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Recent technological developments and challenges for phosphorus removal and recovery toward a circular economy

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ABSTRACT

This review aims to summarise the current state of the art technologies for phosphorus recovery from waste and wastewater. Information corroborated here shows a clear relationship between $\text{PO}_4\text{-P}$ content in the liquid phase and the cost of phosphorus recovery. In fact, all current commercial scale operations in this review involve a phosphorus-rich waste stream. In most cases, phosphorus recovery is achieved via two key steps: solubilising phosphorus into water and then phosphate recovery via chemical precipitation/crystallisation. Recent development has also included enrichment and pre-treatment of the phosphorus rich liquid stream. Phosphorus is also a contaminant in the aquatic environment. Thus, this work also reviews the post-treatment of the liquid stream after phosphorus recovery for environmental discharge or water reuse. This review places a spotlight on the requirement for further research work especially on phosphorus enrichment at pilot- and full-scale level. The review also demonstrates the need for further research on pre-treatment and post-treatment to complement the recovery process via chemical precipitation.

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1. Introduction

Phosphorus is essential to life in many aspects (Liu and Chen, 2014). It is a key element in the molecular structure of nucleotides in DNA and RNA, primary energy carriers in cells (e.g. adenosine triphosphate), bones and teeth (Fan et al., 2020; Jones et al., 2020). In the economy, phosphorus is also a key ingredient of fertilisers for agricultural production (Lusk et al., 2017). Elementary phosphorus is also essential for many high-tech industries such as pharmaceutical, fine chemical, food, and electronic manufacturing (Jones et al., 2020; Jung et al., 2020). Thus, a sustainable supply of phosphorus is essential to future generations.

Phosphate rock, which is a finite resource, is currently the principal source of phosphorus supply (Liu et al., 2008). The global distribution of phosphate rock is geographically uneven. Three quarters of minable phosphate rock are in

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Morocco and Western Sahara (Smol et al., 2020). The remaining is scattered over several countries including China, the USA, and Russia, which also have a significant demand for phosphorus for domestic consumption (Smol et al., 2020). This geographically uneven distribution of such an essential resource is further exacerbated by geopolitical tension to create price volatility and unstable supply, heightening the risk of global food shortage. The price of phosphorus has dramatically increased in recent years, especially since the tension and armed conflict between Russia and Ukraine. Given the absence of any significant renewable phosphorus supplies (through recovery), complete depletion of the current reserve of phosphate rock is imminent although the expected time is still debatable.

In addition to the depletion of phosphorus as an essential resource, phosphorus in the aquatic environment is also problematic. The accumulation of phosphorus, derived from human waste and agricultural runoff, in waterways results in anthropogenic eutrophication and related problems to the environment and human activities (Schindler et al., 2016; Vinçon-Leite and Casenave, 2019). Eutrophication leads to the enormous blooms of algae that cause the depletion of oxygen in water and decimate marine ecosystems (Heffernan et al., 2010; Vu et al., 2020a). Moreover, some blue-green algae (cyanobacteria) can excrete toxins that are toxic to humans and animals (Vu et al., 2020a).

Given the imminent depletion of phosphate rock reserve and ecological issues caused by discharge of phosphorus into the aquatic environment, phosphorus removal and recovery from nutrient-rich streams are urgently needed to ensure sustainable development and food security. Examples of phosphorus-rich stream as a renewable source of phosphorus supply include wastewater and sewage sludge from industrial and urban wastewater treatment plants (WWTPs). Up to 25% of the global phosphorus flow is through wastewater (Egle et al., 2016; Mihelcic et al., 2011).

Several methods for phosphorus removal and recovery from waste and wastewater have been explored and commercialised in recent years. These methods include chemical processes (e.g. precipitation) (Barua et al., 2019; Daneshgar et al., 2018; Vu et al., 2022a), physical processes (e.g. adsorption and ion exchange) (Inoue et al., 2009; Li et al., 2016), biological processes (e.g. constructed wetlands, enhanced biological phosphorus removal, and microalgae cultivation) (Rezk et al., 2019; Nguyen et al., 2020; Vu et al., 2022b). The treatment mechanism and application of each technology to extract phosphorus from waste and wastewater have been extensively discussed and reviewed in the literature. Previous reviews have highlighted the need for phosphorus preservation or focused on specific recovery technologies. To date, there has been very little discussion to guide future research work for integrating all necessary steps in phosphorus recovery for improving economic viability.

This review aims to address the high cost of recovering phosphorus from organic waste and wastewater, which is a major bottleneck to commercial scale phosphorus recovery. This is achieved by first summarising the current state of the art technologies for phosphorus removal and recovery from waste and wastewater. Emerging technologies are then identified and systematically discussed to develop a roadmap for sustainable phosphorus management. The review focuses specifically on integrated technologies for phosphorus recovery from waste and wastewater as well as suitable reuse options.

2. Phosphorus in the economy

2.1. Phosphorus flow

Most of the current global phosphorus consumption is in the form of fertilisers for agricultural production. Yet, there are several major leakage points, through which phosphorus is lost and pollute the aquatic environment. Using annual fertiliser consumption data in the world and the mass flow of phosphorus fertiliser through agricultural production in a well defined region (Fernandez, 2022; Parasana et al., 2022), the magnitude of these leakages at the global scale is illustrated in Fig. 1. These leakage points also present major opportunities for phosphorus recovery for circularity to ensure future availability of this essential mineral.

Approximately 70% of total phosphorus fertiliser is lost to the aquatic environment through agriculture run-off. Phosphorus loss in agriculture run-off from farm land occurs via three processes: (1) attached to the sediment that erodes from the field, (2) dissolved in the surface water runoff, and (3) dissolved in leachate and carried through the soil profile (Alewell et al., 2020; Vaccari et al., 2019). On tilled land, most of phosphorus loss is through erosion, whereas on untilled land, phosphorus loss is mostly through the dissolution in surface water runoff or leachate (Vaccari et al., 2019). Phosphorus loss through agriculture run-off can be minimised by a range of modern cultivation techniques such as soil conservation (to reduce erosion and run-off), on-demand irrigation and water reuse, and controlled release fertiliser (Sharpley et al., 2015).

The remaining 30% of phosphorus loss is via industrial and municipal waste and wastewater discharge (Fig. 1). Although this is smaller than phosphorus loss from agriculture run-off, the opportunity for phosphorus recovery is much more significant. For example, urine and faeces from human and livestock are rich in phosphorus. Each year, through urine and faeces, livestock animals and humans release about 10 Mt of phosphorus to the environment (Mihelcic et al., 2011). If recovered, phosphorus from urine and faeces can offset 25% of the global phosphorus demand for fertiliser (Mihelcic et al., 2011). In practice, only about 10% of phosphorus from these sources is currently recovered and reused (Mihelcic et al., 2011). Most of this is in biosolids from wastewater treatment and is used for land application.

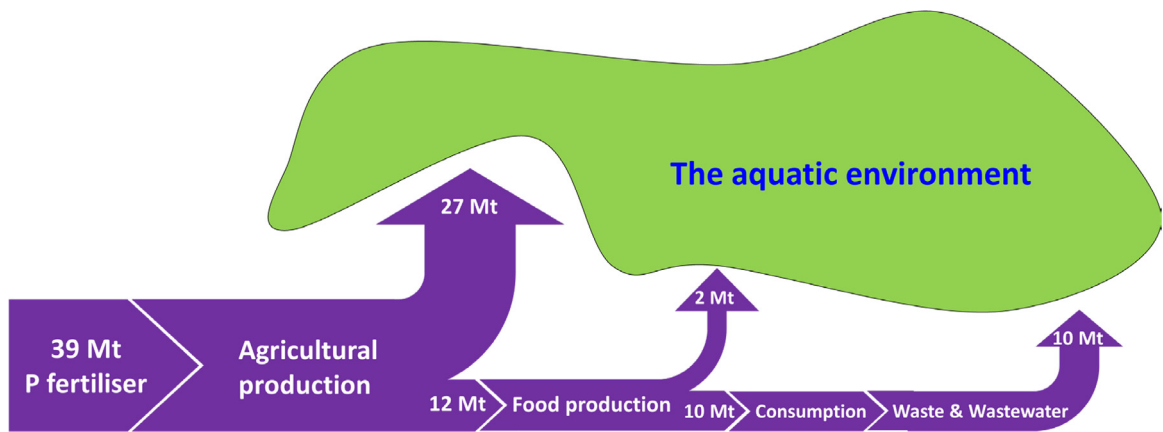


Fig. 1. Mass flow of phosphorus through agricultural production.

2.2. Elementary phosphorus for high-tech industries

In addition to agricultural production, phosphorus is also a critical element to many industries. Elementary (white) phosphorus is a key feedstock for producing high-purity phosphorus compounds for the high-tech industries. Examples of these industrial phosphorus compounds include phosphoric acid, phosphorus trichloride, phosphorus sulphide and sodium hypophosphite. Pure phosphoric acid is indispensable for some food processing and semi-conductor manufacture (Jones et al., 2020; Jung et al., 2020). Phosphorus trichloride is important in producing chlorinating agents, additives for the plastics, lubrication oils, insecticides and flame retardants (Dahlin et al., 2020; Zabihi et al., 2020; Zhang et al., 2020). Phosphorus sulphide is used to produce insecticides and lubrication oils (Dahlin et al., 2020; Valoriani et al., 2020). Sodium hypophosphite is used for nickel plating to produce hard disc drives for computers (Smol et al., 2020).

Although industrial application only accounts for less than 5% of the current phosphorus flow, this is an area of opportunity for phosphorus recovery from waste and wastewater. In some cases, it is more economical to purify and obtain high-purity phosphorus from waste than from phosphorus rock. The cost of recovery and purification can be offset by the high economic value of high-purity and specialised forms of phosphorus such as vivianite for industrial applications (Wu et al., 2019).

2.3. Peak phosphorus

Fig. 2 shows the global consumption of phosphorus since the beginning of the industrial revolution and into future years. The decrease in consumption is driven by supply constraint, showing phosphorus peak by about 2030 and complete phosphorus depletion by 2150 if phosphorus is not recovered from waste and wastewater. The business as usual scenario in Fig. 2 is constructed by accounting for the proven global phosphorus reserve and current phosphorus demand assuming no significant increase in phosphorus recovery (Cordell et al., 2009). If 60% of the current phosphorus consumption can be offset through recovery or loss prevention, the same calculation shows that peak phosphorus can be delayed by several decades and complete phosphorus depletion can potentially be avoided (Fig. 2).

To eliminate the risk of complete phosphorus depletion, some European countries have enacted legislation to mandate phosphorus recovery from wastewater treatment (Sichler et al., 2022). Starting from 2029, all German WWTPs are required to recover phosphorus if the phosphorus content in their sewage sludge is >20 g/kg (in dry weight). Switzerland has also introduced a mandatory phosphorus recovery commencing from 2026 for all WWTPs serving more than 1000 person equivalent, regardless of the phosphorus content in sludge.

In addition to the forecast of phosphorus exhaustion from the peak model, environmental and socio-economical issues associated with the mining activities of phosphate rock (Zhang et al., 2022) are additional drivers for an increasing need to recover phosphorus from other sources especially waste and wastewater. The imminent depletion of global phosphorus reserve can undermine food security and geopolitical stability. The over-exploitation of phosphate rock and the abundance of phosphorus in the aquatic environment due to discharge of wastewater and agricultural run-off into water bodies are major causes of ecological disasters (e.g. irreversible eutrophication, algae bloom, and toxic heavy metal contamination in mining areas) (Liu and Chen, 2014; Liu et al., 2008; Bunce et al., 2018).

3. Technologies for phosphorus management

3.1. Hierarchy for phosphorus preservation

There is a broad range of technologies for phosphorus management from immobilisation to recovery for slow release phosphorus fertilisers (e.g. struvite). In general, the cost of treatment and recovery increases as the phosphorus content in

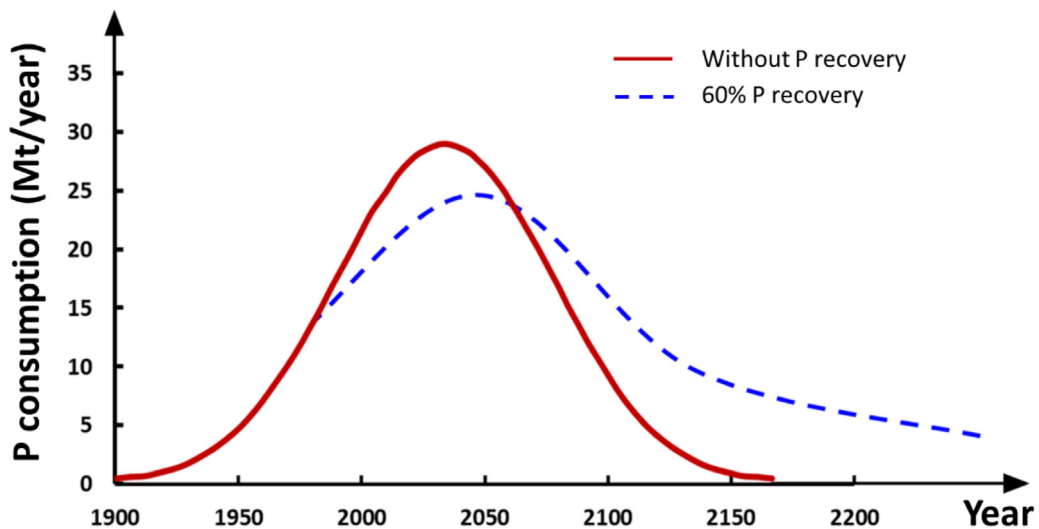


Fig. 2. Global phosphorus consumption as a function of time. Baseline data are from Cordell et al. (2009).

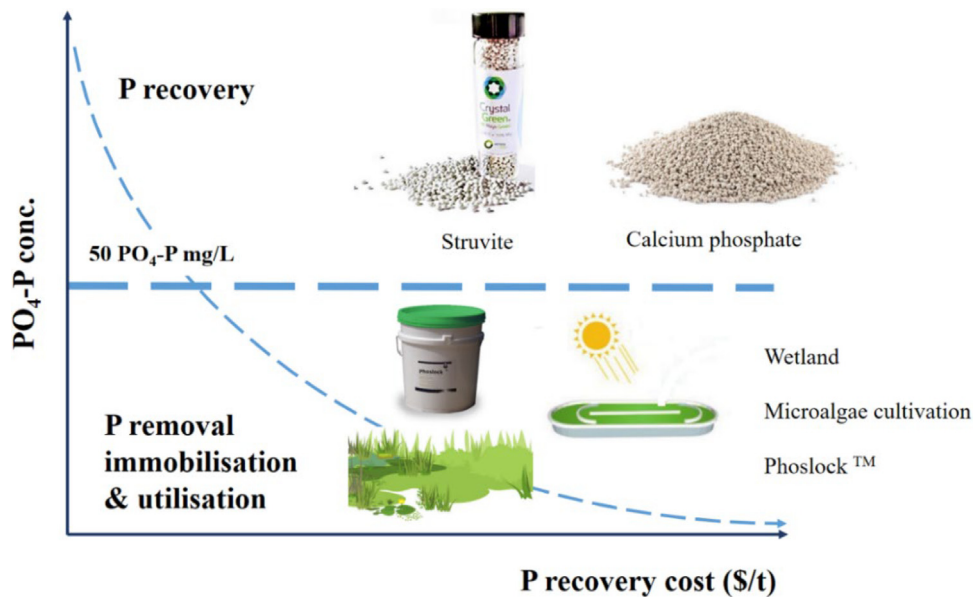


Fig. 3. Relationship between $\text{PO}_4\text{-P}$ content in the liquid phase and cost of phosphorus recovery.

the waste stream decreases (Fig. 3). The $\text{PO}_4\text{-P}$ content of 50 mg/L has been suggested in the literature as the threshold for considering phosphorus recovery (Cordell et al., 2009). In practice, economic viability of phosphorus recovery depends on many factors and will increase significantly as the $\text{PO}_4\text{-P}$ content increase beyond 50 mg/L. At the $\text{PO}_4\text{-P}$ content below 50 mg/L, indirect phosphorus recovery may still be possible using wetland and microalgae cultivation for phosphorus uptake and subsequent utilisation. These indirect phosphorus recovery processes are primarily to polish and remove phosphorus from environmental water to avoid eutrophication.

Phosphorus recovery results in both financial and environmental benefits. Revenue from recovered phosphorus products (e.g. struvite and calcium phosphate as fertilisers and elementary phosphorus for the high-tech industries) can compensate for recovery cost. It has been established that the demand (therefore cost) for chemical (e.g. calcium and magnesium) addition per unit of recovered phosphorus is inversely proportional to phosphorus content in the waste stream. At a low level of phosphorus, the application of wetland, microalgae cultivation, and adsorption technologies is less cost-effective given the large footprint and water requirements and low value of products obtained after treatment (Bunce et al., 2018). In these cases, environmental benefits can justify for phosphorus recovery and removal. For example,

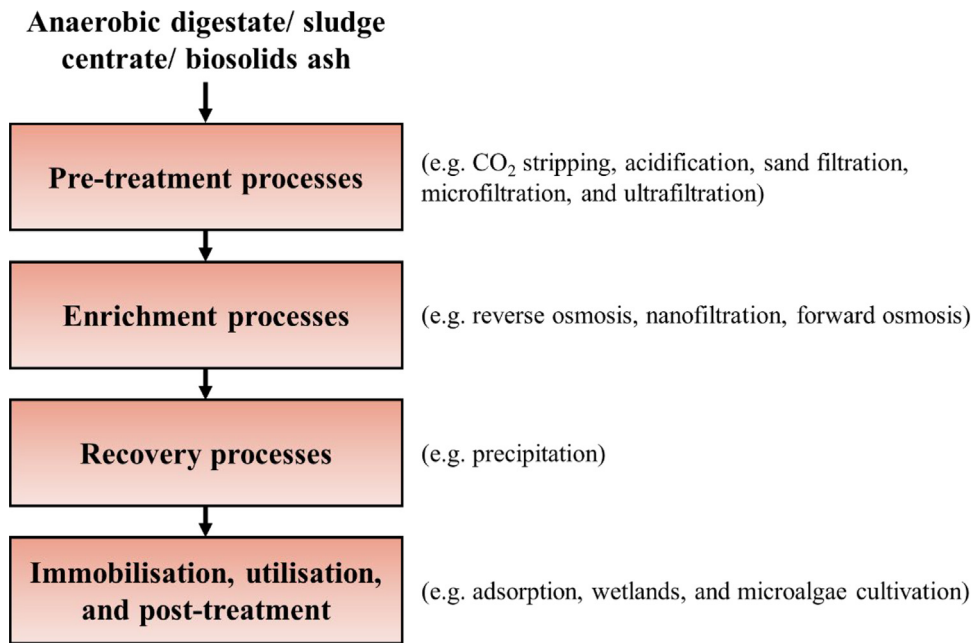


Fig. 4. Schematic diagram showing major steps in phosphorus recovery.

water quality benefits from phosphorus removal is estimated at 9000 \$AUD/kg by the Department of Environment and Conservation NSW (New South Wales et al., 2006).

3.2. Phosphorus removal and recovery from waste and wastewater

3.2.1. Major processes for phosphorus recovery

In the current practice, phosphorus is recovered from nutrient-rich streams released from anaerobic digestion of various feedstock including sewage sludge, food waste, agricultural feedstock, and wastewater from animal farming (Vu et al., 2022a; Salkunić et al., 2022; Ganesapillai et al., 2021). In addition to biogas production, the anaerobic digestion process also converts organic phosphorus to an inorganic form (i.e. PO_4^{3-}) that is soluble in the aqueous phase. The products of the anaerobic digestion that could be used as the input materials for the phosphorus recovery unit involve anaerobic digestate (digested sludge), sludge centrate (the liquid fraction of anaerobic digestate after dewatering), and biosolids (the solid fraction of anaerobic digestate after dewatering) (Salkunić et al., 2022; Saerens et al., 2021). Phosphorus can also be recovered from biosolids after incineration. In this process, organic residues in biosolids are incinerated to produce biosolids ash, which is rich in phosphorus and other minerals (Bagheri et al., 2022). Biosolids ash is then acidified to release phosphate to the aqueous phase in the form of phosphate for subsequent recovery (Bagheri et al., 2022; Muller et al., 2007).

Phosphorus recovery is usually accomplished in four sequential steps, namely pre-treatment, enrichment, recovery (i.e. extraction), and post-treatment (Fig. 4). Pre-treatment is achieved via CO₂ stripping, acidification, sand filtration, microfiltration (MF), and ultrafiltration (UF), or a combination of these processes. The selection of pre-treatment techniques is dependent on the feedstock for the recovery process. For example, CO₂ stripping has been applied to adjust the pH of anaerobic digestate for phosphorus recovery in full-scale NuReSys[®] and AirPrex[®] processes (Saerens et al., 2021; Melgaço et al., 2021). In addition, acidification is used to pre-treat biosolids ash to transfer phosphorus from solid to aqueous phase as well as to remove impurities for subsequent enrichment and recovery (Bagheri et al., 2022; Muller et al., 2007). Sand filtration, MF, or UF can be used to remove suspended solids from sludge centrate to avoid interference with the later process of phosphate enrichment and precipitation. Enrichment is an optional step to increase the phosphorus content in the liquid phase and reduce the demand (thus costs) of chemical addition for recovery by precipitation. High rejection membrane processes such as reverse osmosis (RO), nanofiltration (NF), and forward osmosis (FO) have been considered for pre-concentrating phosphorus prior to recovery (Xie et al., 2016). After the enrichment process, phosphorus is recovered using phosphate precipitation in the form of struvite or calcium phosphate. Finally, post treatment is applied to the spent solution to remove the residual phosphorus prior to effluent discharge. As discussed in Section 3.1, the use of adsorption and biological processes (i.e. wetlands and microalgae cultivation) to immobilise phosphorus for beneficial utilisation is preferred at a low level of phosphorus.

3.2.2. Pre-treatment

Anaerobic digestate has high alkalinity. Thus, CO₂ stripping is sometimes required to adjust the solution pH for subsequent phosphorus recovery. CO₂ stripping has been applied as a pre-treatment step at several full-scale nutrient recovery plants (Yesigat et al., 2022). Anaerobic digestate or sludge centrate is aerated to remove dissolved CO₂ out of the solution, thereby increasing the solution pH and facilitating the subsequent recovery process via precipitation.

Acidification is used as a pre-treatment method to liberate more phosphorus from anaerobic digestate or to dissolve phosphorus of biosolids ash into the solution in order to facilitate the subsequent recovery process. In the Seaborne process at the WWTP Gifhorn, acid sulphuric has been added to anaerobic digestate to solubilise particulate phosphorus and release it in the form of phosphate for subsequent recovery by chemical precipitation (Muller et al., 2007). The acidified stream is then centrifuged to obtain the nutrient-rich liquid fraction for the recovery process via struvite or calcium phosphate precipitation (Muller et al., 2007). Similar to anaerobic digestate, phosphorus in biosolids ash is extracted from solid to liquid phase using acid sulphuric and subsequently recovered from the solution using the precipitation process (Bagheri et al., 2022; Muller et al., 2007).

Sand filtration and MF/UF pre-treatment are used to avoid interference with phosphorus precipitation and prevent membrane fouling during the enrichment process. Pre-treatment by sand filtration and MF can effectively remove suspended solids, thereby mitigating the deposition of these foulants on the membrane surface. Vu et al. (2022a) demonstrated that pre-treatment by sand filtration could reduce the total suspended solid of sludge centrate by eightfold, leading to mitigated membrane fouling and reduced nutrient loss during subsequent FO enrichment.

3.2.3. Phosphorus enrichment

As discussed in Section 3.2.1, the cost of phosphorus recovery decreases as its content in the waste stream increases. Not surprisingly, significant research effort has been devoted toward enrichment by membrane separation, especially RO, NF, and FO. NF can achieve high retention of phosphorus (over 90%) since it exists primarily in the multivalent anionic form of PO₄³⁻ (Barampouti et al., 2020; Thong et al., 2016). Complete or near complete retention (99%–100%) of phosphorus by RO has been reported in the literature (Barampouti et al., 2020). While NF/RO membranes can effectively retain phosphorus for subsequent recovery, they suffer from serious fouling because most feed water is complex and has high fouling propensity. Thus, pressure-driven membrane processes have not been commercially used for enriching phosphorus from highly complex wastewater (e.g. anaerobic digestate and sludge centrate) (Xie et al., 2016). Previous research has shown severe and irreversible fouling when NF/RO membranes are used for phosphorus enrichment (Souza-Chaves et al., 2022; Tian et al., 2023). Significant research effort has been given toward the development of new membrane materials with antifouling or self-cleansing properties as well as new membrane processes. Several techniques to regulate membrane chemistry and surface structure have been explored to reduce fouling and improve selectivity (Guo et al., 2022). Examples include the inclusion of nanoparticles and carbon nanotubes as interplay (Long et al., 2022; Al Ansari et al., 2022) to achieve uniform flux distribution and optimised surface energy (hydrophilicity) for fouling reduction and micro-patterning the membrane surface (Xiao et al., 2019) to create a non-adhesive condition. These techniques are promising, but to date, have only been demonstrated at laboratory scale.

Recent years have seen surging research and advancements in FO, which is a well-established membrane separation process but with potentially new applications for dewatering complex industrial streams such as drilling mud, landfill leachate, fermentation broth, and wastewater sludge. FO is an osmotically driven membrane process with low fouling propensity. Pilot scale demonstrations of FO for dewatering drilling mud and landfill leachate have been reported in peer-reviewed and grey literature (Coday et al., 2014). The potential of FO to enrich phosphorus in sludge centrate has also been reported although mostly at laboratory scale (Jiang et al., 2022; Vu et al., 2021b, 2019; Ansari et al., 2016; Vu et al., 2018).

In the FO process, only water is transported through a semipermeable membrane from a feed solution (e.g. sludge centrate) to a draw solution (i.e. a high salinity solution) due to the difference in osmotic pressure of these two solutions, which leads to increased nutrient contents in sludge centrate (Fig. 5). Unlike the pressure-driven membrane processes, the fouling layer on the FO membrane surface is not compacted, thus, is readily reversible by hydraulic flushing or osmotic backwashing (Vu et al., 2019; Holloway et al., 2007). Several studies demonstrated the feasibility of using FO to concentrate sludge centrate with high enrichment factors (Vu et al., 2022a, 2021b; Xie et al., 2014). Xie et al. (2014) demonstrated five times pre-concentration of sludge centrate using FO driven by MgCl₂ draw solution.

Phosphorus enrichment by membrane separation is a current and active research area with several possible breakthroughs. When one of these breakthroughs is realised, a step decrease in the cost of phosphorus production can be expected to significantly increase the scale and economy of phosphorus recovery from waste and wastewater.

3.2.4. Phosphorus recovery

Chemical precipitation is a core component of phosphorus recovery technology. Indeed, chemical precipitation is used in most commercially available technologies for phosphorus recovery from the liquid phase (e.g. anaerobic digestate, sludge centrate, and acidified biosolids ash) (Table 1). Phosphorus can be precipitated for separation from the aqueous phase in the form of calcium phosphate or struvite. In commercially available technologies, phosphorous precipitation is achieved by either continuously mixed tank (e.g. NuReSys[®] process) (Fig. 6A) (Moerman et al., 2009) or fluidised bed reactor (e.g. Pearl[™] process) (Fig. 6B) (Gysin et al., 2018). The precipitated phosphorus can be readily settled for separation

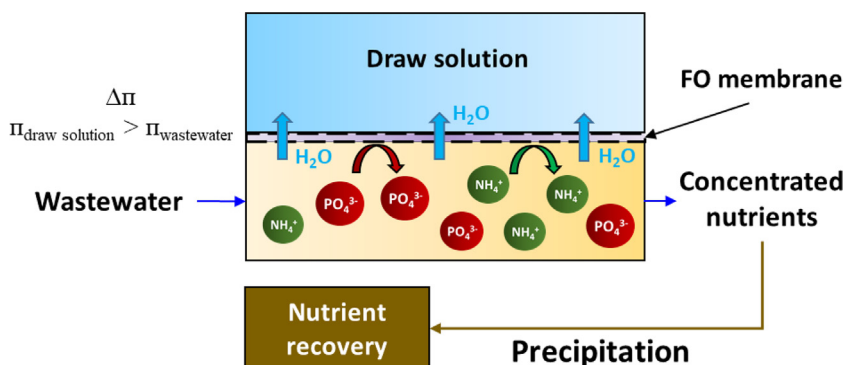


Fig. 5. The schematic diagram of the FO-based system for phosphorus recovery.

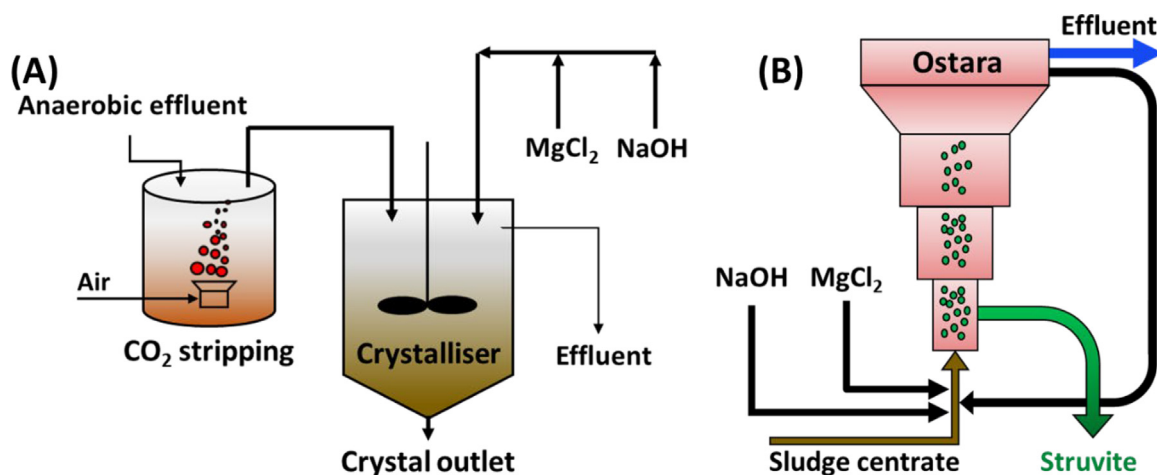
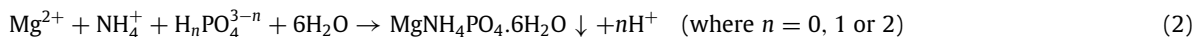


Fig. 6. Schematic diagram of (A) the NuReSys[®] and (B) the Pearl[™] phosphorus recovery processes.

from the remaining liquid. These precipitates can be used directly as fertilisers or further purified for other industrial applications. The formation of phosphorous precipitates can be expressed in Eqs. (1) and (2).



To facilitate struvite precipitation, the addition of magnesium and caustic soda into phosphorus-rich streams is required for the occurrence of the reaction. The optimum pH for struvite precipitation is in the range of 8.5–9.5 (Daneshgar et al., 2018; Günther et al., 2018; Shi et al., 2018). Due to the release of protons during the struvite formation (Eq. (2)), a supply of alkali is needed to maintain the pH in a suitable range for struvite precipitation. In addition to pH, the molar ratio of magnesium to ammonium to phosphate significantly affects the struvite formation efficiency and its purity. At the optimum magnesium to ammonium to phosphate ratio of 1.6:0.6:1.0, between 85%–97% of phosphorus can be achieved via struvite precipitation (Sengupta et al., 2015). In sludge centrate, ammonium and phosphate are abundant while magnesium content is insufficient to ensure an effective reaction (Kataki et al., 2016). For this reason, a magnesium source in the form of MgCl_2 , MgSO_4 , MgO , or $\text{Mg}(\text{OH})_2$ is added into anaerobic digestate (Lee et al., 2003; Quintana et al., 2005). As a high cost commodity, magnesium addition can contribute up to 75% of overall production costs of struvite from waste and wastewater (Barampouti et al., 2020). Several studies have explored the use of low-grade thus cheaper magnesium sources such as seawater and bittern to reduce the cost of phosphorus recovery via struvite precipitation (Kataki et al., 2016; Lee et al., 2003).

Phosphorus recovery in the form of calcium phosphate is an alternative to struvite (Barampouti et al., 2020; Song et al., 2002; Tran et al., 2014; Yeoman et al., 1988). Instead of adding magnesium to form struvite, calcium (which is a much cheaper chemical) is added to induce calcium phosphate formation. The pH range of 8–11 is most favourable for the calcium phosphate precipitation (Yeoman et al., 1988). CO_2 -stripping may be performed prior to precipitation to prevent the competing formation of calcium carbonate. Phosphorus recovery efficiency of 50%–90% can be achieved via calcium phosphate (Lebuf et al., 2017).

Table 1

Current commercial technologies for phosphorus recovery from different types of waste and wastewater (Note: NA = Not available or Not applicable).

Input materials	Name	Devel- oper/Country of origin	Recovery mechanism	Min influent PO ₄ -P (mg/L)	Production capacity (t/yr)	Product
Liquid fraction	Ostara Pearl	Ostara, Canada	Precipitation/ crystallisation	100	>1,000	Struvite
Liquid fraction	MagPrex™	Centrisys/CNP, USA	Precipitation/ crystallisation	NA	NA	Struvite
Liquid fraction	Crystalactor™	Royal Haskoning DHV, Netherland	Precipitation/ crystallisation	25	NA	CaP
Liquid fraction	Phosnix	Unitikia Ltd., Japan	Precipitation/ crystallisation	NA	NA	Struvite
Liquid fraction	Seaborne Gifhorn	Seaborne Laboratory, Germany	Precipitation/ crystallisation	600	NA	Struvite
Liquid fraction	Struvia	Veolia Water	Precipitation/ crystallisation	50	40	Struvite
Liquid fraction/ digestate	ANPHOS [®]	Colsen, Netherland	Precipitation/ crystallisation	50	650	Struvite
Liquid fraction/ digestate	PHOSPAQ™	Paques, Netherland	Precipitation/ crystallisation	50	>1,000	Struvite
Liquid fraction/ digestate	NuReSys [®]	Nutrient Recovery Systems, Belgium	Precipitation/ crystallisation	NA	NA	Struvite
Sludge ash	Ash Dec [®]	Ash Dec (Outotec), Austria	Thermal P recovery	NA	NA	Calcinated P fertiliser Phoskraft [®]

Unlike phosphorus recovery from the liquid phase via precipitation, phosphorus in biosolids ash could be recovered by melting biosolids ash via a dry thermal process (Desmidt et al., 2015). To date, there have been only two thermal processes applied to recover phosphorus at full-scale in Netherland and Austria (Hermann and Schaaf, 2019; Sartorius et al., 2012) (Table 1). The first process called ASH DEC[®] recovers phosphorus under the form of calcinate phosphates (Hermann and Schaaf, 2019). In this process, biosolids ash is mixed with solid chlorine donors (MgCl₂ and CaCl₂) and then combusted at 1000 °C in 30 min to vent out most heavy metals (Desmidt et al., 2015; Hermann and Schaaf, 2019). Under these extreme conditions, new phosphate mineral phases (i.e. calcinate phosphates), which is significantly phosphorus-bioavailable, are formed. These substances could be immediately used as fertilisers.

3.2.5. Immobilisation, utilisation, and post treatment

After the recovery process, the post treatment is required to quench the residual phosphorus (i.e. below 50 mg/L) from the exhausted solution before effluent discharge. In a circular economy, it is desirable to recover phosphorus for beneficial use and utilisation of phosphorus-contained products after the post-treatment. At a low level of phosphorus in the post treatment effluent, the treatment technologies using adsorption and biological processes (e.g. wetlands and microalgae cultivation) are preferred in consideration with the economic viability.

Adsorption has been widely used to capture phosphorus from wastewater at the low level. This method is based on the ion selective retention ability of active sites of sorbents (e.g. biochars (Wang et al., 2020; Yang et al., 2019), steel-making slag (Vu et al., 2021a), red mud (Yin et al., 2022), and activated carbon (Almanassra et al., 2021)). After the adsorption, these sorbents can be either used directly as fertilisers or reused for other applications, thus phosphorus recovery. Biochars produced by thermal treatment of organic waste materials (e.g. coconut shell, rice straw, bamboo wood, corn stalk, and iron-rich sludge) could be used to capture phosphorus in an aqueous solution (Günther et al., 2018; Wang et al., 2020; Yang et al., 2019). The phosphorus capturing capacity of biochars results from the presence of Mg, Ca, and Fe in their composition (Günther et al., 2018; Wang et al., 2020). The phosphorus-captured biochars could be composted and then used as fertilisers, thus phosphorus recovery. In addition to biochars, steel-making slag (a waste product from the steel making industry) has demonstrated its effectiveness in removing phosphorus from an aqueous solution (Vu et al., 2021a; Jonidi Jafari and Moslemzadeh, 2020; Maharaj et al., 2017). The presence of metal oxides (e.g. CaO, Fe₂O₃, Al₂O₃, and SiO₂) in the steel-making slag composition make it an ideal adsorbent for phosphorus immobilisation (Vu et al., 2021a; Lim et al., 2016). Here, chemical precipitation is also the underlying mechanism for phosphorus immobilisation onto the

slag (adsorbent), although the objective is to solely remove rather than recover phosphorus. The weathered and stabilised steel-making slag after phosphorus removal could be used as an aggregate for asphalt roads (Maharaj et al., 2017).

The use of microalgae to uptake phosphorus for biomass production could be a cost-effective and environmentally friendly approach for nutrient recovery from wastewater at a low level. Microalgae are photoautotrophic microorganisms that can be capable of using inorganic carbon (i.e. CO₂), light energy, and nutrients (e.g. nitrogen and phosphorus) to produce biomass via its metabolism. Microalgae biomass is then harvested, whereby phosphorus is extracted from an aqueous solution. Microalgae biomass can be directly applied to the field or used as feedstock for other processes (e.g. biochemical and biofuel productions) (Selvaraj et al., 2021). The abundance of nutrients in sludge centrate is beneficial for microalgae cultivation. Once the nutrient usage alone accounts for 50% total cost of microalgae cultivation, using sludge centrate as an alternative for nutrient supply is expected to reduce significantly the cost of cultivation (Selvaraj et al., 2021). The feasibility of using microalgae to remove phosphorus and produce algal biomass from wastewater has been demonstrated in lab- and pilot-scale studies (Aditya et al., 2022; Nguyen et al., 2022; Vu et al., 2020b). For example, it is reported that approximately 90% of phosphorus could be removed and recovered via algal biomass production when multi-culture of *Chlorellaceae*, *Scenedesmeceae*, *Chlamydomonadaceae* is used to treat piggery farm wastewater in a pilot-scale high rate algal pond.

Constructed wetland can also immobilise phosphorus through a combination of physical, chemical, and biological processes (Kill et al., 2022; Li et al., 2021a; Ung et al., 2022). These processes involve sedimentation, photolysis, hydrolysis, microbial degradation, adsorption, and plant uptake. It is difficult to separate the phosphorus removal efficiency of the individual process as constructed wetland is a complex ecology. The presence of different types of substrates (e.g. biochar, fly ash, steel-making slag, and alum sludge) in constructed wetlands allows for phosphorus removal mostly via adsorption (Ji et al., 2022). The main biological processes for phosphorus removal are uptake (or assimilation) by plants, algae, and bacteria and transformation processes conducted by microbes (Li et al., 2021b). The effectiveness of using constructed wetlands in phosphorus removal has been demonstrated at commercial scales (Bunce et al., 2018; Perdana et al., 2020). A full-scale constructed wetland treating wastewater in Indonesia has shown 90% phosphate removal (Perdana et al., 2020).

4. Future outlook

Chemical precipitation as the core process of phosphorus recovery has been fully developed and used in commercially available systems. In these systems, phosphorus is recovered in the form of struvite or calcium phosphate. Due to low content of alkaline metals in wastewater, additional calcium or magnesium is required to allow for phosphate precipitation. It is a necessary step but will also increase the cost of phosphorus recovery. The economics of phosphorus recovery can be significantly enhanced by enriching phosphorus content in wastewater prior to the recovery process. Yet, technologies for phosphorus enrichment using RO, NF, and FO have been still mostly in the research and development phase. Moreover, these membrane-based enrichment processes are more likely susceptible to severe membrane fouling as they are applied to concentrate the nutrient-rich waste streams, which are usually highly complex. Therefore, the exploration of effective techniques to control membrane fouling during the enrichment process plays an important role in developing a complete and efficient solution for nutrient recovery. Further work on phosphorus enrichment at pilot- and full-scale levels prior to the recovery process is also needed to demonstrate its scalability and techno-economic feasibility.

Prior to the enrichment process, the pre-treatment process has been also limited. This study points out that most of the conventional pre-treatment techniques (e.g. CO₂ stripping and acidification) prior to phosphorus recovery via chemical precipitation are mature as they have been applied on a full scale. The new pre-treatment techniques using sand filtration and MF/UF are still in an early stage of commercialisation. Hence, further research on optimising the system operational parameters, such as system configurations, long-term operation head loss, and fouling resistance is critical to demonstrate their applicability. The implementation of these methods in the pilot- and full-scale should be considered to demonstrate their robustness and techno-economic feasibility.

In addition to the pre-treatment, post-treatment of the recovery process effluent has not received sufficient attention. The effluent of the recovery process via chemical precipitation is still abundant in nutrients, the post-treatment is thus essential. The current techniques using adsorption, microalgae cultivation, and constructed wetland have demonstrated their robustness in removing phosphorus but only from wastewater in general. Further research on their application to post-treat the actual recovery process effluent is necessary to complement the recovery process via chemical precipitation.

5. Conclusion

This review provided a comprehensive and systematic analysis of phosphorus recovery from waste and wastewater under the perspective of the circular economy. Data and information corroborated in this review showed that without effort to recover phosphorus at a large scale, the depletion of this essential element will occur in the coming decades. Phosphorus recovery has already been achieved at scale from nutrient-rich streams from anaerobic digestion of organic feedstock (e.g. human and animal wastes). A practical hierarchy for sustainable phosphorus preservation from immobilisation to recovery with respect to the threshold of phosphorus level (i.e. 50 mg/L) in wastewater was systematically analysed and discussed in this review. Data and information corroborated from this review were used to propose a

roadmap for improving the economic viability and scaling up phosphorus recovery. It is anticipated that future phosphorus recovery plants will include the core step of phosphate recovery by precipitation/crystallisation as well as complementary processes such as pre-treatment, enrichment, and post-treatment of the spent liquid stream. Further work on phosphorus enrichment at pilot- and full-scale would therefore be essential for demonstrating the scalability and techno-economic feasibility of phosphorus recovery from waste and wastewater.

CRedit authorship contribution statement

Minh T. Vu: Data curation, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft, Writing – review & editing. **Hung C. Duong:** Data curation, Visualization, Writing – review & editing. **Qilin Wang:** Validation, Visualization, Writing – review & editing. **Ashley Ansari:** Data curation, Methodology, Validation, Writing – review & editing. **Zhengqing Cai:** Validation, Visualization, Writing – review & editing. **Ngoc Bich Hoang:** Validation, Visualization, Writing – review & editing. **Long D. Nghiem:** Conceptualization, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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