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Technologies to Recover Nutrients from Waste Streams: A Critical Review

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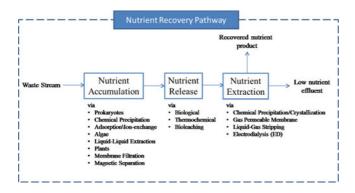
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Technologies to recover nitrogen, phosphorus, and potassium from waste streams have undergone accelerated development in the past decade, predominantly due to a surge in fertilizer prices and stringent discharge limits on these nutrients. This review provides a critical state of art review of appropriate technologies which identifies research gaps, evaluates current and future potential for application of the respective technologies, and outlines paths and barriers for adoption of the nutrient recovery technologies. The different technologies can be broadly divided into the sequential categories of nutrient accumulation, followed by nutrient release, followed by nutrient extraction. Nutrient accumulation can be achieved via plants, microorganisms (algae and prokaryotic), and physicochemical mechanisms including chemical precipitation, membrane separation, sorption, and binding with magnetic particles. Nutrient release can occur by biochemical (anaerobic digestion and bioleaching) and thermochemical treatment. Nutrient extraction can occur via crystallization, gas-permeable membranes, liquid-gas stripping, and electrodialysis. These technologies were analyzed with respect to waste stream type, the product being recovered, and relative maturity. Recovery of nutrients in a concentrated form (e.g., the inorganic precipitate struvite) is seen as desirable because it would allow a wider range of options for eventual reuse with reduced pathogen risk and improved ease of transportation. Overall, there is a need to further develop technologies for nitrogen and potassium recovery and to integrate

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accumulation—release—extraction technologies to improve nutrient recovery efficiency. There is a need to apply, demonstrate, and prove the more recent and innovative technologies to move these beyond their current infancy. Lastly, there is a need to investigate and develop agriculture application of the recovered nutrient products. These advancements will reduce waterway and air pollution by redirecting nutrients from waste into recovered nutrient products that provides a long-term sustainable supply of nutrients and helps buffer nutrient price rises in the future.

Graphical Abstract:



KEY WORDS: nitrogen, nutrient recovery technologies, phosphorus, potassium

1. INTRODUCTION

Nitrogen (N), phosphorus (P), and potassium (K) are critical to intensive agriculture and there are concerns over long-term availability and cost of extraction of these nutrients, particularly with P and K which are predominantly sourced from mineral deposits. The main source of P, phosphate rock, is nonrenewable and is becoming progressively limited with supply uncertainty being reflected in recent price rises. It has been estimated that by 2033 the worldwide demand will progressively outstrip supply, because supply will continue to increase with a growing global population, but the rate of production of phosphorus fertilizer will be in decline when readily accessible phosphorus resources become depleted. In addition, nearly 90% of the world's estimated phosphate rock reserves is found in just five countries, Morocco, Iraq, China, Algeria, and Syria, which may be considered a food security issue for other nations. While N is a renewable resource, the

process by which N (as ammonia) is industrially synthesized (Haber–Bosch process) is energetically intensive, with its cost dependent on the price and supply of natural gas.⁴ Potassium-based fertilizer prices have increased by as much as four times during the period 2007–2009 and there are issues around supply of K-based fertilizers to developing nations.⁵ This is because potash ores (the main source of K) have a limited distribution globally, with the bulk of the world's potash mined in Canada and Europe.³ Thus, there is currently very little scope for many developing countries to be self-sufficient with respect to supply of K via conventional fertilizers. Demand for food for an ever increasing global population and ongoing developments to create energy from biomass (which provide concentrated nutrient side streams) will drive demand for nutrients from alternative sources upwards into the future.

The use of inorganic or synthetic nutrient fertilizers is ubiquitous in modern agriculture, predominantly due to ease of application and lack of organic substitutes. Nearly 90% of the phosphate rock mined worldwide is used for fertilizers⁶ typically in combination with N and K. Typically, crops have limited nutrient uptake efficiency, which is around 40% for N and 45% for P.7 Some of these remaining nutrients are stored in the soil deposits but substantial proportions, particularly of mobile nutrients such as N and K, flow into the environment as atmospheric and aquatic pollutants. Humans and animals consume nutrients from crops and produce nutrient-rich waste streams from processing food. It is estimated globally that the total P content in excreted human waste (urine and feces) can meet approximately 22% of the demand for P.8 Human waste is not generally recycled and is often either discharged (with or without treatment) to waterways or stored in landfills. Animal-derived waste, particularly manure, is widely used as a fertilizer. But the value of these nutrient sources is commonly low or negative (<\$10 tonne⁻¹) because of bulk (moisture content) and low nutrient concentrations. Moreover, the use of this waste as a fertilizer is often complicated by the presence of heavy metals (e.g., Zn and Cu⁹), pathogenic microorganisms, and odor.

Due to limited recycling and inefficient nutrient management, these nutrients are major contributors to the environmental impact of domestic, agricultural, and industrial waste streams. Methane and nitrous oxide, major contributors of greenhouse gases, are generated in large amounts by manure management (stockpiling and treatment) and excess use of N-based fertilizers. Overall, agricultural activities and livestock production are estimated to be responsible for 30–32% of global anthropogenic emissions of greenhouse gases. ^{10,11} Also, there is strong concern of excess nutrients in waterways causing eutrophication. Agricultural runoff of nutrients is associated with oxygen depletion in coastal regions caused by decomposition of dead algal biomass. ¹² Along with environmental impact, eutrophication can have major economic impacts by damaging valuable marine fisheries and impairing water bodies used for potable water supply and recreation. ¹²

Currently, the general objective of waste treatment facilities is to produce an acceptable quality of water for either reuse or discharge. This approach is driven by human health and the minimizing of environmental impact. Most facilities manage carbon and nutrients as wastes to be removed, and are yet to capitalize on nutrients as a substantial resource in waste streams. Removal of nutrients from wastes has largely focused on exploiting nutrient cycling reactions whereby reactive forms of nutrients are converted to unreactive forms (e.g., ammonia to nitrogen gas). Sequestration of nutrients into a form that is not readily bioavailable (such as with strong binding of P to coprecipitated iron) is also commonly used for removing of P from wastes. ¹³ Recycling nutrients through sustainable methods (rather than destruction or emission) is emerging not only for sustainability reasons but also due to economic drivers based around the supply-demand issues outlined above. In the past decade, there have been considerable efforts to improve, demonstrate, and integrate nutrient recovery technologies with existing treatment infrastructure. 14,15 These technologies are specific to a nutrient type, a nutrient form, or a type of waste stream. There have been good reviews on particular relevant technologies, but these have mainly focused on specific technologies or applications, and have generally had a strong focus on phosphorous recovery. 16-22 There is a need for a detailed review of the nutrient recovery field as a whole to outline a holistic and integrated approach to nutrient recovery. This is the motivation for the current review which broadly reviews available nutrient recovery technologies to better understand the opportunities and barriers for widespread adoption and to identify key needs for further targeted research and development. This review focusses on N, P, and K recovery from waste streams, and metal, water, or energy recovery are only considered in terms of how they influence the viability of the nutrient recovery technologies.

2. FRAMEWORK FOR IMPLEMENTING NUTRIENT RECOVERY

Nutrient concentrations in waste streams are relatively low (1–200 mg L⁻¹) when compared with synthetic inorganic fertilizers, and the majority of the available nutrient resources are found in the most dilute waste streams.^{23–25} Since the efficiency of nutrient recovery typically decreases with nutrient concentration in the waste, a three-step framework to nutrient recovery is being proposed here in order to achieve the best overall outcomes. These are the steps (given in sequential order) of: (1) nutrient accumulation, (2) nutrient release, and (3) nutrient extraction. That is, the nutrients in dilute waste streams need to be accumulated in order that subsequent release techniques can mobilize the nutrients for final recovery in the form of concentrated products for beneficial reuse. The advantage of employing this

multistep approach is that each step can be operated and optimized independently. In this review, available technologies are classified into one of these three steps (accumulation, release, and extraction). The reason for this classification, rather than an application-specific focus (e.g., P recovery from manure²¹), is that the different technologies can be assessed from a general adoption perspective rather than an application-specific perspective. The review evaluates the available technologies based on a multicriteria analysis.

3. NUTRIENT ACCUMULATION TECHNOLOGIES

Nutrient accumulation technologies recover soluble nutrients (N, P, and K) from waste streams with low nutrient concentrations (2–20 mg L⁻¹). As nutrient discharge limits can require effluents to contain less than 0.1 mg P L⁻¹ and 1–3 mg N L⁻¹, accumulation options must ideally be capable of sequestering most of the soluble nutrients in order to produce treated effluents that meet these stringent discharge limits.^{26,27} Biological, physical, and chemical techniques can be used for nutrient accumulation. Chemical accumulation techniques have been largely limited to P, whereas biological methods can also be used for accumulation of N and K. Physical accumulation via adsorbents can be used for all three nutrients.

3.1. Prokaryotic Accumulation

Both phototrophic and heterotrophic phototrophic organisms can be potentially used for accumulation of nutrients. Common nutrient accumulating microbes are proteobacteria such as polyphosphate-accumulating organisms (PAOs) and purple nonsulfur bacteria²⁸ and cyanobacteria.²⁹ PAOs are currently extensively used for phosphorous removal and can accumulate up to 20-30% of P by weight³⁰ with solids-retention of less than 10 days,³¹ storing the P in a compound called polyphosphate. Bacterial-accumulation of P through enhanced biological phosphorus removal (EBPR) is widely used in sewage treatment plants to remove 80-90% of soluble P from the effluent. EBPR requires alternating anaerobic and aerobic/anoxic conditions so that the uptake of P by microorganisms is above normal metabolic requirements. 13 The optimum aerobic P uptake occurs at pH 7–8.32 Also wastewater should contain carbon to P ratios of 5 or higher to enhance accumulation of P,³³ with volatile fatty acids being the most effective form of carbon. Pre-fermentation of wastewater to produce volatile fatty acids (VFAs) is often beneficial and sometimes essential for EBPR.34 EBPR removal has also demonstrated >90% P removal from various types of industrial wastewaters.³⁴ Phosphate-rich sludge with PAOs can be separated from the wastewater by settling, and nutrients can then be released and recovered from the settled sludge by the methods outlined in Sections 4 and 5.

Purple nonsulfur bacteria and cyanobacteria can grow with and without light, and consume water, carbon dioxide or oxidized substrate, and nutrients to produce organic matter and oxygen. They have a variety of characteristics that make them well-suited to wastewater treatment to assimilate and accumulate nutrients, and store the nutrients as proteins or polyphosphate. Purple nonsulfur bacteria can be used to treat many kinds of wastewater to produce a smaller quantity (less bulk) but highly nutrient-rich biomass when compared to activated sludge processes. ²⁸ Cyanobacteria such as blue–green algae are suitable for luxury uptake of N. The protein concentration reported for cyanobacteria is up to 80% of the dry weight, and consists of 8-12% N and 1% P.25,35 The nutrient content and removal rate of cyanobacteria depends on the amount, the availability, and the type of the nutrient source.²⁵ Purple nonsulfur bacteria have a high tolerance to heavy metal exposure, but unfortunately accumulate heavy metals along with nutrients from the wastewater.³⁶ The technology may be particularly promising for N recovery and should be considered a high priority for future research.

3.2. Chemical Accumulation via Precipitation

Chemical accumulation of nutrients can be accomplished via coagulation and flocculation, where soluble nutrients and nutrients bound to colloids $(0.01-1 \mu m)$ are precipitated as solids and separated by settling in clarifiers. Aluminum- or iron-based coagulants are commonly used for accumulating of P from dilute wastewater. Other coagulants such as calcium, natural, and synthetic organic polymers, and prehydrolyzed metal salts such as polyaluminum chloride and polyiron chloride¹³ are also used, but generally have a relatively high cost. Metal ions can also be delivered through sacrificial iron or aluminum anode electrodes through electrocoagulation.³⁷ The coagulants, when added to water, hydrolyze rapidly and form multicharged polynuclear complexes with enhanced adsorption characteristics. The efficiency of rapid mixing, the pH, and the coagulant dosage determine which of the hydrolyzed species is effective for treatment.³⁸ Once suspended particles have flocculated into larger particles (sludge) they can usually be removed from the treated water by sedimentation, provided that a sufficient density difference exists between the sludge and the treated water.

The optimum pH is dependent on the type of coagulant used; however, due to the heavy use of biological processes in sewage treatment plants, operation over the pH range of 6.0–8.0 is typical. As this process is effective for removing soluble and particulate P, it is heavily used as part of a multipoint dosing process for controlling P discharge from sewage treatment plants. Along with nutrient removal, the chemical coagulant can also remove organic matter, pathogens, viruses, and other inorganic species such as arsenic and fluoride. Other advantages are ease of operation, flexibility to changing conditions, and low capital cost to reduce effluent P concentration

to less than 1 mg L⁻¹.³⁹ Disadvantages associated with chemical accumulation by precipitation include high operating costs, increased salinity in the effluent (mainly as chloride or sulfate), increased sludge production (up to 35 vol%),³⁹ the addition of heavy metals present in the raw coagulant,⁴⁰ and inhibitory effects on the biological process such as anaerobic digestion following the coagulation process.⁴¹ It should be acknowledged that the sludge produced from chemical accumulation techniques, particularly with aluminum and iron coagulation, is agronomically less useful due to low bioavailability of the strongly bound P.⁴² Consequently, if this accumulation technique is to be applied as part of an overall nutrient recovery strategy, a subsequent release step can be essential to improve bioavailability of the bound nutrients.

3.3. Adsorption/Ion-Exchange

During adsorption and ion-exchange, ions are transferred from the solvent to charged surfaces of insoluble, rigid sorbents suspended in a vessel or packed in a column. The sorbents are made from porous materials containing interconnected cavities with a high internal surface area. A selective preference of an exchange media for a particular ion in aqueous solution (such as phosphate) is based on surface valence (e.g., a higher valence media has a better selectivity for phosphate), diffusivity of the ion, and physical properties of the sorbents such as functional groups and pore size distribution. Adsorption and ion-exchange can accumulate soluble N, P, or K from waste streams. Spent sorbents are regenerated using low-cost, high concentration aqueous solutions of cations or anions such as sodium, sulfate, or chloride. The principle design parameter is bed volumes to breakthrough/the amount of waste stream that a given sorbent can treat (kL per kL).

Adsorption and ion-exchange technology is suitable for waste streams with a range of nutrient concentrations (1–2000 mg L $^{-1}$), but relatively low solids concentrations (<2000 mg L $^{-1}$). For low strength waste streams such as effluent from sewage treatment plants and artificial lakes where nutrient concentrations are less than 5 mg L $^{-1}$, advanced engineered polymeric sorbents are employed. Such sorbents can reduce P load to 50–100 μ g L $^{-1}$. Waste streams with an acidic pH (<8.0) are preferred to improve nutrient solubility and maximize adsorption on the resin. For concentrated waste streams (>2000 mg L $^{-1}$), typically, red mud, metal oxide/hydroxide, and zirconium sorbents are used for P recovery and modified zeolite and clinoptilolite for N and K recovery. Maximum loading capacities have been reported to be 57 g P kg $^{-1}$ for zirconium-loaded orange waste gel, 43 and 21.5 g N kg $^{-1}$ for clinoptilolite. 44

The potential advantages of this technology are the ability to achieve high P accumulation and low P concentrations in the treated effluent of <0.1 mg PO₄–P L⁻¹, even with high-strength waste streams.⁴⁵ No additional

sludge (other than spent media) is produced and the pH of the waste streams remains unaffected. Chemicals required for the regeneration of the sorbents, biofouling, large amounts of resin required for complete removal, limited resin life, and competitive foreign ion adsorption are some of the challenges for full-scale implementation. To reduce regeneration costs, some studies have tried to use biology rather than chemicals to regenerate the media, i.e., bioregeneration. 46,47

Adsorption/ion-exchange can be categorized as a hybrid nutrient accumulation-nutrient recovery technique because the nutrient-laden sorbent/exchange media can potentially be directly applied as a nutrient product in agriculture. 48

3.4. Algae Accumulation

Algae are unicellular or multicellular, autotrophic, photosynthetic eukaryotes. Algae have received significant attention worldwide as a valuable source of biomass for energy because of their high growth rates as compared to terrestrial plants⁴⁹ and their ability to capture large quantities of atmospheric carbon dioxide. These organisms can also be used to accumulate nutrients, as they require less than one-tenth of the area to recover P compared to terrestrial crops and pastures.⁵⁰ Nutrient accumulation is dependent on algal physiology, predominant forms, concentration of nutrients (N and P), light intensity, pH, and temperature. Reports have suggested that the nutrient content of algal dry biomass could reach up to 2% N and 3.3% P.^{51,52}

Algae-based systems can be suspended or nonsuspended. In nonsuspended systems, the algae are immobilized on a resin. The surface-immobilized algae reduce nutrient load in the waste streams via adsorption and/or precipitation on the surface of the material as well as through nutrient uptake by the biomass. Nonsuspended systems have been successfully tested in high-nutrient agriculture streams such as dairy, poultry, and swine manure waste. Suspended algae configurations are used in facultative and high rate algal ponds. In unmixed facultative ponds, the residence time can range from 20 to 100 days; whereas in the high rate shallow ponds, residence times can range between 4 and 10 days while gentle mixing is provided with paddle mixers. In a recent review, a tubular photo-bioreactor with suspended algae was found to be the most promising option for producing algal biomass in full-scale applications.

Optimal pH for growth of algae is in the range of 7.5–8.5 with an optimal temperature between 15 and 30 $^{\circ}$ C, 55 with lower temperatures resulting in decreased growth. As carbon dioxide (CO₂) is consumed by algae during photosynthesis, the pH of the waste stream can increase which can encourage further minerals precipitation of P (such as with calcium or magnesium) and volatilization of N as ammonia. 57

Floating algal farming is an emerging nutrient removal/accumulation process from waste streams. This approach may be most appropriate in coastal regions where nutrients are discharged directly to ocean from agriculture activities⁵⁸ and thus can be recovered. The salinity gradient between waste streams and seawater has the potential to drive osmosis and help concentrate nutrients and dewater harvested algae. To date, environmental and technical issues have limited implementation of ocean-based systems, but economics of ocean-based systems can be relatively competitive in view of the significant land cost associated with onshore algal cultivation.

The nutrient-rich algae can be processed with nutrient release techniques such as anaerobic digestion or thermochemical methods (described in Section 4), or may be used directly as an animal feed or a fertilizer. Due to the small particle size of algal cells and their typical growth as small colonies or single cells, harvesting of algal biomass has been considered to be a major challenge for full-scale nutrient accumulation. Future economic evaluation of algal systems should consider multiple benefits, including nutrient recovery, carbon sequestration, biofuel, and high-value by-products. It is likely that the economics of application will only be positive in scenarios where all the benefits are maximized.

3.5. Liquid-Liquid Extraction

Liquid–liquid extraction is a method of separating compounds based on relative solubility in two immiscible liquids, and can be used to recover soluble nutrients (phosphate or nitrate/nitrite species) from dilute waste streams. In this process, an extractant is dissolved in an organic phase. This organic phase with extractant is brought into contact with the waste streams which causes a transfer of nutrients into the organic phase until equilibrium is reached with the aqueous (wastewater) phase. The organic phase laden with nutrients is then brought into contact with another secondary aqueous phase at conditions where the nutrients are highly soluble in the secondary aqueous phase. This contact causes nutrient transport from the organic phase into the secondary aqueous phase. The organic phase, then stripped of nutrients, can be recycled for contact with more of the waste stream to extract additional nutrients. Figure 1 provides a schematic overview of this process.

Higher nutrient concentrations and lower solids content in the waste stream can improve the efficiency and economics of the liquid–liquid extraction because nutrient extraction will be favored and extractant loss will be minimized. The number of extraction and stripping stages can also vary depending on the degree of accumulation/separation required. The diluent and extractant can be recycled but makeup solution is required to maintain process efficiency. A recent study found that a mixture of kerosene (organic phase) and benzyldimethylamine (extractant) in a 2:1 ratio worked best for

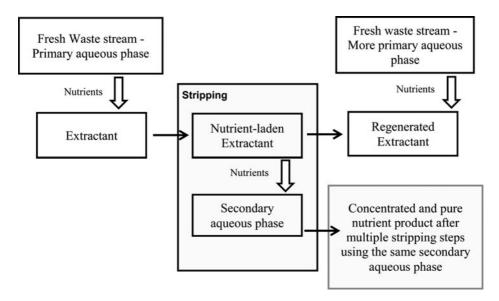


FIGURE 1. A schematic representation of a liquid–liquid extraction process.

phosphate extraction, and that combined use with 6.0 M sulfuric acid as the secondary aqueous phase provided a high P recovery of >93%. 61 In this case, the secondary phase would become a phosphoric acid with a considerably higher phosphorus concentration than the original waste stream. It was observed that the organic mixture could be recycled up to five times and still achieve a reduction in the P concentration of treated water from an initial 21 mg L^{-1} down to below 5 mg L^{-1} . 61

Significant advantages for this technology are the simultaneous accumulation and recovery of nutrients from the waste stream in a single process, the avoidance of waste generation, and the low nutrient concentration levels (${\rm PO_4}^{3-} < 1~{\rm mg~L^{-1}}$) that are achievable in the treated water. ⁶¹ The cost of the chemicals used and carry-over of organic phase into the nutrient products are major hurdles for adoption. Liquid–liquid extraction can be considered a hybrid nutrient accumulation and extraction technique since the concentrated liquid product may potentially be directly applied as a fertilizer in agriculture.

3.6. Plant Accumulation

Nutrient accumulation can also be performed using wetlands. In this system, nutrients accumulate as plants grow on the water surface, creating anaerobic conditions in the surrounding water. The anaerobic conditions drive digestion reactions where organic matter is metabolized to produce nutrients that can then be further accumulated by the plants. These plants, however, must be routinely harvested to ensure that the accumulated nutrients are

not recycled. The wetlands can be subsurface or surface flow type and the plants can be of the submerged, emergent, floating leaved, or free-floating type. ⁶³ Free-floating plants have a higher capacity for nutrient accumulation as they grow on the surface of the water and the roots are kept suspended in the water column to allow accumulation of the nutrients rather than being rooted in the sediments. There are three plants which are currently being examined, on different scales, for the various phases of treating waste streams and recovering nutrients: water hyacinths (*Eichhornia crassipes*), duckweeds (*Lemna minor*, *Landoltia Punctata*, and *Spirodela polyrrhiza*), and various emergent macrophytes. ⁵⁰

Nutrient removal efficiency for plant accumulation varies with each plant type, each characteristic waste stream, environmental factors such as temperature and dissolved oxygen, and type of wetland. The minimum water temperature is typically 7 °C, with optimum temperatures ranging between 25 and 31 °C.64 The optimum pH is 6.0–8.0.64 Both water hyacinths and duckweed can tolerate high-nutrient loads and have a high-nutrient removal capacity (N and P removal >70%)⁶⁵ with N and P accumulation in the range of 1-3%.50 Wetlands are potentially a low-cost option for nutrient recovery with the additional benefit of reducing organic matter from waste streams. Disadvantages include a large footprint and the regular harvesting that is required. The area required by plants to recover nutrients is dependent on nutrient content and areal biomass productivity. Biomass yields (tonne ha⁻¹ year⁻¹) for water hyacinths and duckweed are reported to be as much as 10 times higher than that of terrestrial crops, and require a 100 times smaller footprint while accumulating more P (10 times more) than terrestrial crops (switch-grass and maize).⁵⁰ The plants can be used as animal feed (which directly recycles the nutrients), as a fertilizer, or can be processed through an appropriate nutrient release technology outlined below. 66 Further research is required in plant biotechnology to improve nutrient uptake while minimizing biomass yields and footprint, so that it is more comparable with other biologically based nutrient accumulation systems.

3.7. Membrane Filtration

Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reserve osmosis (RO) are all membrane processes which selectively separate constituents from waste streams, without phase transformation, based on size and reactivity to water, and using semipermeable membranes and differential pressure. Nutrients in particulate form $>0.1~\mu m$ in size (suitable for MF or UF) or in soluble form (suitable for NF or RO)¹⁷ can be selectively removed. The membrane module configurations can be hollow fiber, flat sheet, tubular, or spiral wound.¹⁷ The filtration system can be in a submerged configuration or a pressure vessel configuration (side stream).

Membrane filtration produces a concentrated effluent (N, P, and K) from waste streams and has recently gained importance particularly in manure treatment. The waste stream volumes can be reduced by 4–6 times (concentrate with nutrients is 25–16% of the original volume), while retaining all nutrients and may be suitable for irrigation or subsequent recovery processes. The retention of ammonium and nitrate by NF and RO membranes is >80% and it improves with reduction in pH. 17,67–69 Disadvantages are mainly the high energy costs involved in membrane filtration as well as accumulation of unwanted contaminants and salts, which generally render concentrate unsuitable for direct reuse. Membrane processes are typically operated in a pH range of 6.0–8.0 to reduce inorganic scale formation on the membranes and to maximize nutrient retention. The process requires extensive pretreatment of waste streams to prevent fouling, to maximize membrane life, and to increase membrane flux rates.

3.8. Magnetic Separation

In this approach, soluble nutrients are accumulated from the waste stream by employing adsorption to a carrier material that has magnetic properties (e.g., magnetite, zirconium ferrate, carbonyl iron, and iron oxide). Once sequestered from solution, the nutrients-laden carrier material can be recovered by capturing the magnetic particles with a magnetic field in high gradient magnetic separators (HGMS).^{70,71} The HGMS rely on an electrically generated magnetic field with the electrical wires running parallel to the flow of the suspension carrying the magnetic particles (i.e., magnetic field is perpendicular to the flow field). The nutrients must be adhered to the magnetic particles with sufficient strength to prevent rerelease by hydrodynamic forces acting on the magnetic particles.

The magnetic carrier can be regenerated via chemical release techniques (next section).⁷² This process can simultaneously recover soluble N, P, or K from waste streams using specific adsorbents (refer to Section 3.3) bound to the magnetic carrier. The sequestered nutrients could also be strongly coagulated or precipitated with the magnetic particles. In these ways negatively or positively charged nutrients or uncharged organic nutrient compounds can be sequestered from the original waste stream by binding with the magnetic particles. The process has been tested at full scale to recover P from a sewage treatment plant.⁷² The process had a high recovery of >90% within 1 hr and with effluent P concentrations of <0.5 mg L⁻¹.^{71–74} Notably, this process does not interfere with the biological process and hence can be integrated at any stage of an advanced resource recovery train. The magnetic carriers commonly used are magnetite, ^{72,74} zirconium ferrate, ⁷⁰ carbonyl iron, ⁷¹ and iron oxide. ⁷¹ Perceived advantages of this process are high elimination performance, potentially a small process footprint and low power input per unit

of nutrient recovered.⁷² There is a shortage of published literature on this technology and additional information is needed to fill the knowledge gap.

4. NUTRIENT RELEASE TECHNOLOGIES

Once accumulated, nutrients must be either released or directly extracted into a recovered product. Both biological and chemical release methods can be used. Thermal techniques, discussed separately in this section, are commonly used in conjunction with chemical techniques for complete nutrient release into a soluble form. Animal waste (manure) and biological streams (activated sludge) naturally contain a large quantity of nutrients, but at low concentration with high moisture content and bulk carbon, and are often contaminated or unstable to use directly in land application. The extraction of nutrients from such streams is the focus of this section.

4.1. Biological Release

Anaerobic digestion is the most commonly used process for stabilization of wastes, organic solids destruction, pathogen destruction, and energy recovery from wastes in the form of biomethane.⁷⁵ The digestion process also facilitates the release of nutrients from the biodegradable fraction of the waste. In this process, organic N is converted into ammonium and organic P is hydrolyzed to soluble P with the extent of conversion dependent on the conditions employed during digestion. Anaerobic digester designs vary widely, but for agricultural and high-solids processes, can be divided according to their feed characteristics as 76: largely soluble or low solids (<1% solids; lagoons, high-rate anaerobic processes, anaerobic membrane processes), slurry-based (1-6% solids; complete mix), and high-solids type (>6-10%; plug flow, leach bed). The optimum operating temperature for anaerobic digestion is 35-40 °C for mesophilic bacteria and 55-60 °C for thermophilic bacteria. The optimum pH is in the range of 6.5-7.5. The residence time of the process varies with the substrate and is typically in the range of 20-30 days.

Released nutrients are soluble and tend to form inorganic compounds or adsorb onto solid surfaces in the digestate. Studies on manure showed that most of the organic P is released, however less than 10% of this P remained soluble following digestion.^{77,78} Typically, the soluble P content in most municipally digested wastes range from 50 to 500 mg L⁻¹ and N is often five times higher than soluble P.⁷⁶ To reduce solids handling costs, the digested solids are typically dewatered to produce a soluble nutrient-rich (predominantly N and K) side stream. This nutrient-rich side stream can be a feedstock for nutrient extraction/recovery techniques. The remaining particulate-bound nutrients and residual organics are recovered as biosolids,

which have value as nutrient amendments for agricultural purposes, provided that a suitable biosolids quality can be achieved to match specific application requirements with respect to residual odor, pathogens, and heavy metals.⁷⁹

Processing that selectively enhances solubilization of nutrients can be used to channel a larger portion of nutrients into the valuable nutrient-rich product stream, rather than to the lower-value biosolids. This processing may include the addition of complexing agents, such as ethylenediaminete-tra acetic acid (EDTA), operation at depressed pH, or otherwise modifying operating conditions to reduce the quantity of nutrients being sequestered with the biosolids/sludge.⁷⁷ The so-called waste activated sludge (WAS) enhanced release process⁸⁰ is an example of a process that improves P release from PAOs in WAS, prior to digestion with a short incubation time in the presence of volatile fatty acids. The WAS enhanced phosphorus release process generates a P-rich load that is ammonia-limited, which in turn can be combined with the ammonia-rich (but P-limited) digestate from a conventional anaerobic digestion process to facilitate controlled struvite formation. In this way maintenance issues associated with struvite scale formation in pipes and process infrastructure, can be minimized.

4.2. Thermochemical Stabilization and Chemical Release

Thermochemical processes like thermal hydrolysis, wet oxidation, incineration, gasification, and pyrolysis can greatly reduce the bulk volume of wastes by destroying a large proportion of the carbon, and in the case of incineration, gasification, and pyrolysis, by evaporating off moisture. The processed waste can then be more readily transported and can be further processed by other chemical release technologies to value add to nutrient products. The char/ash/oil that is produced from the thermochemical processes retains most P and K, but N is lost in the gas stream. Wet oxidation is carried out at moderate temperatures (180-315 °C), and at high pressures of 2-15 MPa. 81 Metals are oxidized to their highest valency and P to P₂O₅. 82 The degree of oxidation depends on the temperature and pressure selected, and the quantity of oxygen supplied. Incineration and gasification occurs in the presence of excess oxygen above 800 °C, while pyrolysis operates under a limited supply of oxygen and at relatively low temperatures (<700 °C). Pyrolysis can be designed and operated in such a way as to retain most of the P and K and some of the N in the solid or liquid by-product. For example, pyrolysis of sewage sludge retained 100% of P and K and 55% of N in char. 83

The solid by-products (ash/char) from thermochemical treatment can be further processed thermally in the presence of chloride salts, which convert heavy metals into heavy metal chlorides to be vaporized and removed from the char/ash.⁸⁴ Heavy metals are then captured through flue gas treatment. As a major disadvantage, such processing loses a large proportion of K from the char/ash.⁸⁵ Additionally, incineration ash can only be used if combusted

at low temperatures (<700 °C) to ensure a high fertilizer efficiency of P in ash, ⁸⁶ which is incompatible with minimizing nitrous oxide emissions which requires combustion at >900 °C. ⁸⁷ So, greenhouse gas emissions and fertilizer efficiency are competing factors.

Chemical extraction involves the addition of acids or bases to char, digester reject, solid waste, or waste streams, at moderate temperatures ($<200~^\circ$ C) and/or pressures to release nutrients into a leachate. The chemical extractants typically used are inorganic acids (H_2SO_4 , HCl, HNO₃), organic acids (citric and oxalic acids), inorganic chemicals (e.g., ferric chloride solution), and chelating agents (e.g., EDTA). Unfortunately, undesired compounds, such as heavy metals, are also released into the leachate. Additional processes are often required to extract and recover nutrients from the leachate. There are a number of commercial processes which couple thermochemical stabilization with chemical extraction as shown in Table 1. The major differences between these technologies are the specific extraction chemicals being used, operating pressures and temperatures and the processed feedstock (that is, sludge or char).

The Seaborne, Sesal-Phos, Biocon, Sephos, Pasch, Stuttgarter Verfahren, and Loprox/Phoxnan processes dissolve nutrients and heavy metals using acids at a pH below 3, while the Kreprco and Aquareci processes are operated at high temperatures (>100 °C) and pressures (>5 bar) for nutrient dissolution.⁸⁹ The dissolved ions (nutrients and heavy metals) are subsequently separated by crystallization (Seaborne, Stuttgarter Verfahren, Sephos, Sesal-Phos, and Krepco), membranes (Loprox/Phoxnan), solvent extraction (Pasch), or ion-exchange (Biocon). In these processes, various P-based inorganic compounds are produced, which must be rigorously tested for heavy metal contamination before they can be applied in agriculture. The main challenges to implementing these technologies are the relatively high operating costs (including chemical costs) and high capital costs which limit application to very large commercial installations (centralized processing facilities). The potential need to remove heavy metals from products can also increase the costs associated with implementing these extraction technologies.

4.3. Bioleaching/Extraction

Bioleaching is a release technology that relies on the solubilization of nutrients and heavy metals from solid substrates either directly by the metabolism of leaching microorganisms or indirectly by the products of metabolism. Microorganisms with potential for bioleaching activity include mesophiles such as *Acidithiobacillus thiooxidans* and *Acidithiobacillus ferrooxidans*; thermophiles such as *Sulfobacillus thermosulfidoxidans*; and heterotrophic microbes such as *Acetobacter*, *Acidophilum*, *Fusarium*, *Penicillium*, and *Aspergillus*. ¹⁹ These microorganisms have the unique ability to survive in highly

TABLE 1. Nutrient recovery technology summary

		Nutrients		Level of		Products (wt%	
	Class	(recovery efficiency)	Operating conditions	pretreatment required	Inputs	nutrient by dry mass)	Commercial processes
Embryonic technology Cyanobacteria	Accumulation	d Z	5-40 °C, pH 6.5-8,	Low	Carbon source,	Biomass (8–12% N	l
•			0.5 hr HRT		light	and 1% P)	
Purple nonsulfur bacteria	Accumulation	Х, Р	27–34°C, pH 6–8, 4–7 davs HRT	Low	Carbon source, light	P containing alginate beads	Batelle
Algae	Accumulation	И, Р	15–30 °C, pH	Low	UV light source	Sludge (1–3.2% P	Lemna
			7.5–8.5, 3–15 days HRT			and 1–10% N)	Technologies
Electrodialysis	Extraction/ recovery	N, P, and K	10–40 °C, pH < 8.0, <1 hr HRT	Solid-liquid separation	Electricity	Concentrated nutrient solution	GE Water
Magnetic	Accumulation	N, P (>90%),	25–40°C, pH 8–9,	Solid-liquid	Magnetic material	Leachate from	SIMFLOC, Smit
separation		and K	<1 hr HRT	separation		desorption of magnetic material	Nymegen
Bioleaching/ extraction	Release	N, P (>40%)	20–40 °C, pH 1–3, <3 days HRT	Medium (pH adjustment)	Sulfur and iron source	Leachate	BIOCON
Gas-permeable	Extraction/	N (>90%)	10–80 °C, pH >	High (pH and	Heat	Ammonium sulfate,	
membranes	recovery		9.5, <1 hr HRT	temperature adjustment)		ammonium salts concentrated ammonium liquid	
Innovative technology						,	
Plant	Accumulation	N, P, and K	25–31 °C, pH 6–8, 1–4 months	Low	Light source	Biomass $(1-3\% N)$ and P)	1
Adsorption/ion- exchange	Accumulation	N, P, and K (all >90%)	pH < 8.0, <1 hr HRT	Solid-liquid separation	Adsorbent	57 g P/kg (Zirconium loaded orange waste gel) and 21.52 g N/kg	P-ROC, RECYPHOS, PHOSIEDI, RIM NUT BIOCON
Liquid–liquid extraction	Accumulation/ recovery	N, P (>90%)	15–30°C, pH 2–13, 0.5 hr HRT	Solid-liquid separation	Na ₂ CO ₃ , NaOH, aliphatic, nonvolatile solvents with extractants	(clinoptilolite) Sodium or potassium phosphate (tribasic) solution (0.3%	AD-LLX

SEABORNE, STUTTGARTER VERFAHREN, LO- PROX/PHOXAN, KREPCO, BIOCCON, SEPHOS, AQUARECI, SESAL-PHOS, DASCH	CAMBI, MEPHREC, ASHDEC, THERMPHOS	MEMBOIR	Nonproprietary	Nonproprietary	Nonproprietary	ThermoEnergy Castion	PHOSTRIP, PRISA, DHV CRYSTA- LACTOR, CSIR, KURITA, PHONIX, OSTARA, BERLINER VERFAHEN, FIX-PHOS
Leachate (concentration varies with feed)	Char (concentration	Concentrated	Sludge (5–7% P)	Sludge (1–3% P)	Digested slurry (varies with the feed) biosolids	Ammonium sulfate, ammonium salts concentrated ammonium	Struvite (12% p. 5% N), K-struvite Fe or Ca phosphate
Leaching solution	Heat	1	May require external carbon source	Chemical Al, Fe	I	Heat	Mg or Ca
I	Medium (heating required)	1	Low	Low	Medium (heating may be required)	High (pH and temperature adjustment)	Solid-liquid separation
25–200°C, pH 1–3, <1 hr HRT	150–1100°C, pH all, <1 hr HRT	10–40 °C, pH 6–8, <1 hr HRT	5–40 °C, pH 6.5–8, 0.5 hr HRT	25–40 °C, pH 6–11, <1 hr HRT	35–60°C, pH 6.5–7.5, 20–30 davs HRT	>80 °C, pH > 9.5, <1 hr HRT	25–40°C, pH 8–9, <1 hr HRT
N, P, and K	P and K	N, P, and K	P (15–50%)	N, P (>90%)	N, P, and K	N (>90%)	N, P (>90%), and K
Release	Release	Accumulation	Accumulation	Accumulation	Release	Extraction/ recovery	Extraction/ recovery
Chemical release	Thermochemical release	Membrane filtration Established	EBPR	Chemical precipitation	Anaerobic digestion	Liquid–gas stripping	Crystallization

HRT—hydraulic retention time; embryonic technologies—in laboratory or pilot stage of development; innovative technologies—at demonstration or full scale with limited deployment; established technologies—commonly applied, maybe new to nutrient recovery/waste management sector (in such case commonplace elsewhere).

acidic environments and carry out oxidation of insoluble iron and sulfur compounds, causing the low pH and the release/solubilizing of previously complexed nutrients and heavy metals.

For bioleaching of nutrients from sewage sludge, phosphate rock, and ash, different energy sources such as $FeSO_4$, 90,91 FeS_2 , 92 and elemental sulfur¹⁹ have been provided to a mesophilic mixture containing *At. ferrooxidans* and *At. thiooxidans* strains. ¹⁹ The optimum temperature for growth of these mesophiles is in the range of 20–40 °C and pH in the range of 1.0–4.5. ¹⁹ Because of the low operating pH, the process effectively kills pathogens. ⁹¹ Process configurations that allow continuous bioleaching (e.g., continuous stirred tank reaction with retention times of <3 days) can be superior to batch reactions (with retention times up to 16 days). ¹⁹ Bioleaching is a low-cost process option due to an ability to use elemental or chemically bound sulfur (e.g., mineral metal sulfides) which is usually already present in waste streams in sufficient quantities for the process. The major disadvantage of bioleaching technology is that the release efficiencies for N and P (<40%) are low as compared with that of unwanted heavy metals (>60%), ^{90–95} creating a need for further processing of the leachate.

5. NUTRIENT EXTRACTION AND RECOVERY TECHNOLOGIES

Physicochemical methods can be used to recover the nutrients that were released into a soluble form (e.g., $N-NH_4^+$, $P-PO_4^{3-}$, and $K-K^+$) by the technologies described above. This section focuses on these nutrient recovery techniques which ultimately produce alternative fertilizer products for use in agriculture.

5.1. Chemical Precipitation/Crystallization

Chemical precipitation via crystallization is a phase change process that converts previously dissolved components into a particulate, inorganic compound, for separation from the liquid bulk. During this process, supersaturated conditions (a thermodynamic driving force for precipitation) are created in the waste streams through a change in temperature, pH, and/or by the addition of metal ions. ¹⁶ Because of these conditions, precipitation of selected products can be performed. Struvite (MgNH₄PO₄·6H₂O) crystallization is a well-known example of this technique being applied to simultaneously recover N and P from nutrient-rich streams. ^{96–98} Typically, struvite contains 12% P and 5% N with minimal heavy metal or biological contamination. ⁹⁹ Magnesium is typically limited in waste streams and thus needs to be added in the form of MgCl₂, Mg(OH)₂, or MgO to create supersaturated conditions. The struvite formation reaction is dependent on reactant (NH₄⁺, PO₄³⁻, and K⁺) concentrations and a high pH (pH > 8.0) to ensure that a sufficient

quantity of orthophosphate is in the unprotonated form to participate in the struvite formation reaction.

Chemical precipitation can remove 80-90% of soluble phosphates and 20–30% of soluble ammonia from the waste streams. ¹⁶ The ammonia removal is relatively low, because the waste streams from which nutrients are recovered by struvite often contain a large molar excess of ammonia nitrogen, and due to equimolar stoichiometry of struvite, the excess of ammonia remains in soluble form and is not recovered. Since struvite has a specific gravity of 1.7, the crystals can be readily separated from the liquid bulk by gravity settling, by mechanical separation (filter press) or by the use of an integrated crystallization and separation process. Struvite crystallization is mainly applicable to phosphorous recovery where the waste stream being treated is low in solids content ($<2000 \text{ mg L}^{-1}$)¹⁰⁰ and have a relatively high content $(P-PO_4 > 50 \text{ mg L}^{-1})$. The process is typically operated at short hydraulic residence times of <60 min, a moderately alkaline pH of 8.0-9.0, and an uncontrolled temperature of 25-35 °C. 16 Due to the slow crystal growth rate of struvite, solids-retention times need to be high (>10 days), which assists in the formation of larger aggregated crystals or granules. 16 A fluidized bed design and/or recycle of crystalline product are commonly applied to decouple the solids-retention time (then much longer) from the hydraulic retention time. Alternative products like calcium phosphate, magnesium potassium phosphate, or iron phosphate can be produced in a similar manner, 101 depending on the composition of the waste and the added reagent chemicals.

5.2. Gas-Permeable Membrane and Absorption

Gas-permeable membranes can be used to recover N as ammonia from the liquid phase. In this process, ammonia is transferred by convection and diffusion from the liquid stream across a membrane. Ammonia volatilizes through a hydrophobic membrane and is either condensed or absorbed into an acidic solution. The NH₄–N removal efficiency of a gaspermeable process has been reported to be higher than 90%. A maximum ammonia concentration of 53 g N L⁻¹ (solution containing ammonia) was reported using a gas-permeable membrane with swine waste streams. Following ammonia recovery via membrane concentration, acids such as sulfuric acid are used to recover ammonium as ammonium sulfate ((NH₄)₂SO₄).

Since the process is driven by the difference in partial pressure between the waste streams and the absorbing solution, the performance is better for a higher ammonia concentration in the wastewater. Higher temperatures (up to 80 °C) and pH in excess of 9.0 improves performance by increasing the proportion of ammonia in the free form rather than ammonium ions. 102,104,107 The membranes in this process are typically hydrophobic and may be comprised

of silica, 107 ceramic, 107 polyvinylidene fluoride (PVDF), 104 polypropylene (PP), ¹⁰³ polytetrafluorethylene (PTFE), ^{102,103,105,108} or polymer composites. Asymmetric membranes such as PVDF have a lower mass transfer resistance compared to symmetric membranes. 104 Membranes can be constructed in different configurations including hollow fiber, tubular flat sheet, and spiralwound cylinders and can be used in submerged or external configurations. ¹⁰⁸ Selection and application of these membrane materials and configurations depends on resistance against fouling, flexibility, texture, as well as cost, accessibility, and supply. The main challenges to implementing this technology are the relatively low absorption rate per unit surface area as well as the high capital and operating costs per unit volume of waste streams being treated. 109 These costs arise from the need to adjust pH and temperature. Additionally, this process may not be suitable for recovering ammonia from complex matrices that contain large amounts of hydrophobic compounds such as fats, oils, and grease, due to associated issues with membrane pore blocking and fouling.

5.3. Liquid-Gas Stripping

Gas stripping is a physiochemical process that involves the mass transfer of ammonia from the liquid phase to the gas phase. This process differs from gas-permeable membrane processing, in that all constituents in the waste stream (not just ammonia) are allowed to exert their own partial pressure, thus making the mass transfer less efficient. This transfer is accomplished by contacting the dissolved ammonia with an extractant gas (usually air) and is mainly applicable to situations where the effluent has a relatively high ammonia concentration (NH₄–N > 2000 mg L⁻¹). As with gas-permeable separation, air stripping usually requires an elevated temperature (>80 °C) and pH (>9.5) to increase the proportion of free ammonia in the treated waste streams and in this way decrease the amount of air required. As a result, pretreatment of the feed is critical, involving pH adjustment, solid–liquid separation, temperature adjustment, and carbonate removal. $^{110-113}$

Ammonia removal efficiencies by gas–liquid ammonia stripping of up to 98% have been observed. Papelication of a vacuum can also improve ammonia recovery efficiency. Recovery of the stripped ammonia occurs via condensation, absorption, or oxidation to produce a concentrated fertilizer product. Products from the gas stripping processes include ammonium sulfate ((NH4)₂SO₄), other ammonia salts, or a concentrated ammonia solution. The main challenges to implementing this technology are the relatively high operating cost per unit volume of waste stream treated, the need for a concentrated and pretreated feed, and the production of a spent waste stream (now poor in ammonia and with a high pH) which is not suitable for lagoon storage and/or land application.

5.4. Electrodialysis (ED)

ED is an extraction technology which selectively separates anions and cations across an ion-exchange membrane, driven by an applied electrical field between electrodes. Cationic species ($\rm K^+$ and $\rm NH_4^+$) move toward the cathode passing through cation-exchange membranes (CEM) which allow only positively charged species to pass through while rejecting negatively charged species. Anions (e.g., $\rm PO_4^{3-}$) move toward the anode passing through anion-exchange membranes (AEM) which allow only negatively charged species to pass through while rejecting positively charged species. Through this process, cations and anions are obtained separately in concentrated solutions. ED cells can contain up to several pairs of AEMs and CEMs arranged alternately between the electrodes. Additional cells between the electrodes increase current efficiency as they allow multiple "uses" of the same electron (for each electron that is transferred, ions that match the charge must migrate across all membranes). However, increased membranes also increase the internal resistance and hence power consumption.

ED has the potential to recover all nutrients but is most applicable for N and K, as P can be effectively removed using other lower cost methods. ED is also considered to be appropriate for recovering ions from nutrient streams at low nutrient concentrations (below 2000 mg L⁻¹) and in fact low nutrient concentrations are preferred due to a lower potential for membrane fouling or scale formation. ED has been used to recover ammonia from pig manure, 113,115,116 and source separated urine. 117 A maximum ammonium concentration of 14.25 g L⁻¹ was achieved in the concentrate, which was 10 times that in the manure. ED has achieved K recovery of >99% from winery waste streams and wheat leachates washed from dry wheat biomass. 118,119 Waste streams with an acidic to slightly alkaline pH (<8.0) are preferred due to improved nutrient solubility and ion transfer through membranes. The process requires about 3.25-3.60 and 1.2-1.5 kWh to remove 1 kg of N-NH₃^{113,115,116} and K, ¹¹⁸ respectively. Successful application of this technology in full-scale facilities may be hampered by the high energy consumption and by the requirement for chemical additives for the regeneration of the membranes, and membrane fouling and heavy metal contamination can also be significant issues. Recently, a development was reported where a microbial fuel cell was combined with an ED process in order to harness the current produced by bacteria degrading organic matter in the waste to reduce the external power required for ED. 120

6. NUTRIENT RECOVERY TECHNOLOGY SUMMARY

Table 1 summarizes the current state of development/adoption for each of the nutrient recovery technologies introduced above. The level of adoption

Piggery Poultry Cattle Food industry Meat processing Landfill Tannery Bioaccumulation EBPR Chemical accumulation Algae accumulation Plant accumulation Adsorption/ion-exchange Membrane filtration Magnetic method Anaerobic digestion Thermochemical Chemical release Bioleaching/extraction Crystallization Gas-permeable membranes Liquid-gas stripping Electrodialysis Legend:

TABLE 2. Extent of peer reviewed research performed on nutrient recovery technologies

>1000 citation

0 citation

was categorized as embryonic, innovative, and established. Embryonic technologies are still in the laboratory or pilot stage of development, operating at well below commercial scale. Innovative technologies are operating at a demonstration or full scale with limited deployment, but the level of deployment makes it possible to evaluate the prospect of future wide-spread application at commercial scale. Established technologies include commonly applied processes, even if new to and not fully deployed in the nutrient recovery/waste management sector (in such case the technology is commonplace elsewhere).

Table 2 summarizes an extensive literature analysis that was performed to identify the prominence of research-to-date on the respective nutrient processing technologies for various waste stream types. Table 2 shows that bioaccumulation, anaerobic digestion, and thermochemical release have received considerable research attention across most of the listed waste streams. The relative prominence of research on these technologies may be reflecting drivers to date for nutrient processing technologies, which have largely been nutrient load management and waste volume reduction to reduce environmental impacts and disposal costs. Nutrient reuse has to date been a much lesser focus, and consequently, nutrient recovery processes have generally received much less research attention. Table 2 also appears to reflect typical differences in market drivers for various waste streams, that is, domestic wastewater and food processing have received a substantially greater level of research attention than the other waste streams. Limited land application of nutrients and costs of transporting unwanted nutrients offsite provide some drive for adoption in agriculture. However, reduction in trade waste charges associated with food processing offers much stronger financial incentives for nutrient extraction and recovery prior to sewer discharge.

Moreover, sewage treatment plants have commonly operated under political governance (rather than private sector governance) with the result being that market conditions are more stable within the context of longer term capital planning. Such financial stability is expected to have resulted in more reliable sources of seed funding for ongoing research and development work. Hence, the prominence of research/investigative work on all the treatment technologies as applied to domestic wastewater (Table 2).

7. ANALYSIS

This section identifies and discusses the key technical challenges associated with broad scale adoption of nutrient recovery technologies, including (1) waste stream specific characteristics, (2) technology feasibility based on existing knowledge, and (3) the required characteristics of the nutrient product that is ultimately produced for end use. These issues are again considered for the three-step implementation (accumulation, release, and extraction/recovery). Overall, it is assumed that the simplest and economically most feasible solution that can achieve the desired outcomes for a specific context would be the best overall process solution for that specific context.

7.1 Impact of Waste Stream Characteristics on Technology Selection

As discussed above for the specific technologies, waste stream characteristics heavily impact on the level of complexity required for an overall nutrient recovery system including characteristics such as nutrient concentration, nutrient form, and whether the nutrient is present in particulate or soluble form. To show these aspects, Table 3 summarizes the respective need for nutrient accumulation, release, and concentration technologies based on various waste stream characteristics.

As again highlighted in Table 3, nutrient accumulation techniques are most needed where waste streams have low nutrient concentrations

TABLE 3. An analysis of the need for n	iutrient accumulation,	release, and	extraction	based
on waste stream characteristics				

Waste stream characteri	stics	Nutrient accumulation		Nutrient extraction
Nutrient concentration	High (>100 mg L ⁻¹)	*	**	***
	Low ($<20 \text{ mg L}^{-1}$)	***	*	*
Nutrient form	Soluble	***	*	***
	Particulate or organic substances	**	***	*
Insoluble contaminants	High concentration	**	**	*
Soluble contaminants	High concentration	*	*	*

^{***} Need is high; ** need is medium; * need is low.

(<20 mg L $^{-1}$) and high flows. For example, as shown in Table 2, nutrient accumulation from domestic wastewater has been widely studied for various accumulation techniques, because P in domestic wastewater is primarily in soluble form (>50%) and at low concentrations of 6–8 mg L $^{-1}$, but wastewater flows are high. 30 After accumulation, a release technology such as chemical and thermal treatment is appropriate to produce a liquid effluent with soluble P content in excess of 100 mg L $^{-1}$, from which the P can then be more readily recovered for reuse.

Accumulation techniques are not so essential when nutrients in the waste stream are predominantly present as particulates. Nutrients in the form of inorganic particulate can be readily separated and concentrated via simple gravity separation or filtration. For organic particulates, a release step may be required (Table 3) to further process the nutrients into a final nutrient product with suitable qualities. An example would be the processing of particulate (organic) nutrients in municipal biosolids into soluble form via anaerobic digestion for subsequent recovery.

The presence of contaminants (both soluble and insoluble) can impact on the efficiency and economics of a nutrient recovery process. Table 3 shows that nutrient extraction/recovery technologies are more tolerant to insoluble contaminants. Pretreatment by solid-liquid separation can readily remove such solid impurities from the waste stream to below 2000 mg total suspended solids L⁻¹, so subsequent processing steps are unaffected by the remaining solid impurities. Removal of soluble contaminants prior to nutrient extraction/recovery (e.g., heavy metals, Na⁺, Cl⁻, Ca²⁺, and carbonates) can be more challenging and may require additional pretreatment or posttreatment steps, such as the reduction of water hardness (softening) or elevated temperature treatment to volatilize metal complexes. For example, calcium and magnesium hardness are precipitated as a pretreatment step to reduce the potential for carbonate scale formation during liquid-gas stripping of ammonia. 121 In all cases, additional pretreatment or posttreatment increases the overall cost and complexity of a nutrient recovery processing train.

7.2. Feasibility of Nutrient Recovery Technologies

The level to which specific nutrient recovery technologies will be adopted in the future will depend on: (1) the capital and operating costs of the technologies; (2) the engineering feasibility, maturity, and reliability of the technologies; (3) the credibility and completeness of available information on the technologies; (4) the safety profile of the technologies; and (5) the environmental concerns and benefits associated with the respective technologies. Table 4 provides an overview of the current status of the technologies outlined above, in terms of these feasibility considerations. This

TABLE 4. Summary of technology feasibility

	Engineering feasibility ¹	Capital cost	Technology maturity ²	Operability ³	Information credibility and availability	Operating cost	Safety issues	Environmental concerns ⁴	Other environmental benefits ⁵
Nutrient accumulation Plant accumulation	Med ⁶	Low	Med	Low	Мед	Low	Low	Low	Reduces COD and
Algae accumulation	Med: complex technology	Low	Med	High	Med	Low	Low	Low	Reduces COD and BOD
EBPR accumulation Chemical accumulation	High High	Low	High High	Med High	High High	Low High: chemical	Low	Low High: produces	1 1
Adsorption/ion- exchange	Med	Med	Low	Med	Low	Med: require adsorbent	Low	Low	I
Membrane filtration	High	High	Med	Мед	Мед	High: membrane clogging and cleaning	Low	Low	The filtrate from the process can be recycled
Magnetic separation	Low	High	Low	Low	Low	Low	Low	Low	I
Anaerobic digestion	High	Med^6	High	Med	High	Low	Med: corrosive H ₂ S is produced as	Med	Produces energy, reduces GHG emission, reduces COD and BOD
Thermochemical	Med	High: heat resistant equipment	Med	Low	Мед	High: heat required	High	High: flue gas disposal	Destroys toxic organic matter and biological contaminants, reduces volume of waste
Chemical extraction/leaching	Мед	High: acid resistant equipment	Low	Low	Low	High: acid required	Med	Med: low pH leachate disposal	Heavy metals can be separated from waste
Bioleaching/ extraction	Med	Low	Low	Low	Low	Low	Med		-

(Continued on next page)

TABLE 4. Summary of technology feasibility (Continued)

	Engineering feasibility ¹	Capital cost	Technology maturity ²	Operability ³	Information credibility and availability	Operating cost	Safety issues	Environmental concerns ⁴	Other environmental benefits ⁵
Nutrient extraction Crystallization (struvite)	High	Med^6	Med	High	High	Med: requires	Low	Low	I
Liquid–gas stripping	Med	High	Low	Med	Low		High	High	ı
Electrodialysis	Low	High	Low	Low	Low	red	Med: hydrogen is produced at the electrode	Med (heavy energy user—can be combined innovatively with biofuel cell to reduce energy needs	I
Gas-permeable membranes	Low	High	Low	Low	Low	High: membrane clogging and cleaning costs	Med	Med	I

⁶Med refers to an evaluation of medium.

²Engineering feasibility refers to the level of expertise to design/specify particular technologies and to service operation.

²Technology maturity refers to the level of adoption (e.g., full-scale widespread; high, demo to full-scale sparse; medium, lab-scale to pilot only; low).

³Operability refers to the ease of operating the plant/complexity of the systems and the level of current knowledge and expertise available to ensure ongoing robust operation.

⁴Environmental concerns primarily deals with emissions/recalcitrant by-products.

⁵Environmental benefits captures mitigation of environmental risks.

section discussed these considerations for the three-step implementation of accumulation, release, and extraction/recovery.

7.2.1. ACCUMULATION TECHNOLOGIES

As expected, engineering feasibility is very high for technologies with a proven track record, such as tertiary filtration, chemical precipitation, and EBPR. However, it is noted that the bulk of prior work for these technologies has been on treatment of domestic wastewater (Table 2). As shown in Table 4, high capital cost and low operability are expected to limit the feasibility of magnetic-based accumulation, while large footprint will likely continue to limit plant-based accumulation. As noted above and in Table 2, metals precipitation (with iron and aluminum being most efficient) and bioaccumulation are widely applied in sewage treatment plants. However, in view of a nutrient recovery and reuse perspective, purely bioaccumulation techniques may ultimately outcompete combined chemical and bioaccumulation. This is because phosphorus which is strongly bound to iron and aluminum precipitates is unavailable for crop growth and may require further processing steps which add to complexity and processing cost. Bioaccumulation (EBPR) as a stand-alone technology is cost-effective and widely studied for low margin agriculture waste streams also (Table 2). Environmental benefits such as reduction in COD and BOD levels in treated waste streams are additional benefits of bioaccumulation and plant-based accumulation (Table 4).

7.2.2. Release Technologies

It is expected that anaerobic digestion will continue to be constrained by the biodegradability of the feedstock. For this reason, thermochemical alternatives, which are insensitive to degradability, will always have a place in the market. The loss of P to sludge is another common issue with anaerobic digestion, when high background metals (calcium and magnesium) and P concentrations cause solids precipitation, rather than the preferred scenario of nutrients remaining in soluble form for subsequent extraction/recovery. Particulate nutrients that end up in the unbiodegradable biosolids are not available for recovery into higher value products.

The operability of thermal release technologies was considered to be low compared to anaerobic digestion, reflecting the relative complexity of thermochemical plants. The high treatment temperatures of thermochemical plants and the associated safety, operability, and engineering concerns, result in a much more complex plant with more costly infrastructure (Table 4), lending such facilities to centralized applications. Similar complexities with chemical safety apply to pure chemical release using acid leaching.

7.2.3. Extraction Technologies

Struvite crystallization is relatively simple and easy to scale-up and is increasingly being adopted in both municipal and agro-industrial applications (Table 2). It is currently considered to be the most readily adoptable technology for P recovery. Consequently, a high feasibility was ascribed to struvite crystallization (Table 4) even though application is currently less prominent than other mature technologies. ED and gas-permeable membrane recovery were classed as embryonic extraction/recovery technologies because they were yet to be applied at pilot to demonstration scale (Table 4, low engineering feasibility and technology maturity). However, these two extraction/recovery technologies are likely to be essential for N and K recovery into the future. It is expected that into the future, the application of liquid–gas stripping for N recovery will continue to be limited by the requirements for high concentrations (>2000 mg L⁻¹), high pH, and high temperature resulting in high operating costs and causing safety concerns and operability issues.

7.3. Nutrient Products for Sale

A key requirement for industry-wide adoption of extractive nutrient recovery is the need to produce value-added products that have use in a secondary market. Since over 90% of all P-based products are associated with the agricultural sector, 6 it is appropriate for extractive nutrient recovery options to target products to the agricultural sector. It is expected that in the short-tomedium term, the products from nutrient recovery will mainly offset treatment costs. 122 However, in the longer term, as technologies mature and the value of nutrients increase, the income from alternative fertilizer sales may become a major driver for widespread technology adoption. The initial target should be to continue harnessing the value of existing products such as biosolids (relatively low value but relatively low cost of production), while developing new products that more closely resemble competitor products on the market and that targets increased end-user acceptance. As briefly discussed below, the benefit from nutrient recovery is likely to be site specific and will be based on the products recovered and the local demand for niche products. At present, there are four main nutrient products that are seen to show continued potential. These are: (1) biomass, (2) biosolids, (3) char/ash, and (4) chemical nutrient products. This section considers some of the key characteristics of each of these products

7.3.1. BIOMASS

Nutrient-rich biomass derived from plant, algae, and microbial accumulation techniques can be used as animal feed, as raw material for nutrient release processing, or as feedstock for biofuels production. The application of activated sludge biosolids has been broadly investigated from a contamination

Technology	Existing knowledge	Application development	Product development
Plant accumulation	$\checkmark\checkmark$	$\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{\sqrt{$	$\sqrt{\sqrt{\text{(Identify agronomic release rates)}}}$
Algae accumulation	$\checkmark\checkmark$	$\sqrt{}$	$\sqrt{\sqrt{}}$ (Including high-value products)
EBPR accumulation	$\sqrt{}$	$\sqrt{\sqrt{\text{(Extension and integration only)}}}$	V
Chemical accumulation Adsorption/Ion-exchange Magnetic separation Anaerobic digestion		$\sqrt{}$ $\sqrt{}$ (Improved	$\sqrt{\sqrt{\sqrt{(Improved solids)}}}$
Thermochemical Extraction/leaching Bioleaching/extraction Struvite crystallization Liquid–gas stripping		nutrient release)	√√√ (Char) √ √ √
Electrodialysis Membrane filtration Gas-permeable membranes	\ \/ \/	\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\\	$\sqrt{}$ (N and K concentrated product) $\sqrt{}$

TABLE 5. Summary of existing knowledge and research needs to facilitate widespread adoption of nutrient recovery technologies

 $\sqrt{\sqrt{\sqrt{R\&D}}}$ need is high; $\sqrt{\sqrt{R\&D}}$ need is moderate; $\sqrt{R\&D}$ need is low.

point-of-view and less so from a benefits point-of-view (see next section). The application of other biomass streams is yet to be assessed to the same level of detail as biosolids. Direct application of intact biomass for agricultural purposes has been identified as a possibility; however, research into this application is lacking. For instance, nutrient release rates from different biomass feedstocks applied directly to land are currently not well characterized (Table 5).

7.3.2. Biosolids

Biosolids, a solid product stream produced by anaerobic digestion, can have a high-nutrient content (\sim 4% P and \sim 2% N), making it an attractive product for direct land application of nutrients as well as a soil conditioner to improve soil carbon content. Indeed, studies have found that biosolids have equal or better performance as agricultural amendments when compared with commercial fertilizers. Nevertheless, there continues to be environmental and human health concerns regarding the use of biosolids in agriculture, with pathogens, heavy metals, and trace organic contaminants being key issues. Removal of metals from biosolids can be achieved using chemical extraction but with considerable added cost 125 and codissolution of nutrients and heavy metals can require further posttreatment. Legislation

targets the quality and application rates for biosolids to reduce the associated impacts of the heavy metal content and nutrient loads. Extractive nutrient recovery helps by extracting N and P from biosolids, reducing the load of nutrients in the biosolids, allowing producers of biosolids to better manage the N and P content of the biosolids to match the application needs. The extracted chemical products (such as struvite or other phosphate minerals or aqueous ammonia and derived ammonium salts, see Section 7.3.4) are stable with minimal organic content, and will therefore be less costly to store and transport than the biosolids. The extracted products then can be potentially sold in a secondary market.

One of the major challenges with biosolids as a primary vehicle for nutrients is the expense associated with transport to the site of application/disposal. Moisture content is typically high at 80–90%, ¹³ making biosolids very bulky and costly to transport from urban regions where it is produced to rural regions where the nutrients are used. ¹²⁶ This is clearly shown by comparing the current (2013) value of nutrients in biosolids (approximately \$US8 per tonne biosolids) with the much higher transport cost for a 50 km distance in USA or Australia (about \$US30 per tonne) and transport costs are even higher in Europe. ¹²⁷ For this reason, processes that further dewater digestate/biosolids into pelletized or granulated fertilizer products can be useful. However, importantly, further processing does require significant energy inputs, with a minimum of 600 kWh of energy (as gas) needed to evaporate 1 tonne of water. Solar drying can help to reduce energy demand to 30 kWh of electricity per tonne of water evaporated, ¹²⁶ but is limited to suitable climates.

7.3.3. CHAR AND ASH

The use of char and or ash from thermochemical processes for soil amendment is becoming increasingly popular, because of the potential benefits of soil carbon sequestration, heavy metal immobilization, improvement in soil quality, increased crop yields, mitigation of nutrient leaching, and organic contaminant remediation. Research has indicated short-term benefits of direct application, but additional research is required to determine the long-term effects of char on nutrient availability and soil microbial and fauna communities. Char can also be reused within the construction industry, without exploiting the nutrient content. Similar to biosolids, the reuse of ash and char as agricultural amendments will be limited by heavy metal content. Chemical extraction can be used to process ash and char to extract the remaining nutrients. However, posttreatment of the treated ash/char may then be required for heavy metal removal at greater cost and may limit adoption. 129

7.3.4. CHEMICAL PRODUCTS

Nutrient extraction technologies can recover N and P as particulate or soluble inorganic fertilizers that are readily useable in agriculture. At present, struvite

(magnesium ammonium phosphate) is a primary focus of several commercial technologies. Struvite has been widely cited as a suitable slow-release fertilizer. It is sparingly soluble in water and research has suggested that it has comparable performance to a fertilizer from phosphate rock. ^{130,131} Overuse of struvite can result in magnesium accumulation in soil. However, magnesium levels can be managed using accurate fertilization ¹³² and by selecting crops that tend to accumulate magnesium (e.g., grains, legumes, and dairy cattle). One benefit of struvite recovery is that the process selectively rejects heavy metals to produce a product that easily meets regulatory limits. ^{133–136} Additionally, struvite with low moisture content can have negligible pathogen and trace organic contaminants. ¹³⁷ Other products with potential fertilizer value can include calcium phosphate (hydroxyapatite), iron phosphate (vivianite), phosphoric acid, ammonium sulfate, and ammonium nitrate.

Nitrogen recovery through liquid–gas stripping, gas-permeable membrane, and ED can produce an aqueous ammonia solution which can be used as a fertilizer or for the denoxification of exhaust gases of power stations and waste incinerators. The aqueous ammonia can be further converted into solid inorganic fertilizer such as NH₄NO₃ or (NH₄)₂SO₄. At present, the economic feasibility of N-only recovery is low, largely due to high chemical cost to adjust pH to increase the free ammonia concentration (NH₄⁺ to NH₃), due to the heat required to decrease ammonia gas solubility and drive ammonia stripping, and due to the relatively low cost of competing ammonia products from the Haber–Bosch process. The cost margins may close in the future with the rising costs of treatment of nitrogen and natural gas (gas is used to manufacture ammonia through the Haber–Bosch process). Additionally, it may be possible to target N products to specific niche markets, which may increase the value of the recovered product.

7.3.5. Nonnutrient Products

Use of biological accumulation techniques can allow for the recovery of other byproducts, which can provide add-on value to the technologies. For instance, algae and duckweed can be used as feedstock for energy production (e.g., biofuels or biogas) or as a source of protein for animal feeds due to their high protein content. Biological release methods like anaerobic digestion can also be coupled with nutrient recovery processes to produce methanol, ammonia, or other products from digester gas (e.g., sulfide, sulfur, and hydrogen). These alternative nonnutrient recovery products can be used for a variety of purposes, including use as raw materials for manufacturing of hydrogen peroxide, polymers, solvents, pharmaceuticals, and other products.

8. OPPORTUNITIES AND NEEDS FOR FURTHER WORK

This section outlines some key needs and directions for future research. Overall, this review has identified a need to develop both the respective technologies and the products being produced for the secondary market. Development of the respective technologies (here termed application development) aims to establish viable processing options out of embryonic technologies, aims to better tailor mature technologies by integration using the three-step framework of accumulation, release and extraction/recovery, and aims to better match the technology solutions with the available economic drivers for adoption. Product development targets nutrient products that are of a higher quality and that matches the requirements of the market and also aims at developing high-value by-products to drive initial uptake of nutrient recovery technologies. Table 5 provides an overview of the level of current knowledge, and the needs for further research toward application and product development.

8.1. Application Development

Design, operation, and economic assessment is lacking for many of the innovative and less mature technologies, such as adsorption/ion-exchange, plant accumulation, and chemical extraction applied to nutrient recovery from wastes (primary P extraction is mature in the conventional fertilizer production industry). Full-scale implementation experience is also lacking. Further pilot scale development is required for embryonic technologies such as ED, gas-permeable membrane, and magnetic methods. As discussed above, these technologies are expected to be indispensable for N and K recovery. In this regard, N and K recovery via bioaccumulation using microalgae or purple nonsulfur bacteria is also seen as promising. Further research should aim to seamlessly integrate N and K technologies with established release technologies such as anaerobic digestion and P extraction/recovery processes such as chemical crystallization.

At present, no single technology can effectively recover all the nutrients in a waste stream (N, P, and K). The more likely future scenario will be integrated processes using the three-step framework of accumulation, release, and extraction/recovery. Economic analysis of entire integrated recovery process trains should consider location, because economically feasible pathways may vary at regional, national, and international level. Demands for resources can differ at these respective levels. The optimum technology solution may also depend on the specific context of the nutrient producer. For example, industrial producers (such as food processors or large localized agricultural activity) may harness more complex nutrient recovery technologies, due to the strong financial drivers of reduced trade waste/waste management and the benefits and cost savings of energy recovery. In contrast, rural agriculture contexts may target simple nutrient load management with low-cost treatment systems and predominantly low-value nutrient products.

Nutrient recovery processes must focus on being sustainable by minimizing process inputs (water, chemicals, energy) through better use of the intrinsic resources of the waste. As nutrient management and recovery is interlinked with water and energy issues, nutrient recovery objectives must align with the emerging concept of "plants of the future" whereby advanced waste treatment facilities meet stringent effluent nutrient limits while maximizing water reuse and energy recovery. For this reason, energy recovery technologies such as anaerobic digestion will continue to be common place. Other nonbiological release technologies are also moving more toward energy self-sufficiency or are being smartly integrated with other energy recovery technologies to close the energy loop. An example would be a thermal hydrolysis system, followed by anaerobic digestion with power generation and heat recycling to provide the energy requirements for the thermal hydrolysis. Increasing water awareness will likely increase consideration of water efficient technologies such as solid-phase anaerobic digestion and/or the operation of sludge digestion at higher sludge concentrations.

Further research should target a reduction in operating costs associated with N, P, and K technologies. Options may include the use of alternative sources (potentially waste) of chemical raw materials required by the process. Another option could be to engineer processing technologies to recover additional nonnutrient sale products that improve the economics of nutrient recovery. In this regard, ED, microalgae, and alternate biological release technologies will offer additional value in by-products.

8.2. Product Development

There is a need to diversify the type and quality of recovered nutrient products. It is expected that end users (and environmental legislation) will increasingly require the production of chemical products with high-nutrient content, low moisture, and very low heavy metal and pathogen contamination. In this regard, the coupling of biosolids, manure, and ash/char production with extractive nutrient recovery technologies will help manage the nutrient content of bulky organic products as well as fully harness the benefits of the extracted nutrients. Identification of the most relevant products will require consideration of local agricultural and industrial demands. Emerging technologies that concentrate and repackage nutrients can help decouple end users from source risk, can reduce social taint, and can value add to the original waste streams. The broad range of suitable technologies in the future will be producing a diverse and broad range of marketable products. Importantly, the products that become available will need to undergo extensive agronomic validation.

Into the future, the development of robust integrated technologies and high-value tradable nutrient products will allow the next step of international trade of waste-derived nutrient fertilizers. Such a global nutrient trade can help rectify national nutrient imbalances, and allow net food importers (by mass) such as the Netherlands and Japan to return nutrients to exporters such as Australia.

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