



Review

A review on the integration of mainstream P-recovery strategies with enhanced biological phosphorus removal

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ABSTRACT

Phosphorus (P), an essential nutrient for all organisms, urgently needs to be recovered due to the increasing demand and scarcity of this natural resource. Recovering P from wastewater is a feasible and promising way widely studied nowadays due to the need to remove P in wastewater treatment plants (WWTPs). When enhanced biological P removal (EBPR) is implemented, an innovative option is to recover P from the supernatant streams obtained in the mainstream water line, and then combine it with liquor-crystallisation recovery processes, being the final recovered product struvite, vivianite or hydroxyapatite. The basic idea of these mainstream P-recovery strategies is to take advantage of the ability of polyphosphate accumulating organisms (PAO) to increase P concentration under anaerobic conditions when some carbon source is available. This work shows the mainstream P-recovery technologies reported so far, both in continuous and sequenced batch reactors (SBR) based configurations. The amount of extraction, as a key parameter to balance the recovery efficiency and the maintenance of the EBPR of the system, should be the first design criterion. The maximum value of P-recovery efficiency for long-term operation with an adequate extraction ratio would be around 60%. Other relevant factors (e.g. COD/P ratio of the influent, need for an additional carbon source) and operational parameters (e.g. aeration, SRT, HRT) are also reported and discussed.

1. Introduction

The new paradigm in wastewater treatment proclaims that wastewater treatment plants (WWTPs) should evolve to water resource recovery facilities (WRRFs), where resources and energy are recovered from water. Amongst all the potential resources to be recovered, P arises as a perfect candidate since: i) in the current context of population and food demand growth, P is essential for fertilizers production (Chowdhury et al., 2017; Cieřlik and Konieczka, 2017; Roy, 2017), ii) P is currently a pollutant of our wastewater which may promote eutrophication in water bodies if it is not removed and iii) P is mainly obtained from non-renewable sources such as phosphate rocks (Van Vuuren et al., 2010), which are expected to be depleted in only 50–100 years (Cordell et al., 2009; Desmidt et al., 2015; Rittmann et al., 2011). For the full-scale application of P-recovery processes, one of the main current limitations is that the market price (Table 1) of rock phosphate is lower than recovered products such as struvite. However, if the additional benefits of P-recovery are accounted for, significant incentives to support it emerge: for example, the need to treat the impurities in phosphate

rock (e.g. cadmium or uranium), which can be highly toxic (Cornel and Schaum, 2009), or to avoid the need for external dependence on a strategic resource such as phosphate rock. In addition, P in urban wastewaters has to be removed anyway to avoid eutrophication processes, making P-recovery a win-win scenario.

WWTPs seem to be an adequate place to recover P, since 1.3 Mt of P are removed annually worldwide through wastewater treatment. Some estimates indicate that global demand for P could be covered by 15–20% with this recovery (Li and Li, 2017; Wu et al., 2019; Yuan et al., 2012). Other studies indicate that in Central Europe, P-recovery from municipal wastewater would satisfy 40–50% of the mineral P fertilizer applied annually in agriculture (Egle et al., 2016).

Furthermore, from a life cycle assessment (LCA) point of view, it has been concluded that P-recovery from municipal wastewater sludge, liquor or ash of sludge mono-incineration would provide environmental benefits (Remy and Jossa, 2015). Although differences appear in the amount of P recovered, requirements of energy, chemicals and fuels, or in side effects in the process, P-recovery processes can reduce environmental impacts, mainly on global warming potential, freshwater

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Table 1
The market values of P rock and P-recovery products.

P related species	Market price	Reference
P rock	~120 \$/ton	(Mew, 2016)
Struvite	~500 €/ton	(Wu et al., 2019)
Vivianite	~10,000 €/ton	(Wu et al., 2019)
Hydroxyapatite	~10,000 \$/ton	(Annisa et al., 2021)

eutrophication of and fossil energy demand (Remy and Jossa, 2015). P-recovery as struvite seems to be especially attractive for urban areas, to satisfy the growing trend of local crop production linked to the large volumes of wastewater treated in centralized WWTPs (Ruff-Salis et al., 2020). In this recent LCA study, three recovery technologies were analysed for large WWTPs, showing that were able to recover 5–30 times the amount of P required to fertilize the agricultural area of the studied region annually (36.5 t). Although there was an increase of chemicals and energy requirements per m³ of wastewater due to struvite recovery, it resulted in lower eutrophication potential.

1.1. Fate of P in WWTPs

The fate of the influent P in a WWTP depends on its configuration. Part of the influent P is removed with the primary sludge (around 11%) according to Cornel and Schaum (2009). If the plant does not implement enhanced biological phosphorus removal (EBPR) in the secondary step, approximately 15–30% of the P in the influent becomes part of the biomass (a conventional WWTP sludge contains around 1–2% of P according to mass basis) and removed with the purged sludge (Henze et al., 2008; Parsons and Smith, 2008). Then, if complete P-removal is mandatory, the remaining P should be chemically removed with addition of Fe(III) and Al(III) salts in different dosing points such as the primary sedimentation, before and/or following biological treatment or in a tertiary treatment (Tchobanoglous et al., 2014). However, the chemical sludge produced is not recommended as fertiliser since these salts could reduce the mobility of P in soils where this chemical sludge is applied (Desmidt et al., 2015; Sartorius et al., 2012). The sludge generated is often accumulated in landfills or burnt in incinerators, and may end up contaminating water bodies (such as aquifers, rivers or sea) (Mainstone et al., 2000). Alternatively, the use of ashes from sewage sludge incinerators is proposed for P-recovery (Ma and Rosen, 2021) and to produce fertilizers via a chemical extraction (Fang et al., 2021; J. Li et al., 2018).

The fate of P in WWTPs that include EBPR is very different. EBPR is based on the proliferation of polyphosphate accumulating organisms (PAO), which can accumulate phosphate internally as polyphosphate (poly-P) under aerobic (or anoxic) conditions after being exposed to anaerobic conditions with available carbon source. Then, P is removed as poly-P inside cell biomass by waste sludge (Tchobanoglous et al., 2014). PAO can incorporate up to 0.38 mg P/mg VSS compared to the 0.02 mg P/mg VSS incorporated by the ordinary heterotrophic organisms (OHO) (Gebremariam et al., 2011; Henze et al., 2008; Yuan et al., 2012). EBPR-based WWTPs can typically remove more than 85% of the P from the influent that is stored within the biomass (Bunce et al., 2018; Gebremariam et al., 2011). The reported range of the P content in EBPR-based sludge is around 0.06–0.15 mg P/mg VSS (Henze et al., 2008). However, the use of this sewage sludge in agriculture is nowadays limited in many areas because of the ecological and health threats involved. For example, it is estimated that only about 20% of the sewage sludge meets the EU regulations for fertilizers in terms of concentration of heavy metals or radioactive elements (e.g. radon and uranium) (European Commission, 2019; Pettersson et al., 2008; Weigand et al., 2013).

There is another reason to implement P-recovery strategies. In EBPR-based WWTPs, the clogging of tubes and pumps due to undesired struvite precipitation is observed when the bio-P sludge undergoes

anaerobic digestion and P-recovery is not implemented (D.Wild et al., 1996; Doyle and Parsons, 2002). The repair of this clogging is costly due to the need to shut down and clean the clogged equipment. In a modelling study, Lizarralde et al. (2019) constructed a plant-wide model to understand the effect of several operational parameters on the uncontrolled struvite precipitation when EBPR was implemented. They concluded that struvite recovery has many benefits for the plant not only for its commercial value, but also for the reduction of sludge production, ferric chloride dosage and the prevention of uncontrolled struvite precipitation.

1.2. Most studied technologies for P-recovery in WWTPs

Different technologies have been proposed for P-recovery from flows containing phosphorus in WWTP (i.e. effluent, sewage sludge, sludge ash, dewatering liquor and digester supernatant), providing different operability, costs and potential uses of the recovered material (Cieřlik and Konieczka, 2017; Desmidt et al., 2015; Egle et al., 2016; Law and Pagilla, 2018). Egle et al. (2016) indicated that an ideal technology should provide good P-removal from wastewater, maximize P-recovery rate, destroy hazardous substances and produce a material that could be applied with low environmental risks, good fertilizing effects and high economic efficiency. They also studied all these characteristics from 19 different P-recovery technologies and concluded that the choice of a specific recovery technology is a compromise amongst these factors. Furthermore, the LCA analysis by Amann et al. (2018) showed wide range of changes in gaseous emissions, energy demand and P-recovery potential of different technologies based on liquid phase, sewage sludge and ash, and concluded that LCA should be combined with additional environmental criteria to put the performance of the technologies into perspective.

Considering the wide range of existing P-recovery processes, this section presents the most studied ones classified in two groups, depending on whether they can be obtained from primary/secondary effluents or from the sludge line, and includes another subsection showing the crystallisation processes that could be applied to P-enriched streams to make P-recovery effective. Section 2 then presents the P-recovery processes related to mainstream strategies that are the focus of this review.

1.2.1. P-recovery from primary or secondary effluents

P-recovery from primary or secondary effluents using chemical precipitation does not seem to be a cost-effective alternative because it would require a large amount of chemicals (Cornel and Schaum, 2009; Desmidt et al., 2015; Egle et al., 2016). Several alternative methodologies have been described previously (Egle et al., 2015; Kabdařlı and Tünay, 2018). For instance, Liberti et al. (1979) reported a methodology based on the combination of ion exchange (IX) for phosphate selective exchange and P-recovery from the concentrated stream by struvite precipitation, which was further evaluated in subsequent works (Liberti et al., 2001; Petruzzelli et al., 2003). Williams et al. (2015) studied P and N removal with media based on Fe, Cu, and Al and then precipitation columns with clinoptilolite IX. They achieved almost 84% of P-recovery and a regeneration eluate with concentration up to 560 mg P-L⁻¹. Beaudry and Sengupta (2021) used a novel pyridine-based polymeric ion exchanger and three different fixed-bed columns, producing an effluent concentration with P < 6 µg/L and recovering about 90% of the phosphorus.

The hybrid anion exchanger (HAIX), which consists of a hybrid polymeric base dispersed with hydrated ferric oxide (HFO) nanoparticles, has also been reported for P removal (Martin et al., 2018, 2009; Sendrowski and Boyer, 2013). Phosphate uptake is based on the selective sorption sites with HFO nanoparticles, which form sphere complexes with Fe(III) and phosphate (Blaney et al., 2007). After long-term sorption, phosphate breakthrough occurs, but the exhausted HAIX can be reused with a regeneration phase with NaOH/NaCl solution

and, finally, a rinse with CO₂ sparged filtered water (Blaney et al., 2007). A recent work by Guida et al. (2021) evaluated a large (10 m³/d) HAX system for 2.5 years using treated municipal wastewater, showing high P removal (95%, i.e. from 6 to <0.3 mg PO₄³⁻-P/L) and recovery efficiency (95% of phosphate from the wastewater recovered as hydroxyapatite). In subsequent work, the regeneration of the media and the modelling of this system were also studied (Pinelli et al., 2022). However, the feasibility of this methodology at full-scale has not been proved yet (Egle et al., 2015; Ruff-Salis et al., 2020; Siciliano et al., 2020).

Apart from the above-mentioned approach, microalgae cultivation has been proposed as a promising technology for P-recovery from secondary effluents due to its low cost, eco-friendliness and low technology dependence (Yang et al., 2017). P is taken up biologically by algal growth by assimilation, accompanied by oxygen generation that supports aerobic bacteria growth. Algae can also be used either as a feedstock for biofuel production or directly as a biofertilizer (Chisti, 2013, 2008). El Hamouri (2009) showed that 63% of influent P could be removed and incorporated into algae as a tertiary treatment using a high-rate algal pond. Xu et al. (2016, 2015) also showed that algae could remove about 60% of influent phosphorus for secondary wastewater effluent. However, the application of algae for P-recovery is still limited in large-scale wastewater treatment (Gebremariam et al., 2011).

1.2.2. P-recovery from the sludge line

As mentioned above, the maximum P-recovery potential is from EBPR-based sludge. However, this option is not straightforward. For instance, the agricultural application of stabilized sludge with high P content (Hester and Harrison, 2013) has to consider the strict national health and safety restrictions on nutrients and heavy metals. This hinders the agricultural application of sludge and, thus, this pathway of P-recovery (Law and Pagilla, 2018). Another option is the incinerated sludge ashes, which may contain 4 to 11% of P (Biswas et al., 2009; Donatello et al., 2010; Franz, 2008; Ottosen et al., 2013). However, as a result of the organic matter oxidation, the heavy metal content in these ashes may be even higher than the sludge itself. Therefore, a chemical (Franz, 2008; Ottosen et al., 2013) or thermochemical (Adam et al., 2007) process is needed to separate the P from the heavy metals of the ashes. Wet chemical treatment by acid, alkaline or both have been proposed for the dissolution of P (Law and Pagilla, 2018); acid leaching (sulphuric or nitric acid) is proposed for Fe-rich ashes (Ottosen et al., 2013), while the combination of both acid and alkaline treatment is proposed for Al-rich ashes (Petzet et al., 2012).

The most reported location to recover P is a liquid stream from the sludge line, either from the sludge treatment or the anaerobic digestion (Egle et al., 2015). The treatment of the purged secondary sludge releases soluble phosphate and ammonium, resulting in a supernatant with increased concentrations of these compounds. These streams can contain up to 30% of the influent P loads. Levlin and Hultman (2003) reviewed several side-stream based P-recovery strategies and concluded that P-recovery efficiency about 60–65% is achieved from these streams.

1.2.3. Crystallization processes for P-recovery

Crystallization is nowadays the most accepted strategy for recovering P from these enriched liquors, being struvite the preferred precipitation product (Le Corre et al., 2009; Li et al., 2019; Rittmann et al., 2011). Struvite (MgNH₄PO₄·6H₂O) can be applied directly to the field as a slow release fertiliser, also providing nitrogen (N) and magnesium (Mg) to the soil, and has been reported to be a good option for agricultural uses (Hu et al., 2016). Sometimes, ammonium in struvite can be replaced by potassium and then the precipitate is known as K-struvite (Rontelap et al., 2007). The reactions to precipitate struvite and K-struvite and their corresponding solubility are presented in Table 2.

Fig. 1 presents a graphical representation of the minimum magnesium (Mg²⁺) concentration needed to reach the struvite solubility product at different pH as a function of phosphate and ammonium concentration. As observed, significant concentrations of P, Mg²⁺, NH₄⁺

Table 2
Stoichiometry and solubility for the most usual P-recovery chemical compounds.

Compound	Stoichiometry	pKsp	Reference
struvite	Mg ²⁺ + NH ₄ ⁺ + PO ₄ ³⁻ + 6 H ₂ O →	13.26	(Ohlinger et al., 1998)
K-struvite	Mg ²⁺ + K ⁺ + PO ₄ ³⁻ + 6 H ₂ O →	11.7	(Luff and Reed, 1980)
vivianite	3 Fe ²⁺ + 2 PO ₄ ³⁻ + 8 H ₂ O →	29.03	(Chen and Faust, 1974)
hydroxyapatite	5 Ca ²⁺ + 3 PO ₄ ³⁻ + OH ⁻ →	58.5	(Mcdowell et al., 1977)

and high pH (around 9–10) are needed so that struvite crystallisation becomes thermodynamically favourable. Struvite precipitation in streams with low concentration of phosphate or ammonium or low pH (as the influent or effluent of WWTPs) becomes an unaffordable option because of the requirement of adding high amount of magnesium (e.g. see Fig. 1 at the condition of pH = 7).

In any case, there are successful reports of P-recovery as struvite under full-scale conditions using side-streams obtained from the sludge line (Desmidt et al., 2015; Jabr et al., 2019; Law and Pagilla, 2018; Le Corre et al., 2009; Mayer et al., 2016) and successfully marketed technologies for the recovery of struvite include (amongst others) Ostara®, Airprex™, Multiform™, DHV Crystalactor® and NuReSys™ (Law and Pagilla, 2018). However, struvite crystallization also has some drawbacks in its practical application, for example, an unsatisfactory recovery efficiency related to the total P load in the raw sludge, in the range 45–80% in some full-scale projects for P-recovery from digested sludge or liquor, as reported by Remy and Jossa (2015).

Vivianite (Fe₃(PO₄)₂·8H₂O) is an alternative valuable product for P-recovery. It has a high market value (Table 1) as it is a relatively slow-release fertilizer widely found in soils, lake sediments, hydrothermal deposits or bogs (Wu et al., 2019). Moreover, the precipitating agent, iron, is a common dissolved mineral in natural water and Fe(III) salts are commonly used for improving sludge settleability (Li et al., 2014) and odour control (Zhang et al., 2009). The reaction to precipitate vivianite and its solubility is presented in Table 2.

The theoretical molar ratio of Fe:P to form vivianite is 1.5:1, but the optimal stoichiometric ratio range applied is 1.5–2 considering some of Fe(II) could be transformed to Fe(OH)₂ or oxidized to Fe(III) (Priambodo et al., 2017). The formation condition of vivianite is less dependant on pH (i.e. adequate range is 6–9) compared to struvite (8–9.5). Vivianite can be precipitated from the digested sewage sludge (Prot et al., 2020; Wilfert et al., 2018), wastewater (Wu et al., 2019) and aqueous solutions (Liu et al., 2018). Some researchers have shown that about 62% of P could be precipitated to vivianite from an anaerobic digestate with a pH around 8 (Li, Li and Li, 2018; Priambodo et al., 2017). However, the P-recovery process as vivianite also has disadvantages: i) the formation of vivianite in the sludge produces small crystals or aggregates (10–150 μm) resulting in a complex separation from sludge and ii) vivianite usually contains calcium or magnesium impurities (Robles et al., 2020). Impure vivianite is less stable, since it can be oxidized within 48 h, in contrast to the several weeks of stability reported for pure vivianite (Wilfert et al., 2018). In addition, separation and purification methods for vivianite still need further development (Robles et al., 2020).

Another alternative to struvite is hydroxyapatite (HAP, Ca₅(PO₄)₃OH). HAP can be directly used as a fertilizer in the field of agriculture (Johansson et al., 2017; Mayer et al., 2016) and as an adsorbent for removing dyes, emerging pollutants and heavy metals (Maity et al., 2018; Zhang et al., 2018). It is of great potential for P-recovery since calcium phosphate has a large market (Driver et al., 1999) and, thus, it is expected to be of essential importance for the closure of P cycle (Leinweber et al., 2018). The reaction to precipitate HAP and its solubility is presented in Table 2.

HAP is thermodynamically the most stable phase for orthophosphate

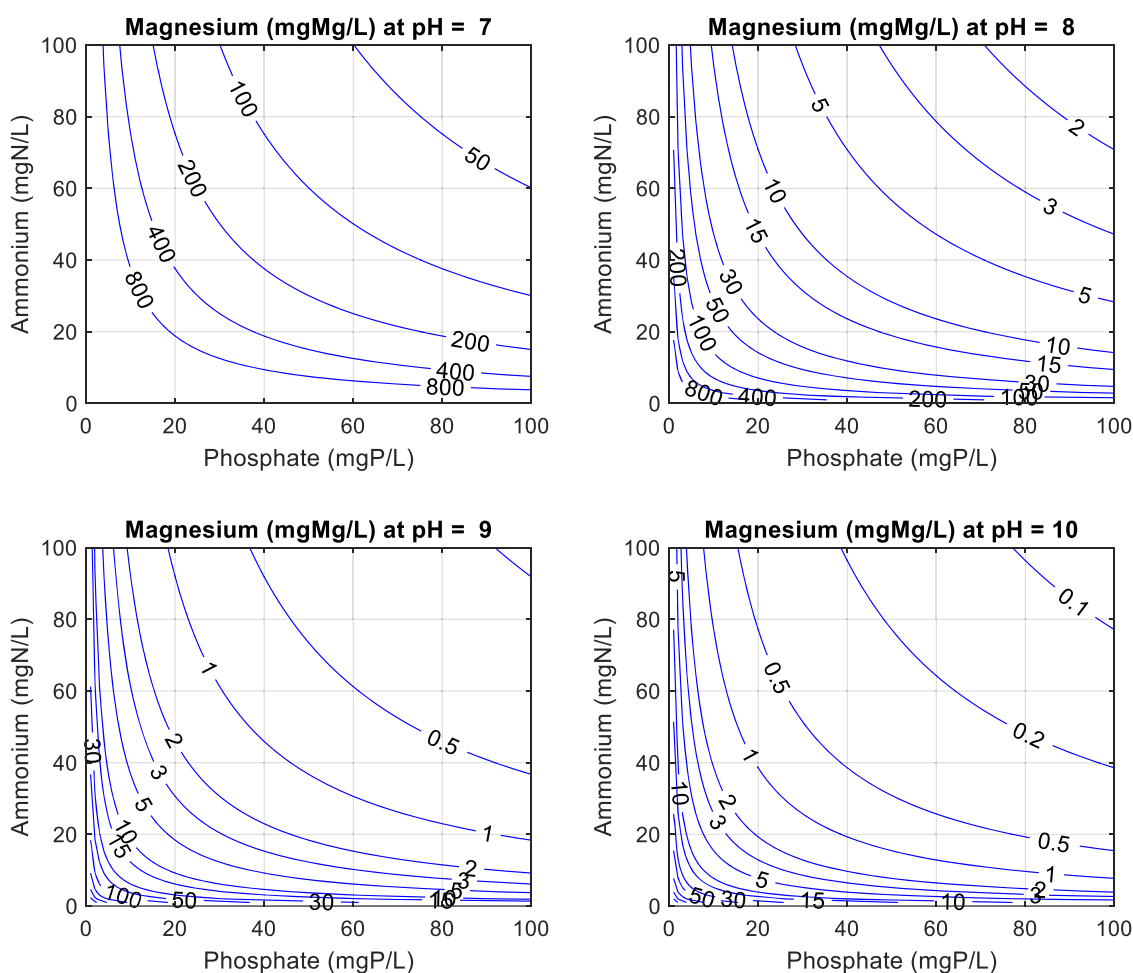


Fig. 1. Minimum magnesium (Mg^{2+}) concentration to reach the K_{sp} value for struvite at different pH and as function of phosphate and ammonium concentration. Addition of magnesium beyond this value will drive struvite precipitation.

and calcium in solution within a broad range of pH of 4–12 (Cichy et al., 2019; Oubagha et al., 2017). When dealing with the common problem of low P concentration in municipal wastewater, HAP is a more attractive option than struvite since struvite demands higher enriched P. For example, although this is highly specific depending on the pH and species concentration, for a typical wastewater composition (8 $mgPO_4^{3-}\text{-P/L}$ and 40 $mgNH_4^+\text{-N/L}$), and based on the HAP and struvite solubility, the addition of 10 $mgCa^{2+}\text{/L}$ and 10 $mgMg^{2+}\text{/L}$ at pH = 8 or higher leads to almost complete precipitation of Ca to produce HAP, whereas struvite is not formed even at a higher pH. Thus, some works have exploited this facility of HAP to precipitate and evaluated P-recovery strategies in lab-scale systems (Berg et al., 2005; Zou and Wang, 2016). However, HAP could lose advantage over struvite precipitation when aiming at a simultaneous recovery of N and P in real industrial wastewater treatment (Cichy et al., 2019). In addition, HAP has lower potential for fertilizer use (Robles et al., 2020).

2. Potential P-recovery from mainstream

P-recovery via precipitation (e.g. struvite, K-struvite, HAP, vivianite) requires a high concentration of P for displacing the solubility equilibrium to the precipitation side (Fig. 1). As described before, in EBPR-based WWTPs, digestate is a common option to precipitate/recover P since the sludge entering anaerobic digestion contains a high amount of internally stored P as poly-P that can be easily released inside the digester and results in a high concentration of P into the digestate. The digestate also contains high ammonia concentration and, thus, seems to

be a favourable option for struvite precipitation. However, these systems also bring disadvantages such as undesired precipitation in the anaerobic digester or in the downstream tubing and instrumentation (Doyle and Parsons, 2002). On the other hand, the high concentration of some ions in the anaerobic digestion sludge (e.g. calcium) could hinder the precipitation of P as struvite, since when P is released it can precipitate as other compounds such as HAP, brushite and other calcium phosphates. In any case, to determine the actual precipitation competition in a given anaerobic digestion scenario, not only solubility has to be taken into account, but also kinetics (D.Wild et al., 1996).

Apart from the digestate, the other location with the highest P concentration in an EBPR plant is the anaerobic reactor and, thus, it can be an alternative for P-recovery. During the anaerobic phase, PAOs have taken up most of the readily biodegradable COD and most of the P has been released into the bulk liquid, reaching levels from 30 to 70 mg P/L (Acevedo et al., 2015; Izadi et al., 2021, 2020; Zhang et al., 2020; Zou and Wang, 2016). Therefore, the anaerobic reactor appears as a good point to implement mainstream P-recovery whilst the digestate seems to be the ideal point for side-stream P-recovery. The integration of EBPR and mainstream P-recovery as struvite would be beneficial to prevent undesired precipitation throughout the plant and to reduce N load when treating wastewater with high ammonium concentration (1 g of P removed as struvite yields 7.92 g of struvite containing 0.45 g of N). Thus, it shows a good potential for improving system performance, and for this reason it has recently received the attention of researchers (e.g., Acevedo et al., 2015; Larriba et al., 2020; Shi et al., 2016; Zou and Lu, 2016).

The first proposal on the potential to obtain a P-rich liquor from the mainstream anaerobic reactor was reported by Van Loosdrecht et al. (1998): the Biological-chemical P and N removal (BCFS®) process (Fig. 2a). BCFS was a modification of the UCT design where a stream from the anaerobic reactor (for instance, an average flow of 10% of the influent flow) was sent to a sludge thickener and a phosphate stripper where FeCl₃ was dosed for P precipitation. Thus, a continuous EBPR operation and a P chemical precipitation system were combined. Modelling was used to better understand this process and to assess its performance under different scenarios (Barat and van Loosdrecht, 2006; Hao and van Loosdrecht, 2006). Hao and van Loosdrecht (2006) simulated the effect of anaerobic stripping on the P-removal and recovery performance within the BCFS® frame and reported an optimal stripping flow rate ratio (flow rate of supernatant/flow rate of influent) of 0.2 to favour P-recovery. Higher values resulted in decreased PAO activity and lower values in low P-recovery (see Section 3.2 for the discussion of this effect). Moreover, they showed that the system could operate with a lower influent COD/P ratio without hindering PAO activity. The ratio could be decreased from 20 to 10 g COD/g P and the system could still meet the discharge requirements of P (1 mg P/L) and maintain 36% of P-recovery.

This first experimental attempt of mainstream P-recovery already showed that there is a maximum threshold value for the amount of P extracted. Barat and van Loosdrecht (2006) implemented the model of the Hardenberg WWTP and simulated different control strategies to explore the P-recovery potential of the BCFS® configuration. They stated that the maximum potential P-recovery without affecting PAO activity was 60% of the influent P. Following this first report, several other works have presented configurations taking advantage of the PAO capability to increase P concentration under anaerobic conditions (Baeza et al., 2017; Shi et al., 2016; Wong et al., 2013).

The P-recovery efficiencies of the evaluated works were calculated according to Eqs. (1), (2) and (3):

$$P_{OUT} = P_{EFF} + P_{BIO} \tag{1}$$

$$P_{AS} = P_{INF} - P_{OUT} \tag{2}$$

$$PR = \frac{P_{AS}}{P_{INF}} * 100\% \tag{3}$$

where P_{OUT} represents the P output per day in the effluent (P_{EFF}) plus in the purged biomass (P_{BIO}), P_{AS} the P in the anaerobic supernatant (thus available for P-recovery), P_{INF} the total input of P in the influent to the secondary treatment per day and PR the P-recovery efficiency. P_{INF} is highly influenced by the configuration of the primary treatment. Typical P removal values around 11% have been reported for conventional primary treatments (Cornel and Schaum, 2009), but these values can be increased considerably to values of 40–99% if chemical dosing is used in the primary settler, as in the chemically enhanced primary treatment (CEPT) processes (Shewa and Dagnew, 2020).

The rest of the manuscript is organized as follows. The studies related to mainstream P-recovery currently available in the literature are briefly described in the following two subsections, classified by their operation mode: Section 2.1 for sequencing batch reactors (SBRs) and Section 2.2 for continuous systems. This classification separates SBR configurations, which offer greater configurability than continuous systems, although they are typically used for lab-scale evaluation of different operating strategies, from continuous systems, which offer more stable operation and are more suitable for full-scale implementation, although the achievable P concentration is typically lower. After this schematic description, Section 3 critically discusses the most important factors affecting P-recovery: SBR vs. continuous configurations, extraction ratio, influent COD/P ratio, concentration of P in the recovered stream, carbon source needs and future outlook.

2.1. SBR configurations

Most of the configurations for mainstream P-recovery studied in previous works have been based on SBR operation (Table 3, Fig. 3). The supernatant after the anaerobic phase is used for P-recovery since it has the highest P concentration in all the cycle. SBRs provide greater flexibility than continuous systems for research studies, and the fact that particular actions can be taken in a single cycle (such as extra COD dosing or supernatant extraction) facilitates the understanding of P-recovery performance under different operation modes. For example,

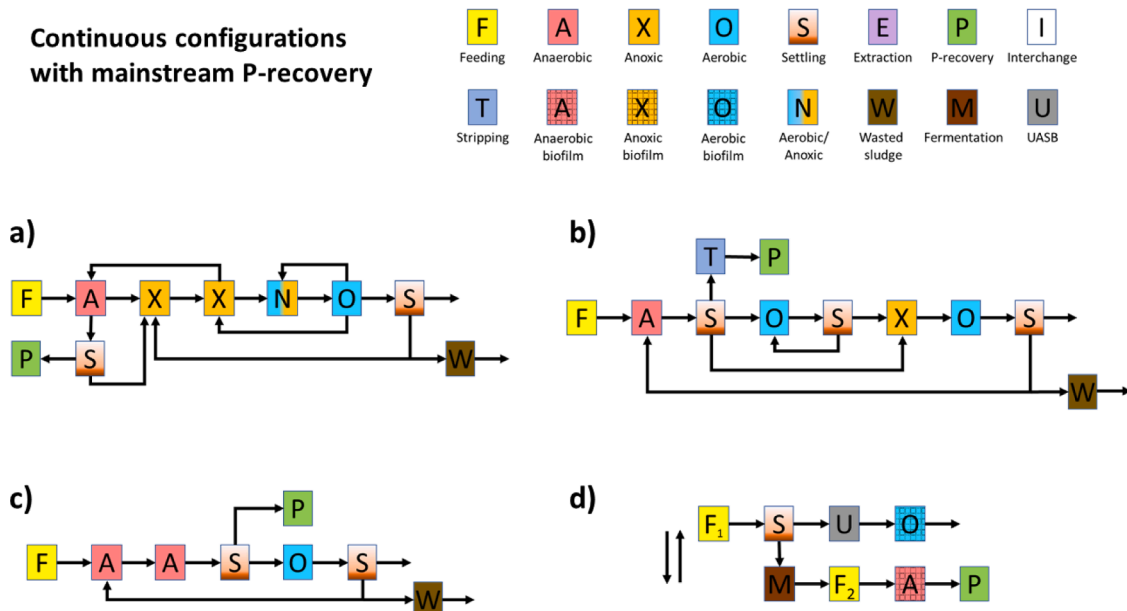


Fig. 2. Schematic representations of the main continuous configurations with mainstream P-recovery found in the literature. a) BCFS process implemented in Hardenberg (Hao and van Loosdrecht, 2006), b) A2N-IC process (Shi et al., 2012), c) EBPR system (Valverde-Pérez et al., 2015), d) trickling filter with alternative anaerobic and aerobic phases and sludge fermentation (Kodera et al., 2013). Please note that the symbols in the legend include those used in Figs. 2 and 3.

Table 3
Summary of P-recovery from mainstream and the related operation parameters based on sequencing batch configurations.

Reference	Configuration	Volume (L)	Nitrification	SRT (d)	HRT (h)	Temperature (°C)	DO (mg/L)	pH	Recovery species	Wastewater	InfluentCOD/ P (gCOD/gP)	Influent P ^a (mg/L)	[P] in the recovery stream (mg/L)	Extraction percentage ^b (%)	P-recovery efficiency ^c (%)	P-removal efficiency ^d (%)	Sporadic external carbon addition
(Xia et al., 2014)	anaerobic/ aerobic	8	no (ATU ^e)	60	24	18–22	1.5–2.5	7.0 - 8.0	Mg ₃ (PO ₄) ₂	synthetic	22	9	240 ^f	12.5	79	100	acetate 640 mg COD/L
(Acevedo et al., 2015)	anaerobic/ aerobic	7	no (ATU)	10	12	20	1.5–2.5	7.0 - 8.5	struvite	synthetic	13	7.5	60–72 110–200	85 ^g	59 ^h 81 ^h	100 100	acetate 100 mg COD/L acetate 350 mg COD/L acetate 370 mg COD/L
(Wong et al., 2013)	anaerobic/ano- xic + biofilm	10	no (ATU)	n.d.	n.d.	22 ± 2	5–8	6.8 - 7.2	n.d.	synthetic	22	8	>90	8	80	79 ± 6	no
(Shi et al., 2016)	A2O + induced crystallization	–	yes	15	n.d.	20	n.d.	8.2 - 8.6	HAP	synthetic /raw	41	6.1	20	20	33	100	no
(Lv and Yuan, 2015)	SBR+EBPR	10	n.d.	11	16	20 ± 1	> 2	7.1 - 8.3	vivianite	synthetic	40	10	39	50	50	95- 97	no
(Zou and Wang, 2016)	anaerobic/ anoxic	6	yes	20	n.d.	16–21	n.d.	>8.5	HAP	synthetic	40	5	15.8	20	59.3	91.6	no
(Baeza et al., 2017)	anaerobic/ aerobic	10	yes	15	n.d.	n.d.	3	n.d.	struvite	urban (model)	33	9	57	5	63	100	no
(Guisasola et al., 2019)	anaerobic/ aerobic	10	no (ATU)	10	12	25	2.5–3.5	7.50 ± 0.05	struvite	synthetic	15 10 10 8	20 30 30 40	40–60 71–123 52–95 59–78	10	54 ^c 66 ^c 69 ^c 33 ^c	98 ± 1 67 ± 10 82 ± 14 77 ± 6	no
(Larriba et al., 2020)	two sludge system	7800	yes	15	24–36	20 ± 6	3	n.d.	struvite	raw	63	3.8	35	8.5	63	93 ± 9	no
(Yu et al., 2021)	anaerobic/ aerobic	5.4	yes	10	24	16 ± 0.5	1 0.6 0.5	n.d.	struvite	synthetic	400/8	8	26–19 25–16 21–6.5	8, 11, 17	10, 24, 50 4, 18, 40 0, 0	>90 >90 60	no

a Influent entering the secondary treatment.

b Calculated as the fraction of the volume of the anaerobic supernatant extracted with respect to the volume of the reactor.

c Calculated as the percentage of P in the anaerobic supernatant extracted with respect to the total input of P (per day).

d Calculated as the fraction of P in the influent that is removed.

e ATU: Allylthiourea.

f P concentration of a parent batch reactor with extra dose of acetate. The anaerobic P concentration is around 80 mg/L.

g Average.

h Weekly average.

n.d. Not described.

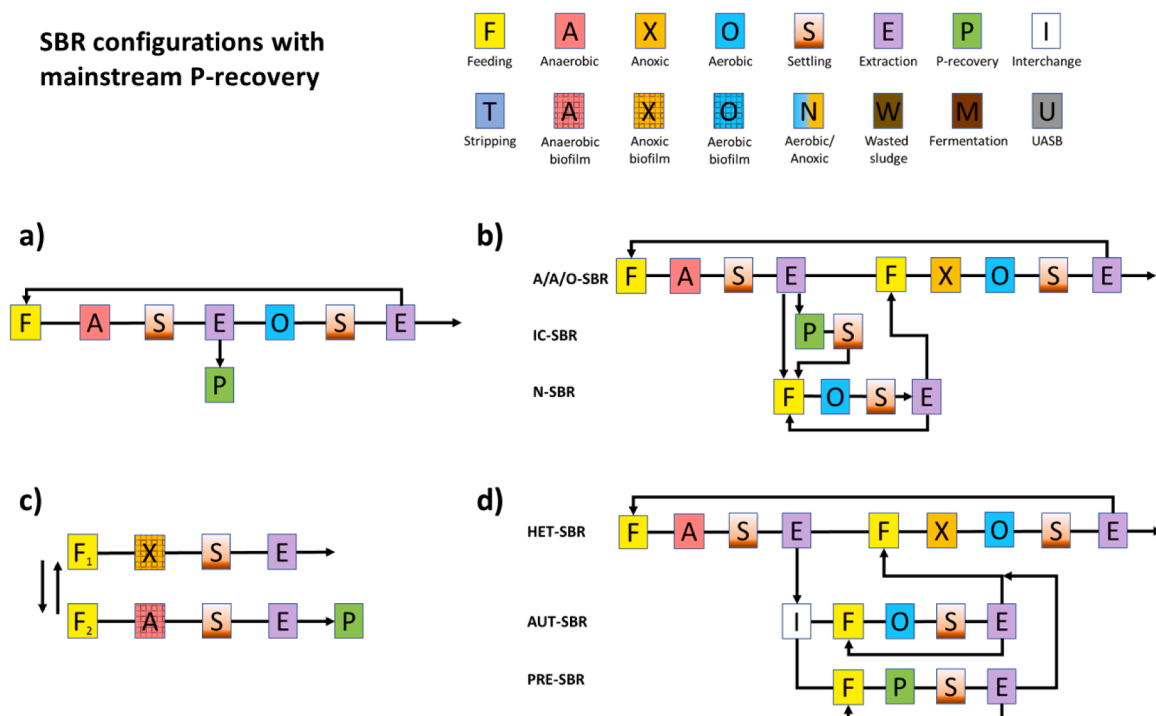


Fig. 3. Schematic representations of the phases used in SBR configurations with mainstream P-recovery found in the literature. a) anaerobic/aerobic SBR EBPR² process (Baeza et al., 2017; Guisasola et al., 2019), b) anaerobic/anoxic/nitrifying-induced crystallization SBR process (Shi et al., 2016), c) two-step biofilm system alternatively exposed to a dilute wastewater stream to facilitate P uptake (under anoxic condition) and to a recovery stream to facilitate P release (under anaerobic condition with external carbon source) (Wong et al., 2013), d) mainstream shortcut enhanced phosphorus and PHA recovery (mainstream SCEPPHAR) configuration (Larriba et al., 2020). Please note that the symbols in the legend include those used in Figs. 2 and 3.

some authors have evaluated the effects of sporadic extractions of anaerobic supernatant and others have tested the automated extraction every single cycle. Long-term operation is in general required to demonstrate the ability of these two different approaches to maintain stable PAO activity despite the extractions, as a decrease in P availability may hinder PAO metabolism.

Wong et al. (2013) reported a novel two-step biofilm (Fig. 3c) process to facilitate anoxic P-recovery. In the first step, the PAO-enriched biofilm was immersed into a diluted wastewater stream (F₁) so that the biomass could take up P and nitrate to remove P and N of the wastewater. In a second step, the biofilm was subjected to a carbon-enriched stream (F₂) under anaerobic conditions, where acetate was taken up and P was released. Then, the biofilm acted under aerobic condition storing P from wastewater and under anaerobic condition releasing P and hence allowing the generation of an enriched P-recovery stream. The anaerobic supernatant volume (1.8 L) was four times lower than the initial volume of wastewater (7.2 L) and achieved a P concentration almost 4 times higher (28 vs 7 mg P/L). An additional experiment was conducted to improve the system efficiency. They applied a highly enriched carbon stream after 10 cycles of normal operation and they obtained an anaerobic supernatant (around 100 mg P/L) which is theoretically high enough for an efficient P-recovery by struvite (Rittmann et al., 2011). Nevertheless, they did not test the effect of extracting this P on PAO survival neither in a short-term nor a long-term basis.

Xia et al. (2014) studied a configuration where a fraction of the anaerobic mixed liquor from a parent EBPR-SBR was treated in a separate batch reactor, and intermittent external carbon source was dosed to stimulate P-release in view of P-recovery. The sludge from this batch reactor was separated with a non-woven cloth and recycled to the parent SBR, whereas the supernatant was treated with MgCl₂·6H₂O to recover P as Mg₃(PO₄)₂. Thus, dosing 10% of additional carbon source in the batch reactor resulted in the recovery of 79% of the P in the influent in single

cycle. The authors applied this new P-recovery sequence for nearly one month in only one out of four cycles although recommended to evaluate this approach in a longer period.

Acevedo et al. (2015) investigated a comparable EBPR-SBR configuration but the extra carbon was dosed in situ in the anaerobic phase. The frequency of this extra carbon addition and subsequent anaerobic extraction for potential P-recovery was varied in different sets of experiments: two times per week with low P extraction or once per week but with high P extraction. Their results were promising since they obtained 59% recovery of the influent P when extracting P two times per week and 81% when they extracted once per week. In the latter case, they used an extra VFA addition of 9% of the usual load. No P-removal deterioration was observed in the cycle after both extractions, but the long-term impact of a P-recovery strategy with continuous extraction of P was not investigated.

Lv et al. (2014) studied a 10 L EBPR-SBR system to investigate the effect of the extraction of 5 L of supernatant enriched in P for subsequent chemical precipitation, supernatant that was obtained after an additional anaerobic settling period. The supernatant was treated with FeCl₃ for P-precipitation and the treated supernatant was returned to the aerobic stage. This extraction was applied to the SBR once every 3 cycles for a long term and observed the decrease of P-release, P-uptake and the concentration of intracellular poly-P. EBPR activity was deteriorated, and P-removal efficiency decreased to about 75%. They disabled the P-recovery strategy and tried to recover the EBPR activity by operating under conventional anaerobic/aerobic conditions, but the system did not recover. After that work, Lv and Yuan (2015) demonstrated that the same system could have a successful short-term performance (40 d) when 50% of anaerobic supernatant (with a concentration of 39.4 mg P/L) was extracted. The recovery process was operated in another separated reaction-sedimentation basin with FeCl₃ as a precipitating agent, and half of the anaerobic supernatant was removed for P-recovery. EBPR activity was strongly limited because of the shortage of poly-P

as a continuous stripping of 40 d, and it could not be recovered even though the stripping was stopped for 60 d

In the same direction, [Guisasola et al. \(2019\)](#) proposed the operation with an automated extraction of anaerobic supernatant every cycle to understand the P-recovery capacity in a long-term basis but without any external dose of additional organic carbon ([Fig. 3a](#)). Thus, the anaerobic P concentration was lower than in the previous reports since it was the routine anaerobic P concentration of the system. They demonstrated that a successful P-recovery strategy could be maintained with 1 L (10% of reactor volume, 20% of the influent) of automated anaerobic extraction per cycle during more than 120 days without any deleterious effect on EBPR activity. They tried to push the system into the limits and operated with other COD/P ratios and different extraction conditions in order to investigate two different aspects: the minimum initial COD/P ratio and the maximum amount of P extraction that the system could tolerate without the loss of the PAO activity. They demonstrated that up to 60% of the influent P could be recovered (when extracting 10% of the anaerobic volume per cycle) without any harmful effect on PAO activity and that the system could be operated under low COD/P ratios (i.e. 7.5 gCOD/gP).

([Zu, Nan, He and Liu, 2020](#)) also used a lab-scale SBR to study the effect of the volume of anaerobic extraction on the biological C and P-removal performance. Different extraction ratio of anaerobic supernatant (30%, 60% and 90%) were tested for P-recovery as vivianite. Extraction was carried out every fourth cycle. P-removal efficiency was improved when increasing from 30 to 60% of the extraction volume. However, the phosphorus balance and the PAO population severely decreased under the extraction ratio of 90%. The harm was irreversible even after a long-term normal operation without stripping process.

In addition to P-removal and P-recovery, some researchers have studied the simultaneous P and N recovery from mainstream in a batch system configuration. [Shi et al. \(2016\)](#) proposed an anaerobic/anoxic/nitrifying-induced crystallization (IC) SBR configuration ([Fig. 3b](#), named as A₂N-IC-SBR) for N, P-removal and P-recovery which was composed of three different SBRs (A/A/O-SBR, N-SBR, and IC-SBR, with 10, 10 and 1.3 L). Nitrifiers oxidised ammonium to nitrate, which was denitrified by denitrifying PAO (DPAO) linked to P uptake. During this process, 1.3 L of anaerobic supernatant extraction was sent to the IC column with calcite seeds for P-recovery. The percentage of chemical P-removal with respect to the total P-removal was 14.9–17.1%.

Similarly, [Zou and Wang \(2016\)](#) implemented a lab-scale EBPR--P-recovery process aiming at removing COD, nutrients and recovering P, which consisted of a DPAO-enriched SBR operated with anaerobic-aerobic/anoxic mode and an IC P-recovery column. The IC column received part of the supernatant obtained after a post-anaerobic settling phase and the IC supernatant was returned to the anoxic phase with external nitrate solution dosage. The system was operated with either synthetic (around 250 mg COD /L) or real domestic wastewater (COD concentration between 153 and 223 mg COD/ L). They achieved successful N and P-removal and P-recovery efficiency via HAP precipitation: the average removal efficiencies were 82.6%, 87.5% and 91.6% for COD, P and N, respectively. The flow ratio of anaerobic supernatant to influent (from 0 to 0.5) was varied and the maximum P-recovery as HAP (up to 59%) was achieved at a ratio of 0.3.

[Salehi et al. \(2019\)](#) explored the possibility of a simultaneous nitrification, denitrification and P-removal (SNDPR) process for nutrient removal and P-recovery. A lab-scale anaerobic/aerobic SBR was employed with synthetic wastewater feeding. They achieved anaerobic liquor with high P concentration (i.e. around 100 mg/L) in granular SNDPR process with an influent P of 10 mg/L and a low COD = 200 mg/L.

[Yu et al. \(2021\)](#) used an anaerobic-aerobic lab-scale SBR (5.4 L) to promote biological C, N and P-removal. Biological nitrogen removal (BNR) was achieved with low DO during the aerobic phase and thus simultaneous nitrification and denitrification was promoted. Successful nutrient removal was stable for a long period (310 d) under different

anaerobic supernatant extraction ratios (0, 1/12, 1/9, 1/6) and low DO conditions (0.6 and 1 mg/L). More than 40% of P could be recovered as struvite due to these extractions. The reactor began to deteriorate and the efficiency of P-removal decreased to 60% when operating the system with the lowest DO of 0.2 mg/L and a relatively high extraction ratio (1/9), and no P-recovery was found afterwards. They proposed a DO of 0.6 mg/L and an extraction ratio of 1/6 in view of saving energy and an optimal condition of DO of 1 mg/L and extraction ratio of 1/9 to maximise the process performance.

Some modelling studies have been reported about mainstream P-recovery with SBR systems. [Baeza et al. \(2017\)](#) simulated an EBPR-based SBR configuration for P-recovery named EBPR². A typical EBPR anaerobic/aerobic cycle configuration was modified including, after the anaerobic phase, a settling and decanting phase where the P-enriched stream was extracted for P-recovery ([Fig. 3a](#)). They simulated different scenarios and showed that it was possible to obtain a stable operation with a high amount of P recovered (up to 63%) when the supernatant extraction in each cycle was 5% of the reactor volume. Moreover, total P-removal was achieved regardless of COD fractionation in the wastewater. In addition, they proposed the inclusion of anaerobic sludge purging, which could be a good option in view of a higher energy recovery (i.e. increasing biochemical methane potential per gram of volatile suspended solid). Sludge obtained from the anaerobic phase contains a higher concentration of poly-hydroxyalkanoate (PHA), which would boost the production of methane during anaerobic digestion ([Chan et al., 2020a](#)). The highest PHA production was obtained by extracting 4.3% of the reactor volume per cycle.

Taking into account the previous simulations and the pilot plant experiments of a two-sludge configuration with EBPR and via-nitrite N-removal ([Marcelino et al., 2011](#)), a new two-sludge process including P-recovery was designed ([Larriba et al., 2020](#)). This novel configuration ([Fig. 3d](#)) was named shortcut enhanced P and PHA recovery (mainstream SCEPPHAR) and was tested in a large demo pilot-scale (total volume of 7.8 m³) fed with real primary influent ([Larriba et al., 2020](#)). It consisted of a two-sludge SBR system: HET-SBR was operated as a conventional anaerobic/anoxic/aerobic (A²O) EBPR-SBR and AUT-SBR was aerobic and aimed at a complete or partial nitrification. A third crystallization reactor (PRE-SBR) was designed so that the supernatant extracted after the anaerobic phase of HET-SBR could be used for P-recovery. An additional interchange vessel was required to interchange supernatants amongst these reactors, in order to avoid idle phases and to ensure that all three reactors could operate simultaneously rather than sequentially. This configuration was operated successfully for a long term for C, N and P-removal (86 ± 12%, 93 ± 9% and 79 ± 6%) with either full nitrification or nitrite shortcut. P-recovery as struvite was carried out in this third reactor by Mg²⁺ dosing at a pH of 8.5, allowing the recovery of an average of 45% of the influent P as struvite precipitate (with a maximum peak of 63%).

2.2. Continuous configurations

This section details the reported alternatives for P-recovery from mainstream with continuous configurations, in addition to the seminal BCFS process discussed above ([van Loosdrecht et al., 1998](#)). Continuous systems are often used to test the P-recovery strategies previously developed in SBR mode. Under these conditions, part of the anaerobic supernatant is diverted to a P-recovery equipment and, in some cases, the flow is returned to the system after P is recovered.

For instance, an anaerobic-anoxic/nitrifying (A₂N) continuous configuration combined with an induced P-crystallisation (IC) column was proposed by [Shi et al. \(2012\)](#) for mainstream P-recovery in addition to removal of COD, N and P ([Fig. 2b](#)). Part of the P enriched anaerobic supernatant was extracted to precipitate calcium phosphate crystals in the IC column. The conventional A₂N system performance was compared to that of A₂N-IC. A₂N-IC showed a more stable performance than A₂N. The authors used a wide range of COD/P ratios (from 17.7 to

Table 4
Summary of P-recovery from mainstream and the related operation parameters based on continuous configurations.

Reference	Configuration	Volume (L)	Nitrification	SRT (d)	HRT (h)	Temperature (°C)	DO (mg/L)	Recovery species	Wastewater	Influent COD/P (gCOD/gP)	Influent P ^a (mg/L)	[P] in the recovery stream (mg/L)	Extraction percentage ^b (%)	P-recovery efficiency ^c (%)	P-removal efficiency ^d (%)	Sporadic external carbon addition
(Barat and van Loosdrecht, 2006)	BCFS-TUD ^e model calibrated for Hardenberg WWTP	1.215·10 ⁷	yes	n.d.	3.8	10–20	0.6 and 2.8	P Stripper (stripping)	model	54	14.2	60 - 70 kg P/d	45	~60	> 90	no
(Shi et al., 2012)	BNR-IC ^f (A2O-crystallization)	31	yes	15	n.d.	n.d.	2–3.8	HAP	synthetic	19.9 ± 1.5 31.0 ± 4.2 42.4 ± 4.4 67.4 ± 10.2	12.7 ± 0.7 7.82 ± 0.68 5.73 ± 0.51 3.75 ± 0.51	n.d.	20	8 9.1 10.8 12.6	> 99	no
(Kodera et al., 2013)	PAOs-enriched biofilm within a modified trickling filter	2.4	n.d.	n.d.	0.5	20	0.9 L/h air ^g	struvite	synthetic	200/5	5	125	10	~60	50–60	no
(Valverde-Pérez et al., 2015)	EBP2R green microalgae cultivation	5.1·10 ⁶	yes	5	4.25	n.d.	0–3	green microalgae	model	79	9	n.d.	30	70	n.d.	no
(Zou and Lu, 2016)	BNR-IC	12.4	yes	n.d.	n.d.	n.d.	>2	HAP	synthetic real domestic synthetic	25 20–30	10 7.08–7.69	28 12	35 35	70 74	95.2 ± 1.0 90.7 ± 1.2	no
(Dai et al., 2017)	BNR-IC	316	yes	12–16	n.d.	n.d.	n.d.	HAP	synthetic	50	5	24	30	83	95	no

a Concentration of P for the EBPR process.

b Calculated as the ratio of the flow rate of anaerobic supernatant diverted to the P-recovery unit and flow rate of the influent.

c Calculated as the percentage of P in the anaerobic supernatant extracted for P-recovery per day in respect of the total input of P in the influent per day.

d Calculated as the removal fraction of phosphate in the effluent with respect to the phosphate in the influent.

e BCFS-TUD: Biological-chemical phosphorus and nitrogen removal system-Model system of Delft University Technology.

f BNR-IC: An anaerobic-anoxic/nitrifying (A2N) continuous configuration for biological nutrients removal (BNR) combined with an induced P-crystallisation (IC) column.

g Aeration flow rate.

n.d. Not described.

67.4 g COD/gP) and found that the P-removal efficiency was consistently maintained at 99.2% for A₂N-IC, regardless of the influent COD/P ratio tested. However, the A₂N process reduced P-removal efficiency from 93.0% to 65.7% when the influent COD/P ratio was decreased from 67.4 to 17.7 gCOD/gP. P-recovery in the A₂N-IC process ranged from 8.9 to 12.9% when the influent COD/P ratio was reduced from 67.4 to 19.9 gCOD/gP. Later, Zou and Wang (2017) used the same configuration and studied the effect of various operational conditions on the IC performance and optimized the crystal dose acting as a seed, pH, temperature, DO, molar Ca/P ratio and reaction time in view of boosting P-recovery. They showed that on the optimal conditions (80 min, pH of 8.5, molar Ca/P ratio of 2.0, 60 g/L seed crystal loads and 4.0 L/min of aeration flow) the system reached an excellent P-recovery of 92.3%. In addition, they found that the temperature had a slight effect on induced HAP crystallization, thus benefiting the integration of EBPR with induced HAP crystallization.

In a similar configuration, Dai et al. (2017) evaluated the influence of the flow of the anaerobic supernatant to IC in a long run (over 180 d) and found that the optimal ratio of anaerobic supernatant extracted to influent was 0.3. Under these conditions, P-recovery efficiency reached 72.4% at the optimal HRT = 0.5 h in IC reactor. Higher extraction ratios could disrupt system stability without enhancing P-recovery and could induce a change of the activated sludge characteristics since it favoured the percentage of extracellular polymeric substances of the sludge. The same authors reported a modelling study about the continuous A₂N-IC aiming at finding the optimal conditions to maximize energy savings and minimize emission reduction (Dai et al., 2019). Their results indicated that P-recovery decreased with the reduction of the extraction ratio and the IC volume.

Kodera et al. (2013) studied a configuration based on a PAO-enriched biotrickling filter, exposed alternatively to anaerobic and aerobic conditions (Fig. 2d). P-uptake by PAO was promoted under aerobic conditions, when the filter treated by recirculation a synthetic stream emulating an urban wastewater, which contained a low concentration of P (5 mg P/L). For the anaerobic phase, the system required a stream with VFA to enhance P-release, with up to 2000 mg COD/L which could be generated by primary sludge fermentation, although in this work a synthetic stream was used. The effluent from the anaerobic phase had a high concentration of P (125 mg P/L) that was adequate for P-recovery. The system operated for a long period of time (250 d) with this configuration, obtaining a P-recovery of almost 60% of the influent P.

In the frame of the cultivation of microalgae for P-recovery, Valverde-Pérez et al. (2015) proposed a novel model-based design of P removal and recovery system consisting of a conventional continuous A²O operation where nitrification was suppressed at low DO (Fig. 2c). The objective was to produce a nutrient medium for microalgae cultivation to produce biofuel or a natural fertiliser for direct use in agriculture. The P recovered for algae cultivation was obtained from the anaerobic effluent as a stream with high P concentration. They investigated the potential P-recovery under different scenarios of DO and SRT and obtained a maximum of 70% P-recovery of influent P, when 30% of the influent flow was diverted as a P-stream and the system was operated at SRT = 5 d. At DO = 1.5 mg/L, nitrification was still being inhibited due to this low DO and P uptake was favoured, increasing P-recovery up to 75%.

3. Discussion

Amongst the different locations for P-recovery in EBPR-based WWTPs, the supernatant of the anaerobic reactor has recently gained attention since mainstream P-recovery would i) mitigate undesired precipitation of P in the sludge line, ii) reduce the COD requirements for P-removal, iii) enable the treatment of wastewater with lower COD/P ratio and iv) depending on the recovery agent, decrease the N load to the plant and, thus, its operational costs. As detailed above, mainstream P-

recovery can be achieved with different configurations and, thus, several operation parameters are essential for an efficient P-recovery performance. Tables 3 and 4 summarize the different reported proposals for the implementation of P-recovery from mainstream and lists, for each case, the applied equipment and recovery methods, basic operation parameters (SRT, HRT, operation temperature, aeration, pH, extraction volume and frequency) and influent characteristics.

3.1. SBR vs continuous configurations for mainstream P-recovery

Mainstream P-recovery under SBR conditions is based on extracting part of the supernatant once the anaerobic phase is finished, whereas it consists of diverting part of the anaerobic liquid to a P-recovery unit under continuous conditions. Most reports on mainstream P-recovery employed lab-scale SBRs (Table 3), as SBRs have several advantages compared to continuous systems. On the one hand, it is easier to obtain a biomass-free anaerobic supernatant as a settling phase can be easily included after the anaerobic phase, whereas in the case of continuous system a membrane or a combined settling reactor+biomass recycle would be needed (Sekine et al., 2018). Moreover, SBRs show a lower space requirement and lower configuration and installation costs, as well as the flexibility to face operational or influent variations (Jiang et al., 2016; Tomei et al., 2016). However, it can also lead to higher operational costs and capital expenditures due to its discontinuous operation, as reported in (Larriba et al., 2020) when comparing the mainstream SCEPPHAR configuration to a conventional continuous A²O configuration.

3.2. Extraction ratio

As mentioned before, the key operational parameter for efficient P-recovery is the extraction ratio, i.e. the percentage of anaerobic volume extracted for an SBR or the flow diverted from the anaerobic reactor to the P-recovery unit for a continuous system. High anaerobic extraction volumes should be chosen to maximise P-recovery, but too high values may hinder P-storage activity of PAO. The rationale is that the effect of internal poly-P limitations is not observed with single periodic extraction or in the short-term, but after several cycles, high P extraction values would deplete the internal poly-P reserves and PAO would lose the competitive advantage versus other microorganisms such as glycogen accumulating organisms (GAO), which are able to uptake VFA as PHA without requiring poly-P reserves. For this reason, many authors have opted for sporadic extraction of anaerobic liquid. Thus, the reported P-recovery efficiencies for single/short-term experiments can be very high, particularly if an extra dose of carbon is added under anaerobic conditions to boost P-release. In this sense, as observed in Table 3, the P-recovery efficiency ranges from 30 to 90% as a function of the SBR operational conditions. For example, Acevedo et al. (2015) reported P-recovery efficiencies around 80% in a single cycle with extraction values of nearly 75% and an external carbon dosage of 350 mg COD/L as acetate. However, it must be carefully considered that even if 100% P-recovery from one out of four cycles was achieved, the average extraction per treated cycle in the plant would only be about 25% of the P input. Moreover, one should realise that these high P extractions are sporadic and could not be sustained in a routine basis due to loss of PAO activity. For example, Lv et al. (2014) operated an EBPR-SBR with an extraction ratio of 0.5 once every 3 cycles. After 83 days of P stripping, P-removal performance decreased, observing a metabolic shift from PAO to GAO and PAO declined to less than 1% of the population. In a subsequent study, Lv and Yuan (2015) extracted half of the anaerobic supernatant every cycle and reached a P-recovery efficiency of 50% in 40 days of operation without additional VFA dosage in the anaerobic stage, but again the internal poly-P levels decreased and the system failed in the next 20 days

On the other hand, if the extraction ratio is properly selected to avoid over-extraction, PAO activity can be maintained for a longer period of

time. In this sense, a maximum automated cycle extraction ratio of 10% was suggested to ensure the stability of PAO activity and thus the feasibility of a long-term P-recovery process (Guisasola et al., 2019), which allowed the recovery of 60% of influent P without any harmful effect on PAO.

In a modelling study, Baeza et al. (2017) evaluated the impact of the anaerobic supernatant extraction ratio on the performance of an EBPR-SBR P-recovery system. If the objective was achieving the highest concentration of P in the supernatant to facilitate precipitation, an extraction ratio lower than 5% of the reactor volume (10% of the influent) was recommended, for which a concentration of 61 mg P/L was obtained. Lower ratios did not increase P concentration and resulted in a lower amount of P redirected to the P-recovery system, due to the lower volume. On the other hand, higher ratios led to a decrease of PAO activity and hence lower P concentration, hindering P-precipitation.

Regarding the experimental studies, the range of extraction ratios that led to a successful long-term P-recovery was 8–15% of reactor volume in batch mode and 20 to 35% of flowrate under continuous mode. Moreover, lower extraction ratios lead to higher anaerobic P concentrations, which from a practical point of view is very interesting since precipitation becomes more feasible (i.e. with less chemical dosage requirements). On the other side, higher extraction ratios enable a higher amount of P-recovery but at expenses of higher costs since the anaerobic P concentration is lower. In summary, choosing the optimal extraction ratio requires a balance between a) optimal PAO activity, high anaerobic P concentration and a low amount of P recovered vs. b) hindering PAO activity, low anaerobic P concentration and a high amount of P recovered. Thus, it is difficult to suggest a general optimal value but, based on the results reported in the literature, we propose 10% as a first initial guess for the extraction ratio when designing batch systems and 25% when designing continuous systems.

3.3. Influent COD/P ratio and N-removal

Another significant parameter is the influent COD/P ratio. The theoretical studies on mainstream P-recovery indicate that wastewater with a low COD/P ratio could be successfully treated in systems where mainstream P-recovery is implemented due to a much better utilisation of the influent C. The range of influent COD/P ratios in Tables 3 and 4 is very wide (from 7.5 to 80 gCOD/gP), since few reports aimed at finding the minimum COD requirements for a successful mainstream P-removal/recovery. In general, continuous systems are reported with a higher COD/P ratio. In addition, some of the reports include simultaneous N and P-removal and, hence, a higher amount of COD is needed. Other reports somehow prevent nitrification from occurring either with a low SRT, with allylthiourea (ATU) addition, low DO setpoint or with a very low influent nitrogen load. For example, Valverde-Pérez et al. (2015) fixed a DO setpoint of 1.5 mg/L and P-recovery efficiency improved up to 75% of the influent P because the activity of nitrifiers was being limited at low oxygen concentrations and P uptake was favoured. In any case, the choice of the DO setpoint is not straightforward since too low DO values may also hinder PAO activity.

Guisasola et al. (2019) prevented nitrifying activity using ATU, tested different influent COD/P ratio and showed that the minimum initial COD/P ratio (7.5 gCOD/gP) allowed 60% of the P-recovery without any harmful effect on PAO activity, which is the value that agrees with the first experiments conducted with the BCFS® methodology (Barat and van Loosdrecht, 2006). This value also agrees with the modelling work of Baeza et al. (2017), where a supernatant extraction of 5% of the reactor volume in each SBR cycle led to a stable operation with up to 63% of P recovered. The rest of P removed was incorporated to the purged biomass, where additional P-recovery strategies reported for the sludge line (i.e. as sewage, sewage ash or digestate) could also be applied in view of boosting the global P-recovery efficiency.

The interaction between mainstream P-recovery and nitrogen removal deserves more attention. If P is recovered as struvite, part of the

influent ammonium (1:1 molar P:N ratio) would be directly recovered without nitrification/denitrification needed. This would decrease the N load to the plant and, thus, the operation costs (e.g. those related to aeration or biomass handling). To the best of our knowledge, an experimental study that analyses these interactions has not been reported yet. When aiming at simultaneous N and P-removal, a higher COD/P ratio is needed in order to divert some electron donor to BNR. BNR results in a certain entrance of nitrate to the anaerobic reactor and, if COD limitations exist, that would hinder the amount of P to be released and, thus, the amount of P to be recovered. Guerrero et al. (2011) already demonstrated that PAO can outcompete denitrifiers under certain operational conditions. Salehi et al. (2019) showed that P-recovery could be achieved together with BNR with synthetic wastewater of only 200 mg COD/L and a COD/P ratio of 20 gCOD/gP.

3.4. Concentration of P in the recovered stream

A high P concentration is critical to guarantee a high P-recovery performance: the Phostrip and BCFS® processes report a minimum phosphate concentration in the stripper stream of 25 mg P/L for an efficient P-recovery during the crystallization process (Barat and van Loosdrecht, 2006; Hao and van Loosdrecht, 2006). Other works report a higher minimum P concentration (more than 50 mg/L) for the struvite precipitation (Cordell et al., 2009; Kodera et al., 2013). Most of the mainstream P-recovery processes listed in Tables 3 and 4 report P concentrations higher than 50 mg/L in the recovery stream. Even so, some works report more than 100 mg/L of P in the enriched stream by repeated exposure of the biofilm to external COD source (acetate or propionate). The typical reported anaerobic P concentrations in EBPR systems are in the range of 30 to 70 mg P/L, so they are in the bottom limit and this is why some of these reports use an external carbon dosage under anaerobic conditions to increase anaerobic P-release. In a real environment, external COD dosage under anaerobic conditions boosts P-release at expenses of higher operating cost. Moreover, this addition should be sporadic or, otherwise, the internal poly-P reserves will be depleted and, thus, this operation would not be sustained in a long-term basis.

3.5. Carbon source needs

Most of the works in Table 3 and 4 were conducted at lab-scale and used synthetic wastewater to mimic urban wastewater. However, some of them prepared a highly loaded wastewater with an extra C inlet, normally acetic acid or propionic acid (Acevedo et al., 2015; Guisasola et al., 2019; Wong et al., 2013) and there was no need for a sporadic carbon dosage. In the case of the reports with real wastewater, the additional carbon source was only supplemented when the influent COD was relatively low (Zou and Lu, 2016). The only work with external COD amendment and a real environment is presented by Larrriba et al. (2020). They treated the effluent from the primary settler of a full scale WWTP by the mainstream SCEPPHAR configuration aiming at P-recovery in a demo scale and studied different strategies (such as nitrite pathway) to improve the efficiency of C usage in view of decreasing the carbon footprint and the operational costs.

This external carbon can also be obtained in a more sustainable way from the own internal plant resources through the fermentation of waste sludge, which is an environmental way to obtain the VFA required to boost anaerobic P-release. In the work of Kodera et al. (2013), an additional fermentation tank was conceptually proposed to obtain enriched VFA by primary sludge fermentation, which was supposed to provide enough COD for anaerobic P-release and subsequent P-recovery. Nowadays, the use of side-stream sludge fermenters to provide COD to fulfil the carbon needs for biological nutrient removal is becoming a trend (Ali et al., 2021; Li, Li and Li, 2018; Wang et al., 2019). Thus, it could also be used as a source for extra carbon dosage in view of P-recovery.

3.6. Effect of other operational parameters

Regarding the effect of other operational parameters on the observed performance, the main general recommendation would be to select them to ensure good PAO activity. The same conditions that would maintain PAO activity in a conventional SBR or A²O continuous system would be recommended in a mainstream P-recovery configuration. For example, (Baeza et al., 2017) reported the effect of SRT in an SBR configuration with P-recovery and found a minimum of SRT = 4 d was required to achieve complete P-removal, but to achieve maximum PHA production, SRT = 10 d was recommended. Therefore, for the specific wastewater composition used and the other particular experimental conditions (e.g. T, DO, pH, volume exchange ratio, operation objective) these results were obtained, but they are case-specific, and hence, for each scenario, they should be determined experimentally or using a proper modelling approach. In fact, mainstream P-recovery is adding an additional limitation to the typical needs that PAO microorganisms also require, such as a minimal T-dependent SRT (Brdjanovic et al., 1998; Chan et al., 2020b), enough DO and aerobic phase length, and typical aerobic and anaerobic HRT (Tchobanoglous et al., 2014).

3.7. Future outlook

P-recovery from the anaerobic supernatant of the mainstream EBPR process is a novel and promising process, as in addition to the advantages of other P-recovery configurations such as alleviating the problem of undesired P precipitation and recovering a valuable resource, it has additional advantages that help to reduce the COD requirements for P-removal, allowing the treatment of wastewaters with lower COD/P ratio, and maintaining high P-removal activity. However, most of the investigations concentrated on lab-scale SBR and almost all recovery processes were based on P crystallization.

In order to optimize these systems, the extraction ratio is the most basic and important operating parameter to achieve high P-recovery without impairing the long-term stability of the system. Anyway, proper simulation studies should be performed, as the operating parameters are case-specific and highly dependant on the operating objective.

To demonstrate that this is a reliable technology for P-recovery, future works should focus on continuous configurations, which seem more suitable for full-scale application, and using real municipal wastewater. The objective is challenging, as it is necessary to obtain an enriched stream with high P concentration to facilitate P-recovery, but without requiring the addition of an external carbon source that would compromise the economics of the process. Moreover, LCA and cost analysis should be used to demonstrate the overall positive effect of P-recovery systems in WRRFs. In any case, policy decisions will be of paramount importance for the implementation of P-recovery, as it has been shown that the costs are affordable, but like many environmental technologies, it is not cost-effective unless it is driven by policy (Nättorp et al., 2017).

4. Conclusions

This paper critically reviews for the first time the performance of the current reported strategies for mainstream P-recovery in EBPR WWTPs and provides the main points to be addressed for its successful implementation. Furthermore, it shows that full-scale mainstream P-recovery is a medium-term possibility.

- It is based on the PAO capability of producing a P-enriched supernatant that enables P-recovery, and SBR and continuous configurations at different scales have been reported.
- It can improve P-removal performance and enable a successful treatment of influents with a relatively low COD/P ratio.

- Configurations combining EBPR with mainstream P-recovery are still at an early stage and their successful full-scale application has not yet been reported.
- The most important parameter to describe these systems is the extraction ratio: the amount of anaerobic supernatant to be extracted or derived to recover P. The choice of this value is a compromise between i) having a low amount of P recovered with high PAO activity and anaerobic P concentration (i.e. easing precipitation) and ii) having a high amount of P recovered with a potential deleterious effect to PAO activity and low anaerobic P concentration.
- A maximum value for the P-recovery efficiency of 60% of influent P seems the most sensitive goal for a long-term operation of these systems. However, higher values can be used for sporadic or single extractions.
- Regarding the preferred agent for P-recovery, most of the works opted for struvite, while vivianite or HAP are less common at this point.

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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