-40 % of the NH<sub>4</sub>-N can be captured.<sup>14</sup> Crystal/pellet sizes range  
223 from 0.5 mm to 5 mm and above, depending on the final end-use. The design involves fluidized  
224 bed reactors and continuously stirred tank reactors.

225 At present, struvite recovery can be economical on side streams from wastewater  
226 treatment with a P-load of more than 20 % by weight, as it has the potential to reduce operating  
227 costs by reducing energy and chemical (iron (Fe)/aluminium (Al)) consumption and minimizing  
228 nuisance struvite formation in piping/equipment. Meanwhile, a high-quality, slow-release granular  
229 fertilizer with agricultural reuse perspectives is produced.<sup>50-53</sup> Assuming that a treatment plant can  
230 recover 1 kg of struvite from 100 m<sup>3</sup> of wastewater<sup>54</sup>, Shu et al. (2006)<sup>52</sup> estimated chemical  
231 savings for P recovery from digester supernatants at € 0.19 d<sup>-1</sup> (compared to alum), savings from  
232 reduced sludge handling at € 0.002 d<sup>-1</sup>, from reduced sludge disposal at € 0.023 d<sup>-1</sup>, from reduced  
233 cleaning of struvite deposits at € 3.8-19.2 d<sup>-1</sup> and savings from reduced landfilling at € 0.002 d<sup>-1</sup>. As  
234 such, the cost of producing struvite could amount to € 0.05 d<sup>-1</sup>, while the gain for the treatment  
235 plant can be € 0.52 d<sup>-1</sup>.<sup>52</sup> Hence, assuming a struvite plant would cost € 1.4 million, the payback  
236 period of a plant processing ± 55,000 m<sup>3</sup> d<sup>-1</sup> of waste could be less than five years according to  
237 this study.<sup>52</sup>

238 However, operational costs and payback times are highly dependent on the input  
239 composition (e.g. available P, Mg, and pH) as it determines the chemical (NaOH, Mg) and energy  
240 costs, which can range between 200 and 75,000 € year<sup>-1</sup>.<sup>55</sup> Dockhorn (2009)<sup>56</sup> estimated operating  
241 and maintenance costs for a plant treating 350,000 person equivalents (PE) at € 2,800 ton<sup>-1</sup> of  
242 struvite if the PO<sub>4</sub>-P-concentration is 50 mg L<sup>-1</sup>, and € 520 ton<sup>-1</sup> if the PO<sub>4</sub>-P-concentration is  
243 800 mg L<sup>-1</sup>. Battistoni et al. (2005a, b)<sup>57,58</sup> estimated operating costs at € 0.19-0.28 m<sup>-3</sup> digestate.

244 Based on budget proposals provided by the above-mentioned suppliers in the context of the  
245 present review, capital costs may range from 2,300 to 24,500 € kg<sup>-1</sup> P d<sup>-1</sup>, while revenues from  
246 struvite valorisation in agriculture range from ± € 45 ton<sup>-1</sup> struvite in Belgium (NuReSys, Waregem,  
247 BE, personal communication 2013) to ± € 109-314 ton<sup>-1</sup> in Australia<sup>59</sup>, and ± € 250 ton<sup>-1</sup> in  
248 Japan<sup>60</sup>. Values of € 736 and € 1,393 ton<sup>-1</sup> have also been reported.<sup>56</sup> As such, overall profits of  
249 struvite production may range from € -7,800 year<sup>-1</sup> (loss) to € 89,400 year<sup>-1</sup> (gain).<sup>54</sup>

250 Although world-wide some utilities have installed these systems, the uptake of this  
251 technology has not been widespread due to market, regulatory, and site-specific conditions. Also,  
252 important technical challenges remain in the reduction of chemical requirements, the guarantee of  
253 a pure product, as well as the stable and controlled production of struvite. Energy-efficient methods  
254 without chemical addition, such as electrochemical<sup>61</sup> and bio-electrochemical<sup>62</sup> struvite recovery,  
255 are under development (Supporting Information 3).

256

### 257 **3.2.2 Calcium phosphate recovery**

258 Next to Mg, calcium hydroxide (Ca(OH)<sub>2</sub>) can also be added to the liquid fraction in order to  
259 increase the pH (> 10.0) and temperature (70 °C), thereby inducing P precipitation as  
260 Ca<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>OH (hydroxyapatite) or CaHPO<sub>4</sub>·2H<sub>2</sub>O (brushite). The reaction is fast (5 min), but often  
261 preceding CO<sub>2</sub>-stripping must be applied to avoid unwanted calcium carbonate (CaCO<sub>3</sub>)  
262 precipitation. Examples of commercial calcium phosphate precipitation processes are: i)  
263 *Crystalactor*, DHV Water (NL),<sup>63</sup> ii) *FIX-Phos*, TU DA (DE), iii) *Kurita*, Kurita Water Industries (JP),  
264 iv) *Phostrip*, Tetra Technologies inc. (USA),<sup>64</sup> and v) *P-Roc*, Kit-CMM (DE)<sup>65</sup>. Removal efficiencies  
265 of 80-100 % P have been achieved, but 50-60 % is more typical. Based on a market demand in the  
266 framework of this study to the suppliers mentioned above, capital costs may range between €

267 2,300-2,900 kg<sup>-1</sup> P d<sup>-1</sup>. Operational costs are mainly determined by the amount of Ca(OH)<sub>2</sub>  
268 required, which on its turn depends on the input waste characteristics. Finally, Quan et al. (2010)<sup>66</sup>  
269 reported on the ability to couple CaNH<sub>4</sub>PO<sub>4</sub>·4H<sub>2</sub>O precipitation and ammonia stripping in a water  
270 sparged aerocyclone (WSA) in order to recover both P and N. To date, this path has only been  
271 examined at lab-scale, but further research on this methodology seems interesting.

272

### 273 **3.3 Ammonia stripping and absorption (NRT 2)**

274 Stripping of ammonia (NH<sub>3</sub>) involves the physical transfer of NH<sub>3</sub> from the aqueous phase (waste  
275 stream) to a gas phase, mostly in a packed bed tower. The gas is then transferred to an air  
276 scrubber (Section 3.4), where mass transfer and absorption of the NH<sub>3</sub> from the gas to a liquid  
277 phase, often sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), takes place in order to form and recover a concentrated solution  
278 of ammonium sulfate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>; AmS) as an end-product<sup>34,67</sup>. AmS is an inorganic salt, which  
279 could be reused as a marketable fertilizer rich in direct available macronutrients, N and S, thereby  
280 providing a valuable substitute for chemical fertilizers based on fossil resources.<sup>20-22</sup> Alternatively,  
281 ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>) fertilizer (by addition of nitric acid, HNO<sub>3</sub>)<sup>68</sup>, a concentrated ammonia  
282 solution (by addition of liquid NH<sub>3</sub>), or other ammonia salts can be produced, depending on local  
283 legislation and options for land application. Ammonia stripping is developed at full-scale, and  
284 sometimes implemented for wastewater treatment. The implementation of this technology for the  
285 treatment of N-rich digestate and manure is on the rise.

286 Commercially available stripping technologies for (digested) sludge and manure treatment  
287 are i) *AMFER*, Colsen (NL), ii) *ANAStrip*, GNS (DE), and the (untitled) stripping processes  
288 developed by the manufacturers: iii) Anaergia (Canada, CA), iv) Branch Environmental Corp  
289 (USA), v) Europe Environnement (France, FR), and vi) RVT Process Equipment (DE).

290 Theoretically, these systems may achieve NH<sub>3</sub>-removal efficiencies up to 98 %, but they are  
291 generally operated to reach 80-90 % removal in order to reduce the operating costs. At present,  
292 most stripping units implemented at full-scale focus on the production of AmS-fertilizer. The AmS-  
293 content in the recovered solution ranges from ± 25 % AmS (*ANAStrip*, GNS) and 30 % AmS  
294 (Branch Environmental Corp) to ± 38 % AmS (Anaergia; RVT Process Equipment) and 40 % AmS  
295 (*AMFER*, Colsen; Europe Environnement).

296 Capital costs of stripping are relatively low compared to biological activated sludge (AS)  
297 systems and depend on the method used for pH-increment. This can occur i) chemically by use of  
298 base, mostly sodium hydroxide (NaOH) (Branch Environmental Corp; Europe Environnement; RVT  
299 Process Equipment), or ii) physically by simultaneous stripping of CO<sub>2</sub> (Anaergia; Colsen),  
300 optionally in combination with the addition of low-quality gypsum (CaSO<sub>4</sub>) for parallel recovery of  
301 CaCO<sub>3</sub> (GNS). Operational costs depend a lot on the operational temperature, pH, and liquid flow  
302 rate. For a 90 % NH<sub>3</sub>-recovery efficiency from leachate at a temperature of 70 °C, a pH of 11, and  
303 a flow rate of 70 m<sup>3</sup> h<sup>-1</sup>, overall costs are estimated at ± € 8.1 (± US \$ 10.2) m<sup>-3</sup>, while at a  
304 temperature of 30 °C this would be ± four times less, , i.e. € 2.0 (± US \$ 2.5) m<sup>-3</sup>.<sup>69</sup>

305 A comparison of budget proposals provided by the above suppliers for NH<sub>3</sub>-stripping and  
306 absorption systems treating a digestate flow of 800 m<sup>3</sup> d<sup>-1</sup> at 2,400 mg NH<sub>4</sub>-N L<sup>-1</sup> (90 % recovery)  
307 resulted in a capital cost ranging from € 500,000 to € 1.58 million if the pH-increase is conducted  
308 chemically, and from € 3.5 million to € 11-15 million if the pH-increase is established physically.  
309 Electricity consumption for this case was estimated by the suppliers at 127-400 kWh<sub>el</sub> h<sup>-1</sup>, heat  
310 consumption at 2,115-2,333 kWh<sub>th</sub> h<sup>-1</sup>, and H<sub>2</sub>SO<sub>4</sub>-consumption (concentrated at 95-97 %) at 5.5-  
311 6.8 ton d<sup>-1</sup> or 7.0-10 kg m<sup>-3</sup> digestate. If NaOH is used for pH-increase, its consumption would  
312 amount to 6.0-6.5 kg m<sup>-3</sup>. As such, operational costs range from € 1.4-2.5 million year<sup>-1</sup> depending  
313 on the system, equivalent to € 4.5-8.6 m<sup>-3</sup> of digestate.

314 Currently, advanced processes for biological removal of  $\text{NH}_3$ , such as deammonification  
315 via Anammox,<sup>70</sup> are often still cheaper (depending on the technology provider). However, as  
316 stripping could (partially) replace a nitrification-denitrification step, additionally remove odorous  
317 compounds and dust particles, and produce a marketable end-product, it is expected that this  
318 technology can be competitive, especially in regions where N-demand is high.<sup>53</sup> Where  
319 commercialisation of the AmS-solution is possible, revenues currently range from € 90 to 120 ton<sup>-1</sup>  
320 FW, which under optimal process conditions should largely compensate the operational costs.  
321 Moreover, the higher process stability (e.g. to input variation, freezing conditions, etc.), lower  
322 surface requirements (for the above case:  $\pm 1,500 \text{ m}^2$ ), higher ability for treatment of high N-flows,  
323 immediate start-up, and ease of automation are all drivers for implementation of stripping units  
324 instead of conventional biological systems.

325 The major technical bottlenecks observed to date in  $\text{NH}_3$ -stripping are scaling and fouling  
326 of the packing material, and the consequent high energy and chemical requirements.<sup>13,32,66</sup> To  
327 avoid scaling, one can install a lime softening step before stripping, which removes a large part of  
328 the Ca, Mg, carbonic acids and carbonates, and increases the pH. In case of high buffering  
329 capacity, a preceding  $\text{CO}_2$ -stripper might also be economical. To avoid fouling, it is important that  
330 during preceding solid-liquid separation as many suspended solids as possible are retained in the  
331 solid fraction. Nonetheless, it is unavoidable that the packing material will have to be cleaned  
332 periodically. Because of these constraints, some of the above manufacturers have developed a  
333 stripping process without internal packing (Anaergia; Colsen). As such, the process developed by  
334 Anaergia would be capable of handling waste flows containing up to 8-9 % total suspended solids  
335 (TSS). Note that both technologies also operate without any chemical addition. Hence, although  
336 capital costs are higher (see above), in terms of sustainability and operational costs, these

337 processes may be of interest. Alternative NH<sub>3</sub>-stripping systems aiming to overcome the above  
338 technical bottlenecks, such as a WSA reactor<sup>66</sup> and the use of rotating disks<sup>71</sup>, are also being  
339 developed (Supporting Information 4).

340

### 341 **3.4 Acidic air scrubbing (NRT 3)**

342 Acidic air scrubbing mostly concerns a packed tower in which sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) is sprayed with  
343 nozzles over the packing material and treatment air is blown into the tower in counter-current.<sup>35,67</sup>

344 As is the case for NH<sub>3</sub>-stripping and absorption (Section 3.3), mostly ammonium sulfate  
345 ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>; AmS) is produced and the wash water is recycled until it is saturated and the removal  
346 efficiency of NH<sub>3</sub> cannot be guaranteed anymore. At that point, the AmS-solution should be  
347 removed and fresh H<sub>2</sub>SO<sub>4</sub> added. The technology is used at full-scale at anaerobic digestion and  
348 sludge/manure processing plants. Examples of well-established technology developers are Dorset  
349 Farm Systems (NL/US), Envitech (CA), and Inno+ (NL). Average NH<sub>3</sub>-recovery efficiencies of 91-  
350 99 % are found in literature.<sup>50,67,72</sup> Investment costs (in case of a new installation for air treatment  
351 of one stable) are estimated at € 18 kg<sup>-1</sup> NH<sub>3</sub>-removal, whereas exploitation costs (including  
352 variable and fixed costs) of an acidic air scrubber are estimated at € 6-7 kg<sup>-1</sup> NH<sub>3</sub>-removal.<sup>73,74</sup> As  
353 these costs are expected to reduce with 50 % (investment) and 14-25 % (exploitation) for large-  
354 scale projects<sup>74,75</sup>, the installation is economically viable at many waste-processing plants.

355 The main operational costs can be attributed to the energy (0.057 kWh 1000 m<sup>-3</sup> air) and  
356 acid (1.5 L H<sub>2</sub>SO<sub>4</sub> at 98 % kg<sup>-1</sup> NH<sub>3</sub> -recovery) requirements.<sup>74</sup> However, power inputs depend a lot  
357 on the reactor type, ranging from 3.8 atm cm<sup>3</sup> air s<sup>-1</sup> for spray-chambers to 260 atm cm<sup>3</sup> air s<sup>-1</sup> for  
358 venturi scrubbers.<sup>76</sup> Interesting advantages of an acidic air scrubber is that odors, dust particles,  
359 and water vapour can also be removed. Technical bottlenecks are mainly related to corrosion



360 problems. The reject AmS-solution is recognised in Flanders and the Netherlands as a mineral  
361 fertilizer in environmental legislation. However, marketing is still hindered due to its variable N and  
362 S content (30-100 kg N ton<sup>-1</sup>; 61-100 kg S ton<sup>-1</sup>), acidic and corrosive features (pH 2.5-7, high salt  
363 content: 100-150 mS cm<sup>-1</sup>) as well as social perception and farmers' distrust in its fertilizer  
364 properties.<sup>21-23</sup> It should be noted that the product properties are highly dependent on the  
365 technology provider, not only in terms of AmS content (see above), but also in terms of pH.  
366 Manufacturers delivering a fertilizer product at relatively high pH, suitable for direct application in  
367 agriculture, are Anaergia (pH 5.5) and RVT Process Equipment (pH 6-7).

368

### 369 **3.5 Membrane filtration (NRT 4)**

370 Pressure-driven membrane filtration, involving microfiltration (MF), ultrafiltration (UF), nanofiltration  
371 (NF), and/or reversed osmosis (RO) is an established technology in wastewater treatment. It has,  
372 however, not yet proven to be a valuable option for the treatment of digestate, manure, and sludge.  
373 Only a few commercial pilots have been installed at full-scale manure and digestate processing  
374 facilities, mostly on a short-term basis because of excessive operational costs. Nevertheless, the  
375 produced membrane filtration concentrates are an interesting nutrient source, which could  
376 potentially be reused as chemical fertilizer substitutes rich in N and K.<sup>19,37,76,77</sup> Examples of  
377 manufacturers for slurry filtration systems are: i) A3 Watersolutions (DE), ii) AquaPurga  
378 International (NL), iii) New Logic (USA), iv) VP Systems (NL), and v) Wehrle Umwelt GmbH (DE).  
379 Operating temperatures range from 10-40 °C, while the pH is usually between 6 and 8. RO has  
380 also been applied at full-scale in combination with NH<sub>3</sub>-stripping of liquid digestate (*Biorek Process*,  
381 BIOSCAN (Denmark, DK)).<sup>79</sup>

382 In reality, the cost of an RO-filtration system for manure and digestate treatment is difficult  
383 to determine, because it depends on the frequency of membrane cleaning and replacement, as  
384 well as the permeate and concentrate end-use, which on its turn are site and region specific.<sup>16</sup>  
385 Gerard (2002)<sup>80</sup> estimated the cost of manure treatment using two RO-cycles at € 12 m<sup>-3</sup> for a 2 m<sup>3</sup>  
386 d<sup>-1</sup> flow at a pilot-scale installation in France. At a pig farm in Canada, the company Purin Pur  
387 estimated the costs of an UF-RO treatment train at € 4.22 (5.97 CAD) m<sup>-3</sup> in 2000.<sup>81</sup> In 2009-2010,  
388 a large pilot project was established in the Netherlands, in which, with authorization of the  
389 European Commission, the RO-concentrate of eight different manure/digestate processing facilities  
390 was applied to agricultural fields. The costs of the installations plus the costs of transporting the  
391 final products amounted to € 9-13 ton<sup>-1</sup> manure/digestate, which was economically feasible for  
392 seven of the eight installations as the price received for treating the manure at that time amounted  
393 to € 11-13 ton<sup>-1</sup> waste.<sup>77,78</sup> The economic value of the RO-concentrates is estimated at 6.1±1.1 €  
394 ton<sup>-1</sup> FW<sup>19</sup>, while the average price paid by farmers during the pilot project was € 1.25 ton<sup>-1</sup> in 2009  
395 and € 1.19 ton<sup>-1</sup> in 2010, yet the standard deviation was high.<sup>78</sup> No other prices for membrane  
396 concentrate marketing have been reported to date.

397 The biggest technical problem stated in membrane filtration is clogging and fouling of the  
398 membrane, resulting in significant chemical and energy requirements.<sup>36,38</sup> The equivalent energy  
399 requirement for an RO-system is estimated at 4-6 kWh m<sup>-3</sup>.<sup>82,83</sup> To reduce cleaning requirements,  
400 vibrating (60-90 Hz) shear enhanced processing (VSEP) has been used for manure and digestate  
401 purification at pilot-scale.<sup>19,84,85</sup> However, little data are available on the energy consumption and  
402 treatment costs of this technology. The energy consumption per vibration is estimated at 8.83 kW  
403 (G. Johnson, New Logic Inc., Ontario, CA, personal communication 2011), while the energy  
404 consumed by the recirculation pump is estimated at 9.4 kWh m<sup>-3</sup> of permeate in a 154 m<sup>2</sup>

405 membrane area unit (VSEP series i-10).<sup>86</sup> This could be reduced to 6 kWh m<sup>-3</sup> if plane ceramic  
406 membranes are used. Energetic calculations based on these data indicate that large VSEP-units  
407 will consume significantly less energy per m<sup>3</sup> of permeate than traditional cross-flow filtration.  
408 Nevertheless, energy consumption and economic performance remain critical points of attention in  
409 the evaluation of membrane technologies for nutrient recovery.

410 Different alternative technologies to improve the performance of membrane filtration in  
411 terms of chemical and energy requirements, as well as operational costs are currently under  
412 development. The most studied examples are forward osmosis<sup>87-90</sup>, electro dialysis<sup>91-94</sup>, and  
413 transmembrane chemisorption<sup>95-97</sup> (Supporting Information 5).

414

### 415 **3.6 Ammonia and P sorption (NRT 5)**

416 A number of materials may be used to selectively adsorb ammonium (NH<sub>4</sub><sup>+</sup>) and P from waste  
417 flows. These materials include zeolites, clays, and resins, whether or not chemically or thermally  
418 modified. Adsorption is carried out in a packed column. Once the adsorption media is saturated,  
419 the column is taken offline and is regenerated to recover the NH<sub>4</sub><sup>+</sup> and/or P. Regeneration can be  
420 achieved by a number of techniques, including nitric acid (HNO<sub>3</sub>) washing, sodium chloride (NaCl)  
421 washing, or biologically. The technique to be used depends on the adsorption material and the  
422 desired end-product. Adsorption can therefore either be operated as a batch process using a single  
423 column, or a series of multiple columns can be sequenced to provide continuous operation.<sup>40,98-99</sup>

424 In the context of wastewater treatment, especially natural zeolites have been successfully  
425 used as adsorption agent for final NH<sub>4</sub>-N-removal.<sup>30,100-102,103-107</sup> Mainly the use of clinoptilolite,  
426 (Na,K,Ca)<sub>2-3</sub>Al<sub>3</sub>(Al,Si)<sub>2</sub>Si<sub>13</sub>O<sub>36</sub>:12(H<sub>2</sub>O), has been studied because of its low-cost availability.<sup>40</sup>  
427 However, full-scale wastewater treatment plants that employ the NH<sub>4</sub><sup>+</sup>-ion exchange technique are

428 scarce, and few applications have been developed to recover  $\text{NH}_4\text{-N}$ , for example, for agricultural  
429 purposes.<sup>40,108</sup> Nevertheless, because the zeolite is porous, the ammonium can leak out of the  
430 zeolite at a much slower rate than it was adsorbed. This makes the ammonium-filled clinoptilolite  
431 itself a potential slow-release fertilizer.<sup>98</sup> An important remark is that the initial N-concentration in  
432 the above-mentioned applications was only a few  $10 \text{ mg L}^{-1}$ . Over the past decade, there has been  
433 increasing interest of using natural zeolite for  $\text{NH}_4\text{-N}$ -removal from waste streams with relatively  
434 high N-concentration or high ionic strength.<sup>109-112</sup> However, its applicability in practice for the  
435 treatment of the liquid fraction of digestate (containing both high N- and ionic concentrations) still  
436 remains to be demonstrated, as does the use of the nutrient-enriched clinoptilolite or other  
437 regenerated N-solutions as a fertilizer.<sup>13,39,40</sup>

438         To date, removal efficiencies of 18 % P (probably due to adsorption) and 15-60 % N (due  
439 to ion exchange) have been reported for the treatment of human urine using clinoptilolite at lab-  
440 scale.<sup>98</sup> This means that the technology would currently not be feasible as stand-alone NRT for  
441 digestate processing, though it may be used as an intermediate step in the digestate treatment  
442 train. As such, some research has been performed towards the combined use of zeolite and  
443 struvite precipitation to obtain a slow-release fertilizer with both high N- and P-concentrations.  
444 Overall recovery efficiencies of 100 % P and 83 % N have been obtained at lab-scale, and the  
445 fertilizer potential of the resulting product has been demonstrated at greenhouse scale.<sup>98</sup> Moreover,  
446 Liberti et al. (1982)<sup>113</sup> investigated a system where  $\text{NH}_4^+$ -ions were concentrated by the  $\text{NH}_4^+$ -ion  
447 exchange technique, followed by air stripping of  $\text{NH}_3$ -gas, which was subsequently absorbed in  
448 sulfuric acid. Zeolites may also be used to further treat the effluent produced by membrane filtration  
449 of the liquid fraction of digestate.<sup>39</sup> Hence, although there is potential to use zeolites as a  
450 technology for nutrient recovery, to date the use of this process for digestate treatment can rather  
451 be considered as an intermediate or final concentration step in the three-step framework (Fig. 1).

452 An important challenge for implementation of zeolites for digestate treatment is fouling of  
453 the adsorbent bed, as well as maintaining the bed capacity after multiple recovery/regeneration  
454 cycles.<sup>41</sup> Moreover, to date, experiments on the operational performance, process optimization,  
455 and recovery/regeneration methodologies are mostly carried out at lab-scale. Hence, further  
456 investigation into the process efficiency at pilot-scale is required.<sup>108</sup>

457 Furthermore, for concentrated waste streams (2,000 mg L<sup>-1</sup>), typically, red mud, metal  
458 oxide/hydroxide and zirconium sorbents are used for P recovery. Phosphorus may be removed  
459 from solution via selective sorption to a solid phase and direct use of the material as a  
460 fertilizer or soil conditioner, or the sorbed P may subsequently be stripped from the solid  
461 sorbent and chemically precipitated as a high-purity fertilizer. However, as is the case for  
462 natural zeolites (see above), further research is required on the use of these sorbents for digestate  
463 treatment.<sup>99</sup>

464 Finally, costs of this technology are expected to be low, but will depend on the nearby  
465 availability of the sorption material used, the required pre-treatment of the packing column to obtain  
466 NH<sub>4</sub><sup>+</sup>- and/or P-selectivity, the recovery/regeneration method (if applicable), and the required  
467 frequency of regeneration. No cost-benefit analyses for nutrient recovery from digestate using  
468 zeolites or other sorbents have been reported on in literature to date.

469

### 470 **3.7 Biomass production and harvest (NRT 6)**

471 Both macrophytes (mostly duckweeds and water hyacinths) and microalgae have been examined  
472 for biological nutrient recovery.<sup>15,114,115</sup> Duckweed (*L. minor*, *L. Punctate*, *S. polyrrhiza*, *S.*  
473 *Oligorrhiza*) ponds have been successfully used as an efficient and potentially low-cost option in  
474 (anaerobically digested) agricultural waste polishing, generating a biomass with high protein

475 content. A detailed overview of the nutrient and heavy metal content of duckweed in function of  
476 water quality has been reported.<sup>116,117</sup> Based on its mineral composition, the plant appears to have  
477 the ability to recover 600, 56-140, 400, 100, 60, 32, and 24 kg ha<sup>-1</sup> year<sup>-1</sup> of N, P, K, Ca, Mg, Na,  
478 and Fe, respectively at a production of 10 tons dry weight (DW) ha<sup>-1</sup>.<sup>117</sup> Xu and Shen (2011)<sup>34</sup>  
479 found removal efficiencies of 83.7 and 89.4 % for total N and P from pig wastewater, respectively,  
480 using *S. oligorrhiza* in eight weeks at a harvest frequency of two times a week. Mohedano et al.  
481 (2012)<sup>118</sup> found an average of 98.0 % total N and 98.8 % total P recovery at full-scale, resulting in  
482 an average biomass (*L. punctate*) protein content of 28-35 %. However, above 60 mg N L<sup>-1</sup> a toxic  
483 effect was noticed perhaps due to high levels of free ammonia in the water, hence levels below this  
484 value should be maintained in order to obtain a consistently high protein content (15-45 % by DW,  
485 depending on the N-supply<sup>114</sup>).

486 Skillicorn et al. (1993)<sup>119</sup> estimated the capital costs for a 0.5 ha large duckweed system at  
487 ± € 2,600 (US \$ 3,300). However, capital costs are significantly influenced by land area  
488 requirements, next to the costs associated with pond inoculation, harvesting, and disposal of  
489 biomass. As such, Mburu et al. (2013)<sup>120</sup> evaluated the capital expenditures (CAPEX) for a full-  
490 scale waste stabilization pond at € ± 705,000 (based on 2,700 person equivalents (PE) at 8.3 m<sup>2</sup>  
491 PE<sup>-1</sup>) and at € ± 276,000 (for 2,700 PE at 3.4 m<sup>2</sup> PE<sup>-1</sup>) for a pilot-scale horizontal subsurface flow  
492 constructed wetland. Maintenance costs for the first design are, however, significantly lower: € 283  
493 compared to € 23,300 for 2,700 PE. As such, the total cost of these “green” nutrient recovery  
494 systems can be evaluated at € 12-33 PE<sup>-1</sup> year<sup>-1</sup>, with an average of € 14.4 PE<sup>-1</sup> year<sup>-1</sup>, whereas  
495 the cost for a traditional activated sludge system is at least three times higher. However, without  
496 water reuse, associated gray-water sales, and duckweed valorization, currently the economic  
497 viability of duckweed systems remains questionable.<sup>114</sup>

498           Furthermore, the ability of microalgae to assimilate excess nutrients from the environment  
499 has been thoroughly studied.<sup>121</sup> However, to date, tests of growing algae in waste(water) are  
500 mostly at laboratory scale. Pilot-scale algae cultivation continues to face many problematic issues,  
501 including contamination, inconsistent waste(water) components, and unstable biomass production.  
502 The major challenge associated with culturing algae in nutrient-rich natural water and slurry comes  
503 from the design of the cultivation system. The addition of polymer that precipitates suspended  
504 solids, thereby allowing light penetration, would improve the technical feasibility of growing algae  
505 on the liquid fraction of (digested) slurry. Nevertheless, Muylaert and Sanders (2010)<sup>122</sup> predict that  
506 breakthrough of algae in the bio-based economy will last another 5-15 years, as currently costs of  
507 algae production are too high as compared to other types of biomass. Estimates of the algal  
508 production cost range from € 3.2-240 (US \$ 4,300) kg<sup>-1</sup> dry biomass.<sup>123-125</sup> Prior economic-  
509 engineering feasibility analyses have concluded that even the simplest open pond systems,  
510 including harvesting and algal biomass processing equipment, would cost at least € 78,000 (US \$  
511 100,000) ha<sup>-1</sup>, and possibly significantly more.<sup>91</sup> To this, the operating costs will need to be added.

512           The harvested algae/macrophytes can serve as a feedstock for the chemical and biofuel  
513 industry, can be used as animal feed (provided that the necessary amendments in legislation are  
514 made), or spread out as a fertilizer.<sup>126,127</sup> As such, El-Shafai et al. (2007)<sup>128</sup> estimated a biomass  
515 value of € 5,300 (US \$ 6,600) year<sup>-1</sup> (by comparison with other feed sources) for a protein yield of  
516 approximately 11 tons ha<sup>-1</sup> year<sup>-1</sup> (*L. gibba*). Next to the large footprint required, biomass  
517 harvesting remains an important technical bottleneck, which requires further research.

518

### 519 **3.8 Phosphorus extraction from ashes/biochar (NRT 7)**

520 The remaining ashes after combustion of biodegradable waste (manure, sludge, digestate) contain  
521 P-, K-, Al-, and silicium (Si) compounds and possibly heavy metals such as Cu, Zn, and cadmium  
522 (Cd). Several companies have designed different processes to extract P from such combustion  
523 ashes.<sup>45</sup> These processes can be subdivided into thermochemical and wet-chemical technologies.  
524 The Finnish company Outotec, for example, adds magnesium chloride (MgCl<sub>2</sub>) and heats the  
525 ashes to 1,000 °C in order to gasify the heavy metals. Phosphorus is bound as CaHPO<sub>4</sub> and sold  
526 as chemical mineral fertilizer substitute. The Belgian company EcoPhos developed and tested, at  
527 lab-scale, a chemical P-extraction process by addition of hydrogen chloride (HCl) to combustion  
528 ashes.<sup>128</sup> Also the Swedish company EasyMining developed a process (*Cleanmap Technology*)  
529 that involves the use of HCl, which is suitable for ashes from incinerated manure. Other P recovery  
530 processes from ash are: *ICI Amfert* (NL), *Mephrec* (DE), *PAsH* (DE), *RecoPhos* project (Austria  
531 (AT), BE, DE, FR, Switzerland (CH)), *sephos* (DE), and *susAN/AsH DEC* (AT, DE, Finland (FI),  
532 NL). P recoveries up to 78 % are found in literature.<sup>45,130</sup> Operational costs for wet extraction itself  
533 can be lower than € 1 m<sup>-3</sup> fresh slurry, whereas net variable costs for combustion (including  
534 revenues from energy and P recovery) range from € 0-10 ton<sup>-1</sup> FW slurry, depending on the water  
535 content. However, a thorough flue gas cleaning system is indispensable and post-processing to  
536 remove heavy metals is often required, hence few full-scale installations currently exist.

537 Experiments with pyrolysis of manure cakes have also been conducted. The fraction of  
538 nutrients recovered in the resulting biochar is larger than in incineration ashes and the plant-  
539 availability of the nutrients tends to be higher, especially for P. It was estimated that the value of P  
540 in bio-char is about five times higher than the value of P in ash: < € 1 m<sup>-3</sup> vs. € 4.25 m<sup>-3</sup>.<sup>45</sup>

541 Nevertheless, as digestate is classified as a waste stream that is eligible for recycling as  
542 soil conditioner, it is in a lot of countries not authorised to convert the product into energy by



543 combustion/pyrolysis according to environmental legislations. Alternatively, P could be extracted  
544 from dried or dewatered digestate, but to date such tests are absent in literature. Some processes  
545 attempting to recover P from dried or dewatered sludge are: *IEACHPOs* (CH), *Mephrec* (DE),  
546 *PHOXNAN/10PROX* (DE), and *seaborne* (DE).

547

### 548 **3.9 Critical comparison**

549 Based on the above compiled information, a critical comparative technology overview is given in  
550 Table 1.

551

552 **Table 1:** Technology overview: technology, mechanism, feed, % recovery, main technical  
553 bottlenecks, economic and ecological evaluation, stage of development, and references.

554

555 At present, only struvite precipitation/crystallization, NH<sub>3</sub>-stripping and absorption in a tower (with  
556 or without packing), acidic air scrubbing, and pressure-driven membrane filtration have been  
557 applied at full-scale for nutrient recovery from digestate. Of these technologies, only the first three  
558 have shown potential to be economically viable for implementation at digestate processing  
559 facilities.

560 Traditional membrane filtration systems often suffer technical problems in wastewater  
561 treatment, making them economically not yet viable for digestate treatment. An interesting solution  
562 may exist in vibrating membrane filtration (VSEP) using RO-membranes. However, further  
563 research is required in order to evaluate the technical and economic performance of this process  
564 (see Vaneeckhaute et al., 2012<sup>19</sup>). Nevertheless, it should be pointed out that membrane filtration  
565 is the most established technology to date for the simultaneous **recovery of both N and K**.

566 Further, Table 1 shows that the NRT that currently achieves the highest simultaneous  
567 nutrient **recovery efficiency of both N and P** would be biomass production and harvest. However,  
568 the overall cost of this treatment is still high and large surface areas are required, making its  
569 potential implementation very region-specific. Further research to improve the economic and  
570 technical feasibility of this technology is recommended.

571 In terms of **costs**, NH<sub>3</sub>-sorption and recovery of N-enriched (natural) zeolites is probably  
572 the lowest-cost option available to date (depending on the nearby availability of the adsorbent).  
573 However, recovery efficiencies are relatively low and further testing is required on the  
574 implementation of this technology for digestate treatment. Moreover, the marketing value of the  
575 recovered N-zeolites remains to be explored. The use of this technology in combination with  
576 struvite or stripping (depending on the purpose) may be interesting, in order to maximize the  
577 overall nutrient recovery potential at minimal costs. Of the technologies applied to date at full-scale  
578 (see above), acidic air scrubbing is likely the most feasible technology, since air treatment is often  
579 required anyway for odor removal at processing facilities. While the investment costs of N stripping  
580 and absorption are relatively low, its operational expenditures (OPEX) are usually much higher  
581 than for P recovery through struvite precipitation. The main operational costs for struvite recovery  
582 are related to chemical dosing, while the main costs for stripping and air scrubbing are related to  
583 the air requirements. Optimization of these parameters is therefore of high interest. Technologies  
584 that are expected to become economically viable in the longer term (after sufficient research and  
585 optimization) are forward osmosis, TMCS, and biomass production.

586 Further, from an **ecological point of view** (chemical and energy use), transmembrane  
587 chemisorption, forward osmoses, (bio)electrochemical struvite recovery, and NH<sub>3</sub>-sorption appear  
588 as the most interesting NRT's. However, none of these technologies is currently applied at full-  
589 scale for the treatment of digestate. Yet, after sufficient testing and optimization, these systems

590 have the potential to become part of commonly used digestate processing technologies. The  
591 extraction of P from ashes or biochars seems the least promising technology, because it can be  
592 questioned whether combustion/pyrolysis of digestate is a sustainable treatment option and if this  
593 should be encouraged.

594 Finally, from a **technical perspective**, it can be stated that further fine-tuning is still  
595 required for all technologies in order to minimize operational costs, especially in terms of energy  
596 and chemical consumption, produce high-quality, pure fertilizers, and economically valorize the  
597 recovered nutrients. The best available and most established technologies for nutrient recovery  
598 from digestate in terms of technical performance and fertilizer marketing potential are struvite  
599 precipitation, ammonia stripping and absorption using a stripping column with or without packing,  
600 and acidic air scrubbing. It is not surprising that these are the only technologies to date that have  
601 been successfully implemented at full-scale digestate processing facilities.

602

## 603 **4. Product quality and markets**

### 604 **4.1 Classification of recovered products**

605 A classification of products that can be recovered from digestate is provided in Table 2.

606 Comparative information on their fertilizer characteristics and marketing value is also presented.

607

608 **Table 2:** Classification recovered end-products: class, technology, feedstock, product,  
609 composition/quality, current marketing value, and reference.

610

611 Based on their fertilizer composition, the current available recovered products can be classified as

612 N/P-, K/P- or P-precipitates, P-extracts, N/S-solutions, N/K-concentrates, N-zeolites, and biomass.

613 The two recovered bio-based products that are currently supplied in the largest quantities and offer  
614 the highest potential for agricultural valorization are struvite from chemical precipitation and AmS  
615 from stripping and acidic air scrubbing. These products can be classified as N/P-precipitate and  
616 N/S-solution, respectively. N/K-concentrates could become an important recovered fertilizer in the  
617 future, if a technical and economic membrane filtration option would become available, for  
618 example, the VSEP-technology.<sup>19</sup> An important aspect for commercialisation of bio-based products  
619 is the guarantee of the product's purity. Pollution with organics, metals, or other contaminants may  
620 occur. In this sense, N recovery through stripping and/or acidic air scrubbing is of high interest  
621 since the purity of the recovered N/S-solutions only depends on the quality of the sulfuric acid  
622 used. Moreover, the development of a model library for resource recovery that allows to predict  
623 and optimize fertilizer quantity and quality under variable conditions (operation, input  
624 characteristics, etc.) is of high value.<sup>12</sup>

625 In-depth product characterisation in time and long-term field trials aiming at the evaluation  
626 of the environmental impact of bio-based products are rare in literature, but highly important in the  
627 development of a market for recovered nutrients. Several researchers have investigated the  
628 fertilizer properties of struvite and the product has been evaluated as an eco-friendly fertilizer for  
629 agricultural production.<sup>131,132</sup> However, the findings reported are mainly based on greenhouse  
630 studies, whereas long-term field trials using recovered struvite from digestate are limited.<sup>132</sup>  
631 Moreover, to reduce costs, these field trials are focussed on plant yield and P-uptake in particular,  
632 but do not investigate the mobility of (other) nutrients and heavy metals.

633 Next, the only reference found in literature on the beneficial value of recovered AmS  
634 fertilizer is the three-year field trial conducted by Ghent University in Belgium.<sup>21-23</sup> For membrane  
635 filtration concentrates, the only study that has been reported on to date is the pilot plant project in

636 the Netherlands, in which the product has been applied during a two-year field experiment.<sup>78,79</sup> In  
637 order to establish the use of bio-based products in the agricultural community and to hasten the  
638 integration of these products in environmental and fertilizer legislations, more in-depth field trials  
639 focusing on the environmental impact of these products, next to their agronomic potential, should  
640 be conducted. Best management practices should also be established, see, e.g., Vaneeckhaute et  
641 al. (2014).<sup>22</sup> All of this may also help to better estimate the economic value of these amendments  
642 compared to the conventional used chemical fertilizers. Indeed, to be economically profitable, the  
643 price allocated to the recovered nutrients should be in accordance to the market price of N, P, K,  
644 and S in mineral fertilizers.

645

## 646 **4.2 Fertilizer market trends and outlook**

647 Of the total world-wide amount of N-fertilizers, only 4 % is AmS<sup>2</sup>, mainly due to its relatively low N  
648 content as compared to that of for instance urea (21 % and 45 % respectively). Recently, however,  
649 the world-wide supply of AmS has increased, in part due to the production of AmS by direct  
650 reaction crystallization from (spent) sulfuric acid and NH<sub>3</sub>. This additional AmS-supply has been  
651 absorbed quickly in the marketplace, because of a general increase in fertilizer demand and an  
652 increased need for S-nutrition in particular. Deficiency of S became a problem for more than 75  
653 countries according to United Nations statistics<sup>134</sup> and supply of this nutrient could be efficient by  
654 using new (recovered) fertilizers containing sulfate.<sup>135</sup> The current additional production capacity of  
655 AmS from waste streams has not even been sufficient to fulfill the market requirements, however,  
656 and naturally, this gap in the supply-demand relationship has led to a rise in AmS-prices. As one  
657 might expect, the price of AmS varies with the various types of product quality available. The  
658 largest disparity is related to particle size, where prices of granular (2-3 mm) crystals are up to

659 three times higher than these of < 1 mm crystals. This price differential can be a strong incentive to  
660 produce large crystals. Hence, the trend of the market is toward the production of the so-called  
661 'granular' AmS-quality, with a coarse fraction of 80 % > 1.8 mm, which has a higher sales return  
662 compared to standard quality, but requires an improvement of the production process.<sup>136</sup>

663         Next, the demand for controlled- and slow-release (CSR) fertilizers, such as struvite, will  
664 continue to grow as they prove to be an efficient alternative to conventional fertilizers because of  
665 their environmentally friendly, resource-saving, and labor-saving (mainly due to the decreased  
666 application frequency) characteristics.<sup>137-138</sup> However, because of the high price of these products  
667 relative to conventional fertilizers, their use is still limited primarily to ornamental, horticultural, and  
668 turf applications. As larger production scales for these materials are achieved, costs will continue to  
669 decline, making them more attractive for commodity/open-field/broad-acre crops such as corn,  
670 wheat, and potatoes. Coated fertilizers, particularly polymer-coated products, have been the  
671 fastest-growing segment of the CSR-fertilizer market, and will continue to grow at a faster rate than  
672 other CSR-fertilizer types. Overall, global demand for these products will continue to increase at  
673 about 2 % annually during 2012–2017 for horticultural and turf applications, including agricultural  
674 crop applications.<sup>137</sup>

675         Considering the full nutrient chain, on average over 80 % of N and 25-75 % of P consumed  
676 end up lost in the environment, wasting the energy used to prepare them and causing emissions of  
677 greenhouse gases and nutrient compounds to water.<sup>2</sup> Hence, if the production price of recovered  
678 AmS and struvite from organic wastes would be competitive with that of chemical fertilizers and if  
679 their application proves not harmful for crop production and soil quality, these products may and  
680 should be used to fulfill future fertilizer market demands, thereby meeting the challenge to produce  
681 more food and energy with less pollution.

682

## 683 **5. Conclusions**

684 Struvite precipitation/crystallization, NH<sub>3</sub>-stripping and absorption, and acidic air scrubbing can be  
685 selected as best available technologies for nutrient recovery from digestate. These technologies  
686 have already been implemented at full-scale and have the ability to produce marketable end-  
687 products. Vibrating membrane filtration (VSEP) also shows potential to become part of the  
688 commonly used nutrient recovery technologies, but its technical and economic performance at full-  
689 scale remains to be demonstrated. All technologies require further technical fine-tuning in order to  
690 minimize operational costs, especially towards energy and chemical use, and to improve the  
691 quality and predictability of the produced fertilizers. To date, recovered bio-based fertilizers can be  
692 classified as renewable N/P-, K/P-, or P-precipitates, P-extracts, N/S-solutions, N/K-concentrates,  
693 N-zeolites, and biomass. Future research should further explore, verify, and improve the fertilizer  
694 characteristics and marketing value of these products towards industrial and agricultural end-users.

695

## 696 **Acknowledgments**

697 This work has been funded by the European Commission under the Interreg IVb Project *Arbor* and  
698 by the Environmental & Energy Technology Innovation Platform (MIP) under the project *Nutricycle*.  
699 The first author is also funded by the Natural Science and Engineering Research Council of  
700 Canada (NSERC), the Fonds de Recherche sur la Nature et les Technologies (FRQNT) and  
701 Primodal Inc. through a BMP Industrial Innovation Scholarship (BMP doctorat 178263). Peter  
702 Vanrolleghem holds the Canada Research Chair in Water Quality Modelling.

703

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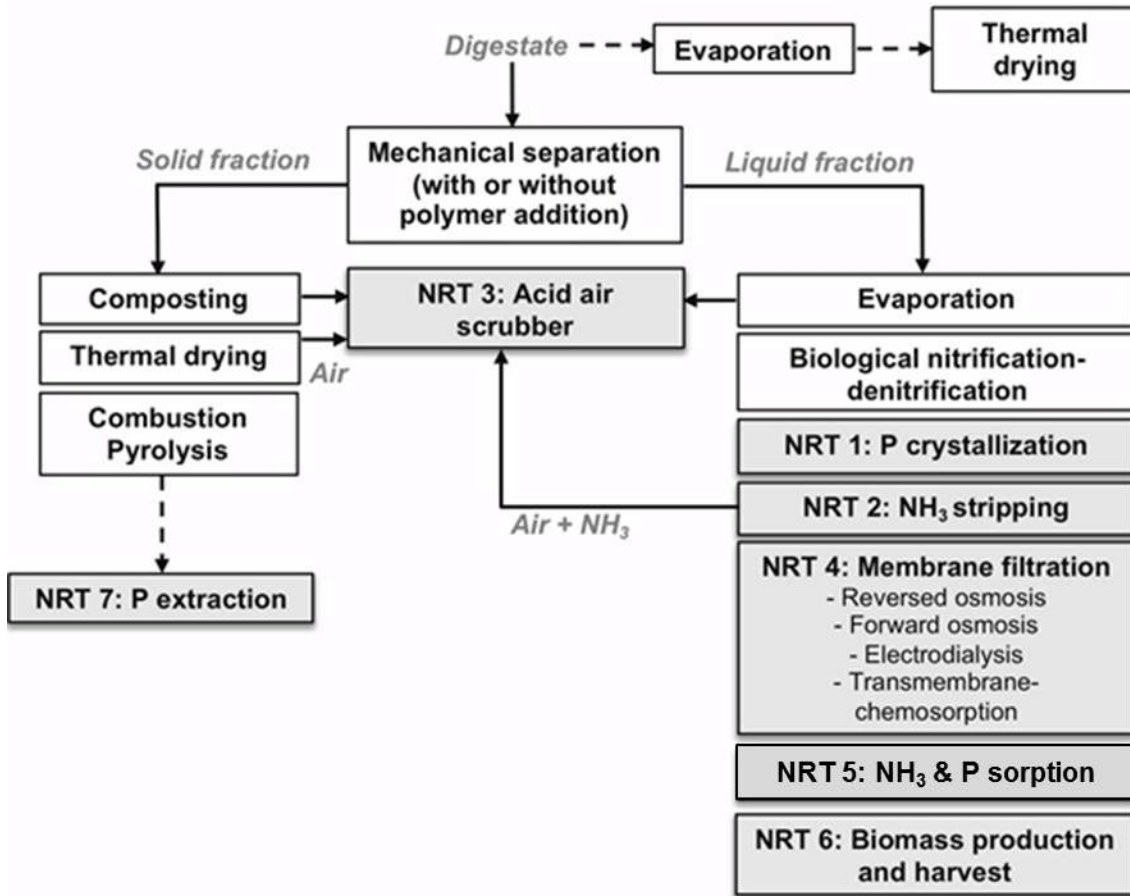
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1090 **Figure 1:** Schematic overview of digestate processing technologies. Shaded boxes: nutrient  
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**Table 1:** Technology overview: technology, mechanism, feed, % recovery, main technical bottlenecks, economic and ecological evaluation, stage of development, and references.<sup>a</sup>

Technology	Mechanism	Feed	% recovery	Main technical bottlenecks	Economic evaluation	Ecological Evaluation	Stage of development	References
Struvite precipitation	Physicochemical	Liquid	80-90 % P 10-40 % N	Precipitation in piping/equipment; Pollution with organic compounds; Stable and controlled production	Can be profitable; CAPEX: € 2,300-24,500 kg <sup>-1</sup> P d <sup>-1</sup> ; OPEX: € -520-2,800 ton <sup>-1</sup> struvite or € -200-75,000 y <sup>-1</sup> Overall: € -7,800-89,400 y <sup>1</sup>	Chemical use (NaOH, Mg) ↑; Fe/Al use ↓; Landfill ↓; Sludge handling and disposal ↓; Cleaning of struvite deposits ↓	Full-scale	14, 52, 55 Technology providers
	Electrochemical	Liquid	> 99 % P	R&D Needed	R&D needed	Chemical use ↓; Energy use ↓: < 70 Wh m <sup>-3</sup>	Lab Pilot: test phase	61
	Bio-electrochemical	Liquid	20-40 % P	R&D Needed	R&D needed	Energy efficiency ↑: 73±4 %; H <sub>2</sub> production	Lab	62
Calcium phosphate precipitation	Physicochemical	Liquid	50-100 % P	Co-precipitation of CaCO <sub>3</sub> ; Preceding CO <sub>2</sub> stripping often required	Can be profitable; CAPEX: € 2,300-2,900 kg <sup>-1</sup> P d <sup>-1</sup> ; OPEX: depends on Ca(OH) <sub>2</sub> use	Chemical use (Ca(OH) <sub>2</sub> ) ↑; Fe/Al use ↓	Full-scale: manure & wastewater; Lab: digestate	63, 65 Technology providers
NH <sub>3</sub> stripping & absorption	Physicochemical: tower (packed bed or no packing)	Liquid	Up to 98 % N; Typical: 80-90 % N	Fouling and corrosion of packing material	CAPEX: € 0.5-15 million, OPEX: € 4.5-8.6 m <sup>-3</sup> , both for 800 m <sup>3</sup> d <sup>-1</sup> at 2.4 g N m <sup>-3</sup> (90 % recovery); Overall: € 2.0-8.1 m <sup>-3</sup> for 70 m <sup>3</sup> h <sup>-1</sup> ; Depends on pH and temperature; Can (partially) replace activated sludge system; Interest in S ↑	Odors ↓; Energy use (air/heat) ↑: 1.54-12 kWh <sub>air</sub> m <sup>-3</sup> and 62-69 kWh <sub>th</sub> m <sup>-3</sup> ; Acid use ↑: 7-10 kg H <sub>2</sub> SO <sub>4</sub> m <sup>-3</sup> ; Base use (↑): 0-6.5 kg NaOH m <sup>-3</sup> , all for 800 m <sup>3</sup> d <sup>-1</sup> at 2.4 g N m <sup>-3</sup> (90 % recovery); Chemical use for cleaning ↑	Full-scale	13, 32, 69 Technology providers
	Physicochemical: water-sparged aerocyclone	Liquid	> 97 % NH <sub>3</sub>	R&D needed; Scale-up?!	Interest in S ↑; Potential for simultaneous P recovery	Self-cleaning; Energy use ↓; Acid use ↑; Chemical use ↓	Lab	66
	Physicochemical: rotating disks	Liquid	± 80 % NH <sub>3</sub>	R&D needed; Scale-up?!	Interest in S ↑; No air scrubber required	Energy use ↓: < 1 kW; Acid use ↑; Chemical use ↓	Pilot	71
Acidic air scrubbing	Physicochemical	Gas	91-99 % N	Fouling/corrosion of packing material; Performance under freezing conditions?!	Can be profitable; CAPEX: € 13 kg <sup>-1</sup> NH <sub>3</sub> removal; OPEX: € 6-7 kg <sup>-1</sup> NH <sub>3</sub> removal; Interest in S ↑	Odor ↓; Energy use (air) ↑: min. 0.057 kWh kg <sup>-1</sup> NH <sub>3</sub> ; Acid use ↑: min. 1.5 L H <sub>2</sub> SO <sub>4</sub> kg <sup>-1</sup> NH <sub>3</sub>	Full-scale	35, 67, 72, 74
Membrane filtration	Physical: pressure-driven membrane filtration	Liquid	N and K; % depends on pre-treatment	Membrane blocking & scaling; High maintenance and power requirements	High CAPEX & OPEX: € 4-13 m <sup>-3</sup>	Energy use ↑: 4-6 kWh m <sup>-3</sup> (RO); Chemical use (cleaning) ↑	Full-scale	36, 38, 77, 78, 80, 81, 82, 83

Technology (Continuation)	Mechanism	Feed	% recovery	Main technical bottlenecks	Economic evaluation	Ecological Evaluation	Stage of development	References
	Physical: forward osmosis	Liquid	R&D needed	Reverse solute diffusion; Need for new membrane development and draw solute design	Costs ↓ (no data available)	Energy use ↓ (to be confirmed): 3-8 kWh m <sup>-3</sup> ; Chemical use ↓: less cleaning	Full-scale: desalination, food processing; Full-scale digestate/ manure: in 5-10 y	83, 89, 90
	Physicochemical: transmembrane chemisorption	Liquid	70-99 % NH <sub>3</sub>	Membrane clogging	Depends on mass transfer; Similar costs as stripping	Energy use ↓: < 1 kWh m <sup>-3</sup> ; Chemical use (acid) ↑	Pilot	95, 96, 97
	Electrochemical: electrodialysis	Liquid	80-83 % NH <sub>3</sub>	High energy consumption; Variable costs for membranes, electrodes, cases; Acid NH <sub>3</sub> trap required; Post-treatment (RO/stripping) required	High costs (no data available)	Energy use ↑: 3.25-3.60 kWh kg <sup>-1</sup> NH <sub>4</sub> -N or 1.2-1.5 kWh kg <sup>-1</sup> K; NH <sub>3</sub> volatilization	Full-scale: limited; Lab: digestate/manure	80, 91, 92, 94
NH <sub>3</sub> sorption	Physicochemical	Liquid	18 % P 15-60 % NH <sub>3</sub>	Fouling of the packing column; Regeneration and maintenance; Post- and/or pre-treatment required	Potentially low costs relative to other technologies (depending on availability, pre-treatment, and regeneration); Further research needed for digestate treatment	Energy use ↓; Chemical use ↑	Full-scale: limited for wastewater; Lab: digestate	13, 39, 40, 41, 98
Biomass production and harvest	Biological	Liquid	84-98 % N 90-99 % P	Harvest method; Reduced light penetration; Dilution often required; Large surface area; Toxic if N > 60 mg L <sup>-1</sup>	Capex: > € 80,000 ha <sup>-1</sup> ; Overall (macrophytes): € 12-33 PE <sup>-1</sup> y <sup>-1</sup> Overall (algae): € 4-300 kg <sup>-1</sup> dry weight	Surface ↑: 3.4-8.3 m <sup>2</sup> PE <sup>-1</sup> ; Energy use ↑ (CO <sub>2</sub> addition); Polymer use ↑	Pilot/Full-scale: duckweed; Mostly lab: algae	32, 117, 118, 119, 122, 123, 124
P extraction from ashes/ bio-char	Thermochemical / Wet-chemical	Solid	Up to 78 % P	Often heating, flue gas cleaning, and heavy metal removal required; pH, temperature, and chemical choice are critical	< € 1 m <sup>-3</sup> slurry (wet extraction); € 3 ton <sup>-1</sup> slurry (pyrolysis); € 0-10 m <sup>-3</sup> slurry (combustion)	Combustion = sustainable?!; Energy use ↑: temperature- dependent; Chemical use ↑: process- dependent	Full-scale: incinerated sludge; Lab: incinerated digestate, but often not authorized!	45, 128, 129

<sup>a</sup> ↑ = relatively high; ↓ = relatively low; CAPEX = capital expenditures; OPEX = operational expenditures; PE = person equivalent; RO = reversed osmosis; R&D = research and development.

**Table 2:** Classification of recovered end-products: class, technology, feedstock, product, composition/quality, current marketing value, and reference.

Class	Technology	Feedstock	Product	Composition/quality	Marketing value	References
N/P-precipitates	Struvite precipitation (chemical)	Liquid fractions, (acidified) raw digestate	MgNH <sub>4</sub> PO <sub>4</sub> ·6H <sub>2</sub> O	Pure struvite: 6 % N, 29 % P <sub>2</sub> O <sub>5</sub> , 10 % Mg; Ostara: 5 % N, 28 % P <sub>2</sub> O <sub>5</sub> , 10 % Mg; Crystalactor: 9 % N, 46 % P <sub>2</sub> O <sub>5</sub> , 16 % Mg; Crystals (0.5-5 mm); Slow-release; Purity?!	€ 45-1,393 ton <sup>-1</sup>	12, 54-60 Technology providers
	Struvite precipitation (electrochemical)	Liquid fractions	MgNH <sub>4</sub> PO <sub>4</sub> ·6H <sub>2</sub> O	> 90 % purity; R&D needed	R&D needed	61, 139
	Struvite precipitation (bio-electrochemical)	Liquid fractions	MgNH <sub>4</sub> PO <sub>4</sub> ·6H <sub>2</sub> O	R&D needed (40 % soluble P removal)	R&D needed	62
K/P-precipitates	Struvite precipitation (chemical)	Liquid fractions, (acidified) raw digestate	KMgPO <sub>4</sub> ·6H <sub>2</sub> O	R&D needed	R&D needed	49
P-precipitates	Calcium phosphate precipitation	Liquid fractions, (acidified) raw digestate	Ca <sub>5</sub> (PO <sub>4</sub> ) <sub>3</sub> OH CaHPO <sub>4</sub> ·2H <sub>2</sub> O	P <sub>tot</sub> : 10-11 %; Crystals; Purity?!	R&D needed	63, 65
P-extracts	P extraction	Ashes/biochar	Acid P-extract, CaHPO <sub>4</sub> , struvite, Fe/Al-PO <sub>4</sub>	P <sub>2</sub> O <sub>5</sub> : 15-35 %; High P bio-availability; Purity?!	€ 0.89-4.25 m <sup>-3</sup>	45, 129
N/S-solutions	Stripping & absorption	(Decarbonated) liquid flows	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> -solution	AmS: 25-38 %; N: 30-100 kg m <sup>-3</sup> ; S: 61-100 kg m <sup>-3</sup> ; pH: 3-7; High salt content: 30-150 mS cm <sup>-1</sup>	€ 90-120 ton <sup>-1</sup>	12, 20, 21, 22, 34, 67 Technology providers
	Acidic air scrubbing	Strip gas, air	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> -solution	N <sub>tot</sub> : 30-70 kg m <sup>-3</sup> ; S: 61-100 kg m <sup>-3</sup> ; pH: 3-7; High salt content: 30-150 mS cm <sup>-1</sup>	€ 90-120 ton <sup>-1</sup>	12, 20, 21, 22 Technology providers
	Transmembrane chemisorption	Tested on urine; Potential for liquid fractions of manure/digestate	(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> -solution	Several 100 g NH <sub>4</sub> <sup>+</sup> L <sup>-1</sup>	R&D needed	95
N/K-concentrates	Reversed osmosis	Permeate from ultrafiltration, microfiltration or dissolved air flotation	N/K-concentrates	N <sub>tot</sub> : 3-11 kg ton <sup>-1</sup> ; 92 % NH <sub>4</sub> -N, 8 % organic N; K <sub>2</sub> O <sub>tot</sub> : 5.0-13.6 kg ton <sup>-1</sup> ; P <sub>2</sub> O <sub>5</sub> <sub>tot</sub> : 0-1.4 kg ton <sup>-1</sup> ; Purity?!	€ 1.19-1.25 ton <sup>-1</sup>	19, 77, 78
	Forward osmosis	Liquid fractions	N/K-concentrates	R&D needed; Potential for high-quality product through high rejection	R&D needed	83, 89, 90
	Electrodialysis	(Filtrated) liquid fractions	N/K-concentrates	R&D needed; 7 x the input concentration	R&D needed	80, 91, 92, 94
N-zeolites	NH <sub>3</sub> sorption	(Filtrated) liquid fractions (to be confirmed for digestate)	N-enriched Clinoptilolite	Slow-release fertilizer; Potential contamination (metals, etc.); R&D needed	R&D needed	13, 39, 40, 98
Biomass	Biomass production & harvest	Diluted liquid fractions	Biomass (algae, macrophytes)	Duckweed: 30 % P on dry weight; High content of proteins, N, P, K, C; Potential for biofuel and chemical industry, or as animal feed	€ 5,300 y <sup>-1</sup> for a protein yield of ± 11 ton ha <sup>-1</sup> y <sup>-1</sup>	113, 115, 125, 126, 127

