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REVIEW

Nutrient Recovery from Digestate: Systematic Technology Review and Product Classification

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

Keywords Anaerobic digestion · Bio-based fertilizers · Residuals management · Sustainable agriculture · Sustainable resource management · Waste valorization

Introduction

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

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

So far, the technical approach for digestate processing was similar to the approach for the treatment of manure and wastewater. This means that the focus was on low cost-effective, energy-intensive, and non-sustainable nutrient removal practices through destruction or emission, e.g. biological nitrification/denitrification [13]. The challenge for anaerobic digestion plants now is to achieve optimal recovery and recycling of nutrients from the digestate in a sustainable way. As such, regulatory drivers can be met and an internal revenue source can be produced, i.e. the present 'waste' problem can be turned into an economic opportunity.

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

The aim of this review paper is to provide a systematic overview and a critical comparison of technologies for the recovery of macronutrients from digestate, as well as a classification of the resulting end-products based on their fertilizer characteristics. The focus is on the recovery of N, P, and potassium (K), but parallel attention is given to sulfur (S), calcium (Ca), and magnesium (Mg). The systematic review involves an exhaustively search of all the relevant peer-reviewed literature, as well as grey literature and unpublished research findings. Moreover, an intensive discussion platform was established involving existing international academic experts, administrations, and companies active in the field of nutrient recovery. Included and excluded data were discussed with the expert panel and selected on the basis of predefined criteria. Financial and technical aspects for the most established technologies at full-scale were further investigated by requesting budget proposals using a predefined questionnaire to key technology suppliers in the field. The survey involved capital and operational costs, use of consumables, recovered product quality, and potential revenues, among other technical items.

Based on the findings, first the general composition of digestate is briefly discussed ("Digestate Characteristics" section). In the core of this paper, the technical and economic state-of-the-art of the existing technologies and those under development is explored ("Technology Overview" section), and available information on product quality and value is compiled ("Classification of Recovered Products" section). Market trends and outlook are also discussed ("Fertilizer Market Trends and Outlook" section). As such, this review paper can provide the fundamental basis to classify and categorize recycled products in environmental and fertilizer legislations, thereby stimulating their economic valorization as marketable commodities. This, in turn, may foster the development and implementation of innovative technologies for nutrient recovery from digestate.

Digestate Characteristics

Digestate is the remaining product after biogas production in an anaerobic digester. It contains the non-digested recalcitrant organic fraction, water, micro- and macronutrients [19, 21–23]. The composition of digestate varies strongly according to the composition of the feedstock (biodegradable waste) that is digested, next to the digester type and process parameters. Hence, giving a standard composition of digestate is not possible. Because of this constraint, 213 digestates from different (co-)digestion plants in Flanders (Northern part of Belgium confronted with high nutrient pressure) were sampled and analyzed during 4 years (2008–2011) [24]. Product quality ranges are compiled in Table 1. Based on the results, a short overview of how physicochemical characteristics change during the digestion process and how the feedstock influences the digestate composition is given below.

During anaerobic digestion, easily degradable organic matter is converted into methane (CH_4) and carbon dioxide (CO_2), while complex organic matter, such as lignin, remains in the digestate, thereby increasing its amount of effective organic carbon (OC). This is the OC that remains in the soil after 1 year and thus contributes to the humus built-up [average: 33.7 kg ton^{-1} in digestate vs. 20.0 kg ton^{-1} in pig manure on fresh weight (FW)] [24]. As such, the digestate contains important soil improving qualities [25]. The percentage of organic dry matter can vary between 30 and 80 %, with lower values for increasing slurry fractions and higher values for increasing fractions of kitchen and garden waste (KGW). The average dry matter content of 100 % KGW was estimated at 21 % [25], whereas the median dry matter content of the 213 studied digestates amounted to 8.7 % (Table 1).

Due to the degradation of more than 90 % of volatile fatty acids (VFA's), the pH is increased and odor emissions are significantly lower during the application of digestate on agricultural fields as compared to pig slurry [26]. The pH of slurry is on average 7 [125], whereas the median digestate pH amounts to 8.3 (Table 1). However, the higher pH causes an increased risk for NH_3 volatilisation. This is why injecting the digestate is strongly advised [25].

Next, during the digestion process, organically bound N is released as ammonium (NH_4^+), which is directly available for crop uptake. The higher the share of NH_4-N, the higher the efficiency of the digestate as a N-fertilizer. An input stream with a high N-level is, for example, pig slurry (average: 6.78 kg N ton^{-1} FW), in comparison to cattle slurry (3.75 kg N ton^{-1} FW) and maize (4.00 kg N ton^{-1} FW) [25]. The N-content of sewage sludge usually ranges between 0.1 and 1 kg N ton^{-1} FW [12]. The Vlaco [24] data showed a median total N-content of 4.2 kg N ton^{-1} FW (Table 1). When digesting raw pig slurry, more than 80 % of the N becomes available as NH_4^+. However, for digestates produced from organic waste such as KGW, the share of NH_4^+ is often not higher than 44–47 %, which is even lower than the value for raw pig slurry (±60 %). Digestates with a low NH_4-N-content are mostly originating from organic food/industrial wastes, including KGW [25].

Furthermore, the total P-content of the input streams is not changed during the digestion process, but the organically bound P becomes available for the plant during digestion. Pig slurry has a high P_2O_5-content of about 5 kg ton^{-1}. By adding co-products to pig slurry the P_2O_5-content of the digestate is somewhat lowered. The P_2O_5-content of sewage sludge usually ranges between 0.04 and 0.7 kg ton^{-1}, with exception of sludge produced by enhanced biological phosphorus removal (EBPR), which can contain up to 15 kg P_2O_5 ton^{-1}. The 213 studied digestates showed a median total P_2O_5-content of 3.9 kg ton^{-1} FW (Table 1).

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

Finally, impurities such as weed seeds and pathogens can be killed off during the digestion process. The extent to which this inactivation is sufficient depends entirely on the temperature and residence time in the digester and on the type of organism [25]. The effect of anaerobic digestion on persistent organic pollutants requires further study.

Table 1 Composition (10th percentile, median, 90th percentile; No. of observations: 213) of unprocessed digestate [24]

Parameter	Unit	Unprocessed digestate		
		10-perc.	Median	90-perc.
Dry weight	w%	4.98	8.70	12.0
Organic matter	w%	2.8	5.3	7.6
pH (H_2O)	–	8.1	8.3	8.6
Electrical conductivity	mS cm^{-1}	20	32	45
Total N	w%	0.17	0.42	0.75
NH_4-N	g L^{-1}	0.52	2.15	3.41
NO_3-N	mg L^{-1}	3.10	5.85	10.0
C:N-ratio	–	3.89	6.58	13.7
Total P_2O_5	w%	0.14	0.39	0.65
Total K_2O	w%	0.20	0.35	0.50
Total CaO	w%	0.16	0.30	0.55
Total MgO	w%	0.03	0.09	0.20

w% = % on fresh weight

Technology Overview

Digestate Processing Technologies

In the past decade, a diverse range of technologies that can be applied for digestate processing has been developed. However, certainly not all of them can be considered as an NRT. To date, there is no straightforward definition of an NRT. In this review we consider an NRT as a process that: (1) creates an end-product with higher nutrient concentrations than the crude digestate (=concentrated product that contains both minerals and organics), or (2) separates the envisaged nutrients from organic compounds, with the aim to produce an end-product that is fit for use in the chemical or fertilizer industry or as a chemical fertilizer substitute. The breakthrough of such technologies would make it possible to reuse reactive nutrients locally and close nutrient cycles in a cross-sectorial cradle-to-cradle approach (Supporting Information 1). An overview of existing digestate processing technologies is presented in Fig. 1. The technologies delineated as NRT are indicated in shaded boxes. It must be noted that these technologies could also be applied on undigested manure, sludge, and wastewater. However, as mentioned above, implementing anaerobic digestion as an intermediate step can highly improve the overall process efficiency. In some cases opportunities for direct reuse, e.g. through fertigation or reuse of nutrients via irrigation with (waste)water containing nutrients, may also exist [28, 29].

In general, digestate processing starts with a mechanical separation into a liquid fraction and solid fraction aiming at dewatering [30]. The solid fraction is rich in recalcitrant organic matter, Ca, Mg, and often P, but offers limited possibilities to recover nutrients, since they are largely organically bound. Soluble N, P, K, organics and mineral salts are present in the liquid fraction. The potential to recover soluble nutrients from the liquid fraction by use of extraction techniques is high [19].

From literature, the technologies for nutrient recovery from the liquid fraction available or under development today are: (1) chemical crystallization [2, 14, 31], (2) gas stripping and absorption [32–34], (3) acidic air scrubbing [32, 35], (4) membrane separation [36–38], (5) ammonia sorption [39–41], and (6) biomass production and harvest [42, 43]. For the solid fraction, only P extraction from ashes produced by combustion or pyrolysis can be considered a potential NRT [44, 45]. However, as it is critically questioned whether incineration is a sustainable technology, the interest is growing to extract nutrients from the crude digestate, thereby eliminating the solid–liquid separation step or producing an organic, P-poor thick fraction.

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

Phosphorus Precipitation and Crystallization (NRT 1)

Struvite Recovery

Nutrient recovery through P precipitation and crystallization is a mature technology, mostly involving the addition of Mg ($MgO/MgCl_2$) and caustic soda (NaOH) to a solution containing soluble PO_4-P (ortho-P) and ammonium, thereby increasing the pH to 8.3–10 and inducing the precipitation of struvite, $MgNH_4PO_4 \cdot 6H_2O$ [14]. The process has been implemented at full-scale installations for wastewater (digested) sludge, and manure treatment, as well as at pilot-scale for the treatment of crude digestate. The most established processes commercially available today are: (1) *AIRPREX*, Berliner Wasserbetriebe (Germany, DE), (2) *ANPHOS,* Colsen (the Netherlands, NL), (3) *CAFR,* NALVA (DE), (4) *Ceres*, Ceres Milieutechniek (Belgium, BE), (5) *NuReSys*, Akwadok (BE) [46], (6) *Nutritec*, Sustec (NL), (7) *Pearl,* Ostara (Canada, CA), (8) *Phosnix*, Unitika (Japan, JP), (9) *PHOSPAQ,* Paques (NL) [47], and (10) *PRISA*, Aachen University (DE) [48]. Moreover, in Gelderland (NL) four installations are available for the production of K-struvite ($KMgPO_4 \cdot 6H_2O$) from calf manure [49]. These processes have the ability to remove and recover over 80–90 % of the soluble P in the waste(water) flow, yet only 10–40 % of the NH_4-N can be captured [14]. Crystal/pellet sizes range from 0.5 to 5 mm and above, depending on the final end-use. The design involves fluidized bed reactors and continuously stirred tank reactors.

At present, struvite recovery can be economical on side streams from wastewater treatment with a P-load of more than 20 % by weight, as it has the potential to reduce operating costs by reducing energy and chemical (iron (Fe)/aluminium (Al)) consumption and minimizing nuisance struvite formation in piping/equipment. Meanwhile, a high-quality, slow-release granular fertilizer with agricultural reuse perspectives is produced [50–53]. Assuming that a treatment plant can recover 1 kg of struvite from

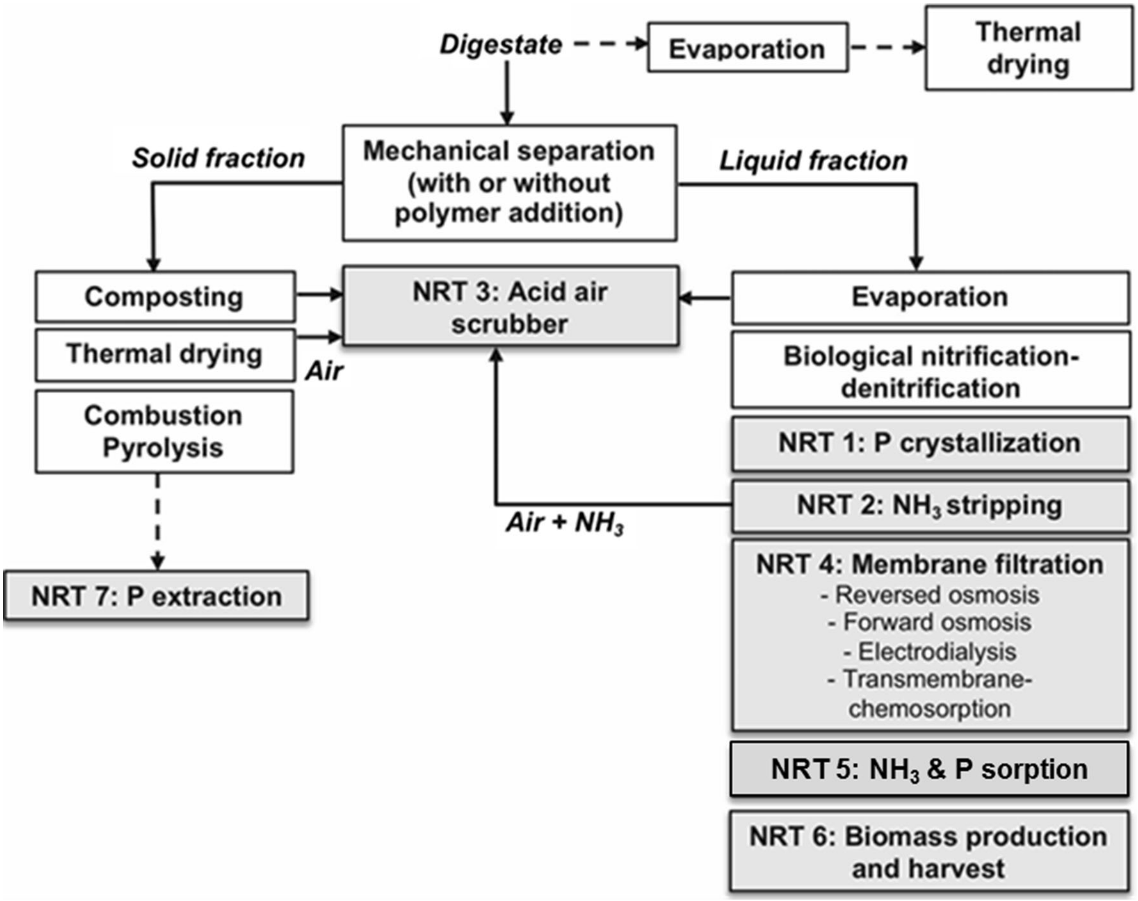

Fig. 1 Schematic overview of digestate processing technologies. *Shaded boxes* nutrient recovery technology (NRT)

100 m^3 of wastewater [54], Shu et al. [52] estimated chemical savings for P recovery from digester supernatants at € 0.19 day^{-1} (compared to alum), savings from reduced sludge handling at € 0.002 day^{-1}, from reduced sludge disposal at € 0.023 day^{-1}, from reduced cleaning of struvite deposits at € 3.8–19.2 day^{-1} and savings from reduced landfilling at € 0.002 day^{-1}. As such, the cost of producing struvite could amount to € 0.05 day^{-1}, while the gain for the treatment plant can be € 0.52 day^{-1} [52]. Hence, assuming a struvite plant would cost € 1.4 million, the payback period of a plant processing ±55,000 m^3 day^{-1} of wastewater could be less than 5 years according to this study [52].

However, operational costs and payback times are highly dependent on the input composition (e.g. available P, Mg, and pH) as it determines the chemical (NaOH, Mg) and energy costs, which can range between 200 and 75,000 € $year^{-1}$ [55]. Dockhorn [56] estimated operating and maintenance costs for a plant treating 350,000 person equivalents (PE) at € 2800 ton^{-1} of struvite if the PO_4-P-concentration is 50 mg L^{-1}, and € 520 ton^{-1} if the PO_4-P-concentration is 800 mg L^{-1}. Battistoni et al. [57, 58] estimated operating costs at € 0.19–0.28 m^{-3} digestate. Based on budget proposals provided by the above-mentioned suppliers in the context of the present review, capital costs may range from 2300 to 24,500 € kg^{-1} P day^{-1}, while revenues from struvite valorisation in agriculture range from ± € 45 ton^{-1} struvite in Belgium (NuReSys, Waregem, BE, personal communication 2013) to ±€ 109–314 ton^{-1} in Australia [59], and ±€ 250 ton^{-1} in Japan [60]. Values of € 736 and € 1393 ton^{-1} have also been reported [56]. As such, overall profits of struvite production may range from € −7800 $year^{-1}$ (loss) to € 89,400 $year^{-1}$ (gain) [54].

Although world-wide some utilities have installed these systems, the uptake of this technology has not been widespread due to market, regulatory, and site-specific conditions. Also, important technical challenges remain in the reduction of chemical requirements, the guarantee of a pure product, as well as the stable and controlled production of struvite. Energy-efficient methods without chemical addition, such as electrochemical [61] and bio-electrochemical [62] struvite recovery, are under development (Supporting Information 2).

Calcium Phosphate Recovery

Next to Mg, calcium hydroxide ($Ca(OH)_2$) can also be added to the liquid fraction in order to increase the pH (>10.0) and temperature (70 °C), thereby inducing P precipitation as $Ca_5(PO_4)_3OH$ (hydroxyapatite) or $CaHPO_4 \cdot 2H_2O$ (brushite). The reaction is fast (5 min), but often preceding CO_2-stripping must be applied to avoid unwanted calcium carbonate ($CaCO_3$) precipitation. Examples of commercial calcium phosphate precipitation processes are: (1) *Crystalactor*, DHV Water (NL) [63], (2) *FIX-Phos,* TU DA (DE), (3) *Kurita*, Kurita Water Industries (JP), (4) *Phostrip*, Tetra Technologies inc. (USA) [64], and (5)

P-Roc, Kit-CMM (DE) [65]. Removal efficiencies of 80–100 % P have been achieved, but 50–60 % is more typical. Based on a market demand in the framework of this study to the suppliers mentioned above, capital costs may range between € 2300–2900 kg^{-1} P day^{-1}. Operational costs are mainly determined by the amount of $Ca(OH)_2$ required, which on its turn depends on the input waste characteristics. Finally, Quan et al. [66] reported on the ability to couple $CaNH_4PO_4 \cdot 4H_2O$ precipitation and ammonia stripping in a water sparged aerocyclone (WSA) in order to recover both P and N. To date, this path has only been examined at lab-scale, but further research on this methodology seems interesting.

Ammonia Stripping and Absorption (NRT 2)

Stripping of ammonia (NH_3) involves the physical transfer of NH_3 from the aqueous phase (waste stream) to a gas phase, mostly in a packed bed tower. The gas is then transferred to an air scrubber (Sect. 3.4), where mass transfer and absorption of the NH_3 from the gas to a liquid phase, often sulfuric acid (H_2SO_4), takes place in order to form and recover a concentrated solution of ammonium sulfate ($(NH_4)_2SO_4$; AmS) as an end-product [34, 67]. AmS is an inorganic salt, which could be reused as a marketable fertilizer rich in direct available macronutrients, N and S, thereby providing a valuable substitute for chemical fertilizers based on fossil resources [20–22]. Alternatively, ammonium nitrate (NH_4NO_3) fertilizer (by addition of nitric acid, HNO_3) [68], a concentrated ammonia solution (by addition of liquid NH_3), or other ammonia salts can be produced, depending on local legislation and options for land application. Ammonia stripping is developed at full-scale, and sometimes implemented for wastewater treatment. The implementation of this technology for the treatment of N-rich digestate and manure is on the rise.

Commercially available stripping technologies for (digested) sludge and manure treatment are (1) *AMFER*, Colsen (NL), (2) *ANAStrip*, GNS (DE), and the (untitled) stripping processes developed by the manufacturers: (3) Anaergia (Canada, CA), (4) Branch Environmental Corp (USA), (5) Europe Environnement (France, FR), and (6) RVT Process Equipment (DE). Theoretically, these systems may achieve NH_3-removal efficiencies up to 98 %, but they are generally operated to reach 80–90 % removal in order to reduce the operating costs. At present, most stripping units implemented at full-scale focus on the production of AmS-fertilizer. The AmS-content in the recovered solution ranges from ±25 % AmS (*ANAStrip*, GNS) and 30 % AmS (Branch Environmental Corp) to ±38 % AmS (Anaergia; RVT Process Equipment) and 40 % AmS (*AMFER*, Colsen; Europe Environnement).

Capital costs of stripping are relatively low compared to biological activated sludge (AS) systems and depend on the method used for pH-increment. This can occur (1) chemically by use of base, mostly sodium hydroxide (NaOH) (Branch Environmental Corp; Europe Environnement; RVT Process Equipment), or (2) physically by simultaneous stripping of CO_2 (Anaergia; Colsen), optionally in combination with the addition of low-quality gypsum ($CaSO_4$) for parallel recovery of $CaCO_3$ (GNS). Operational costs depend a lot on the operational temperature, pH, and liquid flow rate. For a 90 % NH_3-recovery efficiency from leachate at a temperature of 70 °C, a pH of 11, and a flow rate of 70 m^3 h^{-1}, overall costs are estimated at ±€ 8.1 (±US \$ 10.2) m^{-3}, while at a temperature of 30 °C this would be ±4 times less, i.e. € 2.0 (±US \$ 2.5) m^{-3} [69].

A comparison of budget proposals provided by the above suppliers for NH_3-stripping and absorption systems treating a digestate flow of 800 m^3 day^{-1} at 2400 mg NH_4-N L^{-1} (90 % recovery) resulted in a capital cost ranging from € 500,000 to € 1.58 million if the pH-increase is conducted chemically, and from € 3.5 million to € 11–15 million if the pH-increase is established physically. Electricity consumption for this case was estimated by the suppliers at 127–400 kW_{el}, heat consumption at 2115–2333 kW_{th}, and H_2SO_4-consumption (concentrated at 95–97 %) at 5.5–6.8 ton day^{-1} or 7.0–10 kg m^{-3} digestate. If NaOH is used for pH-increase, its consumption would amount to 6.0–6.5 kg m^{-3}. As such, operational costs range from € 1.4–2.5 million $year^{-1}$ depending on the system, equivalent to € 4.5–8.6 m^{-3} of digestate.

Currently, advanced processes for biological removal (note: no recovery) of NH_3, such as deammonification via Anammox [70], are often still cheaper (depending on the technology provider). However, as stripping could (partially) replace a nitrification–denitrification step, additionally remove odorous compounds and dust particles, and produce a marketable end-product, it is expected that this technology can be competitive, especially in regions where N-demand is high [53]. Where commercialisation of the

AmS-solution is possible, revenues currently range from € 90 to 120 ton^{-1} FW, which under optimal process conditions should largely compensate the operational costs. Moreover, the higher process stability (e.g. to input variation, freezing conditions, etc.), lower surface requirements (for the above case: ±1500 m^2), higher ability for treatment of high N-flows, immediate start-up, and ease of automation are all drivers for implementation of stripping units instead of conventional biological systems.

The major technical bottlenecks observed to date in NH_3-stripping are scaling and fouling of the packing material, and the consequent high energy and chemical requirements [13, 32, 66]. To avoid scaling, one can install a lime softening step before stripping, which removes a large part of the Ca, Mg, carbonic acids and carbonates, and increases the pH. In case of high buffering capacity, a preceding CO_2-stripper might also be economical. To avoid fouling, it is important that during preceding solid–liquid separation as many suspended solids as possible are retained in the solid fraction. Nonetheless, it is unavoidable that the packing material will have to be cleaned periodically. Because of these constraints, some of the above manufacturers have developed a stripping process without internal packing (Anaergia; Colsen). As such, the process developed by Anaergia would be capable of handling waste flows containing up to 8–9 % total suspended solids (TSS). Note that both technologies also operate without any chemical addition. Hence, although capital costs are higher (see above), in terms of sustainability and operational costs, these processes may be of interest. Alternative NH_3-stripping systems aiming to overcome the above technical bottlenecks, such as a WSA reactor [66] and the use of rotating disks [71], are also being developed (Supporting Information 3).

Acidic Air Scrubbing (NRT 3)

Acidic air scrubbing mostly concerns a packed tower in which sulfuric acid (H_2SO_4) is sprayed with nozzles over the packing material and treatment air is blown into the tower in counter-current [35, 67]. The technology can be applied for air treatment, for example, from stables or following NH_3-stripping from liquid waste flows ("Ammonia Stripping and Absorption (NRT 2)" section). In most cases, ammonium sulfate ($(NH_4)_2SO_4$; AmS) is produced and the wash water is recycled until it is saturated and the removal efficiency of NH_3 cannot be guaranteed anymore. At that point, the AmS-solution should be removed and fresh H_2SO_4 added. The technology is used at full-scale at anaerobic digestion and sludge/manure processing plants. Examples of well-established technology developers are Dorset Farm Systems (NL/US), Envitech (CA), and Inno + (NL). Average NH_3-recovery efficiencies of 91–99 % are found in literature [50, 67, 72]. Investment costs (in case of a new installation for air treatment of one stable) are estimated at € 18 kg^{-1} NH_3-removal, whereas exploitation costs (including variable and fixed costs) of an acidic air scrubber are estimated at € 6–7 kg^{-1} NH_3-removal [73, 74]. As these costs are expected to reduce with 50 % (investment) and 14–25 % (exploitation) for large-scale projects [74, 75], the installation is economically viable at many waste-processing plants.

The main operational costs can be attributed to the energy (0.057 kWh 1000 m^{-3} air) and acid (1.5 L H_2SO_4 at 98 % kg^{-1} NH_3-recovery) requirements [74]. However, power inputs depend a lot on the reactor type, ranging from 3.8 atm cm^3 air s^{-1} for spray-chambers to 260 atm cm^3 air s^{-1} for venturi scrubbers [76]. Interesting advantages of an acidic air scrubber is that odors, dust particles, and water vapour can also be removed. Technical bottlenecks are mainly related to corrosion problems. The reject AmS-solution is recognised in Flanders and the Netherlands as a mineral fertilizer in environmental legislation. However, marketing is still hindered due to its variable N and S content (30–100 kg N ton^{-1}; 61–100 kg S ton^{-1}), acidic and corrosive features (pH 2.5–7, high salt content: 100–150 mS cm^{-1}) as well as social perception and farmers' distrust in its fertilizer properties [21–23]. It should be noted that the product properties are highly dependent on the technology provider, not only in terms of AmS content (see above), but also in terms of pH. Manufacturers delivering a fertilizer product at relatively high pH, suitable for direct application in agriculture, are Anaergia (pH 5.5) and RVT Process Equipment (pH 6–7).

Membrane Filtration (NRT 4)

Pressure-driven membrane filtration, involving microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and/or reversed osmosis (RO) is an established technology in wastewater treatment. It has, however, not yet proven to be a valuable option for the treatment of digestate, manure, and sludge. Only a few commercial pilots have been installed at full-scale manure and digestate processing facilities, mostly on a short-term basis because of excessive operational costs. Nevertheless, the produced membrane filtration concentrates are an interesting nutrient source, which could potentially be reused as chemical fertilizer substitutes rich in N and K [19, 37, 76, 77]. Examples of manufacturers for slurry filtration systems are: (1) A3 Watersolutions (DE), (2) AquaPurga International (NL), (3) New Logic (USA), (4) VP Systems (NL), and (5) Wehrle Umwelt GmbH (DE). Operating temperatures range from 10 to 40 °C, while the pH is usually between 6 and 8. RO has also been applied at full-scale in combination with NH_3-stripping of liquid digestate (*Biorek Process*, BIOSCAN (Denmark, DK)) [79].

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

The biggest technical problem stated in membrane filtration is clogging and fouling of the membrane, resulting in significant chemical and energy requirements [36, 38]. The equivalent energy requirement for an RO-system is estimated at 4–6 kWh m^{-3} [82, 83]. To reduce cleaning requirements, vibrating (60–90 Hz) shear enhanced processing (VSEP) has been used for manure and digestate purification at pilot-scale [19, 84, 85]. However, little data are available on the energy consumption and treatment costs of this technology. The energy consumption per vibration is estimated at 8.83 kW (G. Johnson, New Logic Inc., Ontario, CA, personal communication 2011), while the energy consumed by the recirculation pump is estimated at 9.4 kWh m^{-3} of permeate in a 154 m^2 membrane area unit (VSEP series i-10) [86]. This could be reduced to 6 kWh m^{-3} if plane ceramic membranes are used. Energetic calculations based on these data indicate that large VSEP-units will consume significantly less energy per m^3 of permeate than traditional cross-flow filtration. Nevertheless, energy consumption and economic performance remain critical points of attention in the evaluation of membrane technologies for nutrient recovery. There are also concerns about the potential pollution of the recovered concentrates, e.g. with heavy metals and other contaminants.

Different alternative technologies to improve the performance of membrane filtration in terms of chemical and energy requirements, as well as operational costs are currently under development. The most studied examples are forward osmosis [87–90], electrodialysis [91–94], and transmembrane chemosorption [95–97] (Supporting Information 4).

Ammonia and P Sorption (NRT 5)

A number of materials may be used to selectively adsorb ammonium (NH_4^+) and P from waste flows. These materials include zeolites, clays, and resins, whether or not chemically or thermally modified. Adsorption is carried out in a packed column. Once the adsorption media is saturated, the column is taken offline and is regenerated to recover the NH_4^+ and/or P as a concentrated solution and to reuse the adsorption media. Regeneration can be achieved by a number of techniques, including nitric acid (HNO_3) washing, sodium chloride (NaCl) washing, or biologically. The technique to be used depends on the adsorption material and the desired end-product. Adsorption can therefore either be operated as a batch process using a single column, or a series of multiple columns can be sequenced to provide continuous operation [40, 98, 99].

In the context of wastewater treatment, especially natural zeolites have been succesfully used as adsorption agent for final NH_4-N-removal [30, 100–107]. Mainly the use of clinoptilolite $(Na, K, Ca)_{2-3}Al_3(Al, Si)_2Si_{13}O_{36}\cdot 12(H_2O)$, has been studied because of its low-cost availability [40]. However, full-scale wastewater treatment plants that employ the NH_4^+-ion exchange technique are scarce, and few applications have been developed to recover NH_4-N, for example, for agricultural purposes [40, 108]. Nevertheless, because the zeolite is porous, the ammonium can leak out of the zeolite at a much slower rate than it was adsorbed. This makes the ammonium-filled clinoptilolite itself a potential slow-release fertilizer [98]. An important remark is that the initial N-concentration in the above-mentioned applications was only a few 10 mg L^{-1}. Over the past decade, there has been increasing interest of using natural zeolite for NH_4-N-removal from waste streams with relatively high N-concentration or high ionic strength [109–112]. However, its applicability in practice for the treatment of the liquid fraction of digestate (containing both high N- and ionic concentrations) still remains to be demonstrated, as does the use of the nutrient-enriched clinoptilolite or other regenerated N-solutions as a fertilizer [13, 39, 40].

To date, removal efficiencies of 18 % P (probably due to adsorption) and 15–60 % N (due to ion exchange) have been reported for the treatment of human urine using clinoptilolite at lab-scale [98]. This means that the technology would currently not be feasible as stand-alone NRT for digestate processing, though it may be used as an intermediate step in the digestate treatment train. As such, some research has been performed towards the combined use of zeolite and struvite precipitation to obtain a slow-release fertilizer with both high N- and P-concentrations. As such, total recovery efficiencies of 100 % P and 83 % N have been obtained at lab-scale, and the fertilizer potential of the resulting product has been demonstrated at greenhouse scale [98]. Moreover, Liberti et al. [113] investigated a system where NH_4^+-ions were concentrated by the NH_4^+-ion exchange technique, followed by air stripping of NH_3-gas, which was subsequently absorbed in sulfuric acid. Zeolites may also be used to further treat the effluent produced by membrane filtration of the liquid fraction of digestate [39]. Hence, although there is potential to use zeolites as a technology for nutrient recovery, to date the use of this process for digestate treatment can rather be considered as an intermediate or final concentration step in the three-step framework (Fig. 1).

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

Furthermore, for concentrated waste streams (>2000 mg solids L^{-1}), typically, red mud, metal oxide/hydroxide and zirconium sorbents are used for P recovery. Phosphorus may be removed from solution via selective sorption to a solid phase and direct use of the material as a fertilizer or soil conditioner, or the sorbed P may subsequently be stripped from the solid sorbent and chemically precipitated as a high-purity fertilizer. However, as is the case for natural zeolites (see above), further research is required on the use of these sorbents for digestate treatment [99].

Finally, costs of this technology are expected to be low, but will depend on the nearby availability of the sorption material used, the required pre-treatment of the packing column to obtain NH_4^+- and/or P-selectivity, the recovery/regeneration method (if applicable), and the required frequency of regeneration. No cost-benefit analyses for nutrient recovery from digestate using zeolites or other sorbents have been reported on in literature to date.

Biomass Production and Harvest (NRT 6)

Both macrophytes (mostly duckweeds and water hyacinths) and microalgae have been examined for biological nutrient recovery [15, 114, 115]. Duckweed (*L. minor*, *L. Punctate*, *S. polyrrhiza*, *S. Oligorrhiza*) ponds have been successfully used as an efficient and potentially low-cost option in (anaerobically digested) agricultural waste polishing, generating a biomass with high protein content. A detailed overview of the nutrient and heavy metal content of duckweed in function of water quality has been reported [116, 117]. Based on its mineral composition, the plant appears to have the ability to recover 600, 56–140, 400, 100, 60, 32, and 24 kg ha^{-1} $year^{-1}$ of N, P, K, Ca, Mg, Na, and Fe from water bodies, respectively at a production of 10 tons dry weight (DW) ha^{-1} [117]. Xu and Shen [43] found removal efficiencies of 83.7 and 89.4 % for total N and P from pig wastewater, respectively, using *S. oligorrhiza* in 8 weeks at a harvest frequency of two times a week. Mohedano et al. [118] found an average of 98.0 % total N and 98.8 % total P recovery from swine wastewater at full-scale, resulting in an average biomass (*L. punctate*) protein content of 28–35 % on FW. However, above 60 mg N L^{-1} a toxic effect was noticed perhaps due to high levels of free ammonia in the water, hence levels below this value should be maintained in order to obtain a consistently high protein content (15–45 % by DW, depending on the N-supply [114]).

Skillicorn et al. [119] estimated the capital costs for a 0.5 ha large duckweed system at ±€ 2600 (US $ 3300). However, capital costs are significantly influenced by land area requirements, next to the costs associated with pond inoculation, harvesting, and disposal of biomass. As such, Mburu et al. [120] evaluated the capital expenditures (CAPEX) for a full-scale waste stabilization pond at € ±705,000 (based on 2700 person equivalents (PE) at 8.3 m^2 PE^{-1}) and at € ±276,000 (for 2700 PE at 3.4 m^2 PE^{-1}) for a pilot-scale horizontal subsurface flow constructed wetland. Maintenance costs for the first design are, however, significantly lower: € 283 compared to € 23,300 for 2700 PE. As such, the total cost of these "green" nutrient recovery systems can be evaluated at € 12–33 PE^{-1} $year^{-1}$, with an average of € 14.4 PE^{-1} $year^{-1}$, whereas the cost for a traditional activated sludge system is at least three times higher. However, without water reuse, associated gray-water sales, and duckweed valorization, currently the economic viability of duckweed systems remains questionable [114].

Furthermore, the ability of microalgae to assimilate excess nutrients from the environment has been thoroughly studied [121]. However, to date, tests of growing algae in waste(water) are mostly at laboratory scale. Pilot-scale algae cultivation continues to face many problematic issues, including contamination, inconsistent waste(water) components, and unstable biomass production. The major challenge associated with culturing algae in nutrient-rich natural water and slurry comes from the design of the cultivation system. The addition of polymer that precipitates suspended solids, thereby allowing light penetration, would improve the technical feasibility of growing algae on the liquid fraction of (digested) slurry. Nevertheless, Muylaert and Sanders [122] predict that breakthrough of algae in the bio-based economy will take another 5–15 years, as currently costs of algae production are too high as compared to other types of biomass. Estimates of the algal production cost range from € 3.2–240 (US $ 4300) kg^{-1} dry biomass [123–125]. Prior economic-engineering feasibility analyses have concluded that even the simplest open pond systems, including harvesting and algal biomass processing equipment, would cost at least € 78,000 (US $ 100,000) ha^{-1}, and possibly significantly more [91]. To this, the operating costs will need to be added.

The harvested algae/macrophytes can serve as a feedstock for the chemical and biofuel industry, can be used as animal feed (provided that the necessary amendments in legislation are made), or spread out as a fertilizer [126, 127]. As such, El-Shafai et al. [128] estimated a biomass value of € 5300 (US $ 6600) $year^{-1}$ (by comparison with other feed sources) for a protein yield of approximately 11 tons ha^{-1} $year^{-1}$ (*L. gibba*). Next to the large footprint required, biomass harvesting remains an important technical bottleneck, which requires further research.

Phosphorus Extraction from Ashes/Biochar (NRT 7)

The remaining ashes after combustion of biodegradable waste (manure, sludge, digestate) contain P-, K-, Al-, and silicium (Si) compounds and possibly heavy metals such as Cu, Zn, and cadmium (Cd). Several companies have designed different processes to extract P from such combustion ashes [45]. These processes can be subdivided into thermochemical and wet-chemical technologies. The Finnish company Outotec, for example, adds alkaline additives and heats the ashes to 1000 °C in order to gasify the heavy metals. Phosphorus is bound as $CaHPO_4$ and sold as chemical mineral fertilizer substitute. The Belgian company EcoPhos developed and tested, at lab-scale, a chemical P-extraction process by addition of hydrogen chloride (HCl) to combustion ashes [128]. Also the Swedish company EasyMining developed a process (*Cleanmap Technology*) that involves the use of HCl, which is suitable for ashes from incinerated manure. Other P recovery processes from ash are: *ICI Amfert* (NL), *Mephrec* (DE), *PAsH* (DE), *RecoPhos* project (Austria (AT), BE, DE, FR, Switzerland (CH)), *sephos* (DE), and *susAN/AsH DEC* (AT, DE, Finland (FI), NL). P recoveries up to 78 % are found in literature [45, 130]. Operational costs for wet extraction itself can be lower than € 1 m^{-3} fresh slurry, whereas net variable costs for combustion (including revenues from energy and P recovery) range from € 0 to 10 ton^{-1} FW slurry, depending on the water content. However, a thorough flue gas cleaning system is indispensable and post-processing to remove heavy metals is often required, hence few full-scale installations currently exist.

Experiments with pyrolysis of manure cakes have also been conducted. The fraction of nutrients recovered in the resulting biochar is larger than in incineration ashes and the plant-availability of the nutrients tends to be higher, especially for P. It was estimated that the value of P in biochar is about five times higher than the value of P in ash: <€ 1 m^{-3} versus € 4.25 m^{-3} [45].

Nevertheless, as digestate is classified as a waste stream that is eligible for recycling as soil conditioner, it is in a lot of countries not authorised to convert the product into energy by combustion/pyrolysis according to environmental legislations. Alternatively, P could be extracted from dried or dewatered digestate, but to date such tests are absent in literature. Some processes attempting to recover P from dried or dewatered sludge are: *lEACHPOs* (CH), *Mephrec* (DE), *PHOXNAN/10PROX* (DE), and *seaborne* (DE).

Critical Comparison

Based on the above compiled information, a critical comparative technology overview is given in Table 2.

At present, only struvite precipitation/crystallization, NH_3-stripping and absorption in a tower (with or without packing), acidic air scrubbing, and pressure-driven membrane filtration have been applied at full-scale for nutrient recovery from digestate. Of these technologies, only the first three have shown potential to be economically viable for implementation at digestate processing facilities.

Traditional membrane filtration systems often suffer technical problems in wastewater treatment, making them economically not yet viable for digestate treatment. An interesting solution may exist in vibrating membrane filtration (VSEP) using RO-membranes. However, further research is required in order to evaluate the technical and economic performance of this process [19]. Nevertheless, it should be pointed out that membrane filtration is the most established technology to date for the simultaneous recovery of both N and K.

Further, Table 2 shows that the NRT that currently achieves the highest simultaneous nutrient recovery efficiency of both N and P would be biomass production and harvest. However, the overall cost of this treatment is still high and large surface areas are required, making its potential implementation very region-specific. Further research to improve the economic and technical feasibility of this technology is recommended.

In terms of costs, NH_3-sorption and recovery of N-enriched (natural) zeolites is probably the lowest-cost option available to date (depending on the nearby availability of the adsorbent). However, recovery efficiencies are relatively low and further testing is required on the implementation of this technology for digestate treatment. Moreover, the marketing value of the recovered N-zeolites remains to be explored. The use of this technology in

Table 2 Technology overview: technology, mechanism, feed, % recovery, main technical bottlenecks, economic and environmental evaluation, stage of development, and references

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: € 2300–24,500 kg^{-1} P day^{-1} OPEX: € −520 to 2800 ton^{-1} struvite or € −200 to 75,000 $year^{-1}$ Overall: € −7800 to 89,400 $year^{-1}$	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^{-3}	Lab Pilot: test phase	[61]
	Bio-electrochemical	Liquid	20–40 % P	R&D needed	R&D needed	Energy efficiency ↑: 73 ± 4 % H_2 production	Lab	[62]
Calcium phosphate precipitation	Physicochemical	Liquid	50–100 % P	Co-precipitation of $CaCO_3$ Preceding CO_2 stripping often required	Can be profitable CAPEX: € 2300–2900 kg^{-1} P day^{-1} OPEX: depends on $Ca(OH)_2$ use	Chemical use ($Ca(OH)_2$) ↑ Fe/Al use ↓	Full-scale: manure and wastewater Lab: digestate	[63, 65] Technology providers
NH_3 stripping and 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^{-3}, both for 800 m^3 day^{-1} at 2.4 g N m^{-3} (90 % recovery) Overall: € 2.0–8.1 m^{-3} for 70 m^3 h^{-1} Depends on pH and temperature Can (partially) replace activated sludge system; Interest in S ↑	Odors ↓; Energy use (air/heat) ↑: 1.54–12 kWh_{el} m^{-3} and 62–69 kWh_{th} m^{-3}; Acid use ↑: 7–10 kg H_2SO_4 m^{-3}; Base use (↑): 0–6.5 kg NaOH m^{-3}, all for 800 m^{-3} day^{-1} at 2.4 g N m^{-3} (90 % recovery); Chemical use for cleaning ↑	Full-scale	[13, 32, 69] Technology providers
	Physicochemical: water-sparged aerocyclone	Liquid	>97 % NH_3	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_3	R&D needed Scale-up?!	Interest in S ↑ No air scrubber required	Energy use ↓: <1 kW Acid use ↑; Chemical use ↓	Pilot	[71]

Table 2 continued

Technology	Mechanism	Feed	% recovery	Main technical bottlenecks	Economic evaluation	Ecological evaluation	Stage of development	References
Acidic air scrubbing	Physicochemical	Gas	91–99 % N	Fouling/corrosion of packing material; Performance under freezing conditions?!	Can be profitable CAPEX: € 13 kg^{-1} NH_3 removal OPEX: € 6–7 kg^{-1} NH_3 removal Interest in S ↑	Odor ↓; Energy use (air) ↑: min. 0.057 kWh kg^{-1} NH_3 Acid use ↑: min. 1.5 L H_2SO_4 kg^{-1} NH_3	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^{-3}	Energy use ↑: 4–6 kWh m^{-3} (RO) Chemical use (cleaning) ↑	Full-scale	[36, 38, 77, 78, 80–83]
	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^{-3} Chemical use ↓: less cleaning	Full-scale: desalination, food processing; Full-scale digestate/manure: in 5–10 y	[83, 89, 90]
	Physicochemical: transmembrane chemosorption	Liquid	70–99 % NH_3	Membrane clogging	Depends on mass transfer Similar costs as stripping	Energy use ↓: <1 kWh m^{-3} Chemical use (acid) ↑	Pilot	[95–97]
	Electrochemical: electrodialysis	Liquid	80–83 % NH_3	High energy consumption; Variable costs for membranes, electrodes, cases Acid NH_3 trap required Post-treatment (RO/stripping) required	High costs (no data available)	Energy use ↑: 3.25–3.60 kWh kg^{-1} NH_4–N or 1.2–1.5 kWh kg^{-1} K; NH_3 volatilization	Full-scale: limited Lab: digestate/manure	[80, 91, 92, 94]
NH_3 sorption	Physicochemical	Liquid	18 % P 15–60 % NH_3	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, 39–41, 98]

Table 2 continued

Technology	Mechanism	Feed	% recovery	Main technical bottlenecks	Economic evaluation	Ecological evaluation	Stage of development	References
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^{-1}	Capex: >€ 80,000 ha^{-1} Overall (macrophytes): € 12–33 PE^{-1} $year^{-1}$ Overall (algae): € 4–300 kg^{-1} dry weight	Surface ↑: 3.4–8.3 m^2 PE^{-1} Energy use ↑ (CO_2 addition); Polymer use ↑	Pilot/Full-scale: duckweed Mostly lab: algae	[32, 117–119, 122–124]
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^{-3} slurry (wet extraction) € 3 ton^{-1} slurry (pyrolysis) € 0–10 m^{-3} 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]

↑ = relatively high; ↓ = relatively low; CAPEX = capital expenditures; OPEX = operational expenditures; PE = person equivalent; RO = reversed osmosis; R&D = research and development; R&D = research and development; ?! = questionable

combination with struvite or stripping (depending on the purpose) may be interesting, in order to maximize the overall nutrient recovery potential at minimal costs. Of the technologies applied to date at full-scale (see above), acidic air scrubbing is likely the most feasible technology, since air treatment is often required anyway for odor removal at processing facilities. While the investment costs of N stripping and absorption are relatively low, its operational expenditures (OPEX) are usually much higher than for P recovery through struvite precipitation. The main operational costs for struvite recovery are related to chemical dosing, while the main costs for stripping and air scrubbing are related to the air requirements. Optimization of these parameters is therefore of high interest. Technologies that are expected to become economically viable in the longer term (after sufficient research and optimization) are forward osmosis, transmembrane chemosorption (TMCS), and biomass production.

Further, from an ecological point of view (chemical and energy use), transmembrane chemosorption, forward osmosis (bio)electrochemical struvite recovery, and NH_3-sorption appear as the most interesting NRT's. However, none of these technologies is currently applied at full-scale for the treatment of digestate. Yet, after sufficient testing and optimization, these systems have the potential to become part of commonly used digestate processing technologies. The extraction of P from ashes or biochars seems the least promising technology, because it can be questionned whether combustion/pyrolysis of digestate is a sustainable treatment option and if this should be encouraged.

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

Product Quality and Markets

Classification of Recovered Products

A classification of products that can be recovered from digestate is provided in Table 3. Comparative information

Table 3 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	$MgNH_4PO_4 \cdot 6H_2O$	Pure struvite: 6 % N, 29 % P_2O_5, 10 % Mg Ostara: 5 % N, 28 % P_2O_5, 10 % Mg Crystalactor: 9 % N, 46 % P_2O_5, 16 % Mg; Crystals (0.5–5 mm); Slow-release; Purity?!	€ 45–1393 ton^{-1}	[12, 54–60] Technology providers
	Struvite precipitation (electrochemical)	Liquid fractions	$MgNH_4PO_4 \cdot 6H_2O$	>90 % purity; R&D needed	R&D needed	[61, 139]
	Struvite precipitation (bio-electrochemical)	Liquid fractions	$MgNH_4PO_4 \cdot 6H_2O$	R&D needed (40 % soluble P removal)	R&D needed	[62]
K/P-precipitates	Struvite precipitation (chemical)	Liquid fractions (acidified) raw digestate	$KMgPO_4 \cdot 6H_2O$	R&D needed	R&D needed	[49]
P-precipitates	Calcium phosphate precipitation	Liquid fractions (acidified) raw digestate	$Ca_5(PO_4)_3OH$ $CaHPO_4 \cdot 2H_2O$	P_{tot}: 10–11 %; Crystals; Purity?!	R&D needed	[63, 65]
P-extracts	P extraction	Ashes/biochar	Acid P-extract, $CaHPO_4$, struvite, Fe/Al-PO_4	P_2O_5: 15–35 % High P bio-availability Purity?!	€ 0.89–4.25 m^{-3}	[45, 129]
N/S-solutions	Stripping & absorption	(Decarbonated) liquid flows	$(NH_4)_2SO_4$-solution	AmS: 25–38 % N: 30–100 kg m^{-3} S: 61–100 kg m^{-3} pH: 3–7 High salt content: 30–150 mS cm^{-1}	€ 90–120 ton^{-1}	[12, 20–22, 34, 67] Technology providers
	Acidic air scrubbing	Strip gas, air	$(NH_4)_2SO_4$-solution	N_{tot}: 30–70 kg m^{-3}; S: 61–100 kg m^{-3} pH: 3–7 High salt content: 30–150 mS cm^{-1}	€ 90–120 ton^{-1}	[12, 20–22] Technology providers
	Transmembrane chemosorption	Tested on urine; Potential for liquid fractions of manure/digestate	$(NH_4)_2SO_4$-solution	Several 100 g NH_4^+ L^{-1}	R&D needed	[95]
N/K-concentrates	Reversed osmosis	Permeate from ultrafiltration, microfiltration or dissolved air flotation	N/K-concentrates	N_{tot}: 3–11 kg ton^{-1}: 92 % NH_4–N, 8 % organic N K_2O_{tot}: 5.0–13.6 kg ton^{-1} P_2O_{5tot}: 0–1.4 kg ton^{-1}; Purity?!	€ 1.19–1.25 ton^{-1}	[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× the input concentration	R&D needed	[80, 91, 92, 94]
N-zeolites	NH_3 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	€ 5300 year^{-1} for a protein yield of ±11 ton ha^{-1} year^{-1}	[113, 115, 125–127]

R&D = research and development; ?! = questionable on their fertilizer characteristics and marketing value is also presented.

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

In-depth product characterisation in time and long-term field trials aiming at the evaluation of the environmental impact of bio-based products are rare in literature, but highly important in the development of a market for recovered nutrients. Several researchers have investigated the fertilizer properties of struvite and the product has been evaluated (by means of greenhouse and field trials) as an eco-friendly (low leaching potential and environmental pollution) fertilizer for agricultural production [131–133]. However, the findings reported are mainly based on greenhouse studies, whereas long-term field trials using recovered struvite from digestate are limited [132]. Moreover, to reduce costs, these field trials are focussed on plant yield and P-uptake in particular, but do not investigate the mobility of (other) nutrients and heavy metals.

Next, the only reference found in literature on the beneficial value of recovered AmS fertilizer is the three-year field trial conducted by Ghent University in Belgium [21–23]. For membrane filtration concentrates, the only study that has been reported on to date is the pilot plant project in the Netherlands, in which the product has been applied during a 2-year field experiment [78, 79]. In order to establish the use of bio-based products in the agricultural community and to hasten the integration of these products in environmental and fertilizer legislations, more in-depth field trials focusing on the environmental impact of these products, next to their agronomic potential, should be conducted. Best management practices should also be established, see, e.g., Vaneeckhaute et al. [22]. All of this may also help to better estimate the economic value of these amendments compared to the conventional used chemical fertilizers. Indeed, to be economically profitable, the price allocated to the recovered nutrients should be in accordance to the market price of N, P, K, and S in mineral fertilizers.

Fertilizer Market Trends and Outlook

Of the total world-wide amount of N-fertilizers, only 4 % is AmS[2], mainly due to its relatively low N content as compared to that of for instance urea (21 and 45 % respectively). Recently, however, the world-wide supply of AmS has increased, in part due to the production of AmS by direct reaction crystallization from (spent) sulfuric acid and NH_3. This additional AmS-supply has been absorbed quickly in the marketplace, because of a general increase in fertilizer demand and an increased need for S-nutrition in particular. The latter is related to a general improvement of air quality world-wide, resulting in lower deposition of S on agricultural fields. Deficiency of S became a problem for more than 75 countries according to United Nations statistics [134] and supply of this nutrient could be efficient by using new (recovered) fertilizers containing sulfate [135]. The current additional production capacity of AmS from waste streams has not even been sufficient to fulfill the market requirements, however, and naturally, this gap in the supply–demand relationship has led to a rise in AmS-prices. As one might expect, the price of AmS varies with the various types of product quality available. The largest disparity is related to particle size, where prices of granular (2–3 mm) crystals are up to three times higher than these of <1 mm crystals. This price differential can be a strong incentive to produce large crystals. Hence, the trend of the market is toward the production of the so-called 'granular' AmS-quality, with a coarse fraction of 80 % >1.8 mm, which has a higher sales return compared to standard quality, but requires an improvement of the production process [136]. To date, recovered AmS from stripping and air scrubbing is most often marketed as a liquid fertilizer. The technical and economic feasibility of producing granular AmS from waste required further investigation.

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

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

Conclusions

Struvite precipitation/crystallization, NH_3-stripping and absorption, and acidic air scrubbing can be selected as best available technologies for nutrient recovery from digestate. These technologies have already been implemented at full-scale and have the ability to produce marketable end-products. Vibrating membrane filtration (VSEP) also shows potential to become part of the commonly used nutrient recovery technologies, but its technical and economic performance at full-scale remains to be demonstrated. All technologies require further technical fine-tuning in order to minimize operational costs, especially towards energy and chemical use, and to improve the quality and predictability of the produced fertilizers. To date, recovered bio-based fertilizers can be classified as renewable N/P-, K/P-, or P-precipitates, P-extracts, N/S-solutions, N/K-concentrates, N-zeolites, and biomass. Future research should further explore, verify, and improve the fertilizer characteristics and marketing value of these products towards industrial and agricultural end-users.

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