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Review

Second-Generation Phosphorus: Recovery from Wastes towards the Sustainability of Production Chains

Camila Ester Hollas [1], Alice Chiapetti Bolsan [2], Bruno Venturin [1], Gabriela Bonassa [1], Deisi Cristina Tápparo [1], Daniela Cândido [3], Fabiane Goldschmidt Antes [4], Matias B. Vanotti [5], Ariel A. Szögi [5] and Airton Kunz [1,3,4,*]

[1] Center for Exact and Technological Sciences, PGEAGRI, Western Paraná State University, Cascavel 85819-110, PR, Brazil; camila.hollas@gmail.com (C.E.H.); brunoventurin583@gmail.com (B.V.); gabrielabonassa@gmail.com (G.B.); deisictapparo@gmail.com (D.C.T.)
[2] Graduate Program in Biotechnology, Universidade Tecnológica Federa do Paraná, Dois Vizinhos 85660-000, PR, Brazil; alice1bolsan@gmail.com
[3] Graduate Program in Environmental Science and Technology, Universidade Federal da Fronteira Sul, Erechim 99700-970, RS, Brazil; danielacandido@unochapeco.edu.br
[4] Embrapa Suínos e Aves, Concórdia 89715-899, SC, Brazil; fabiane.antes@embrapa.br
[5] Coastal Plains Soil, Water and Plant Research Center, USDA-ARS, Florence, SC 29501, USA; matias.vanotti@usda.gov (M.B.V.); ariel.szogi@usda.gov (A.A.S.)
* Correspondence: airton.kunz@embrapa.br

Abstract: Phosphorus (P) is essential for life and has a fundamental role in industry and the world food production system. The present work describes different technologies adopted for what is called the second-generation P recovery framework, that encompass the P obtained from residues and wastes. The second-generation P has a high potential to substitute the first-generation P comprising that originally mined from rock phosphates for agricultural production. Several physical, chemical, and biological processes are available for use in second-generation P recovery. They include both concentrating and recovery technologies: (1) chemical extraction using magnesium and calcium precipitating compounds yielding struvite, newberyite and calcium phosphates; (2) thermal treatments like combustion, hydrothermal carbonization, and pyrolysis; (3) nanofiltration and ion exchange methods; (4) electrochemical processes; and (5) biological processes such as composting, algae uptake, and phosphate accumulating microorganisms (PAOs). However, the best technology to use depends on the characteristic of the waste, the purpose of the process, the cost, and the availability of land. The exhaustion of deposits (economic problem) and the accumulation of P (environmental problem) are the main drivers to incentivize the P's recovery from various wastes. Besides promoting the resource's safety, the recovery of P introduces the residues as raw materials, closing the productive systems loop and reducing their environmental damage.

Keywords: waste treatment; struvite; chemical precipitation; biological recovery; nutrient recovery; phosphate

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1. Introduction

Phosphorus (P) has gained increasing attention in the world scenario in the last few years. The concern with the limitation of natural sources, associated with the constant demand beyond the environmental impacts, has attracted attention to this element [1,2]. Besides essential for life, the P has a fundamental role in the industry and the world food production system, directly influencing the economic sector [3].

The mineral extraction of P from phosphate rocks is the primary source of this resource, which configures the first-generation phosphorus, that is, the one that comes from rock mining. Considering that, the worldwide reserves of phosphate rocks are irregularly distributed worldwide, concentrated in countries like Morocco, Iraq, China, Algeria, Syria, Russia, and South Africa, which together hold 88% of world reserves, that, in turn, have been being strongly affected due to the high consumption of the resource [4].

The growing demand for food, boosted by population growth, also increased the agricultural sector's pressure to produce more, to meet global needs. Consequently, the agricultural sector is the one that most consumes this resource, with up to 80–90% of the P flow sent to this sector as a result of crop production expansion [5]. According to Ulrich and Frossard [6], the most optimistic projections estimate that natural sources of P would be depleted in 100 years. However, other estimates predict the depletion of natural reserves much earlier, in 50 years or less, which draws attention to the sustainability of this scenario, of limited sources and high demand for the resource.

According to a report released by FAO [7], with the future perspectives for the three main fertilizer (nitrogen, P, and potassium—PK), until the year 2022, the global supply capacity is estimated at 52,066 million tonnes, while the world demand for fertilizers is 49,095 million tonnes. As such, the balance is positive. However, if it is stratified by regions across the globe, the balance became negative for many regions, as for Latin America and the Caribbean (−5991 thousand tonnes), South Asia (−7859 thousand tonnes), Central Europe (−526 thousand tonnes), West Europe (−1519 thousand tonnes) and Oceania (−515 thousand tonnes). This scenario is worrying and very close since the positive balance is low.

Added to these factors, P's actual use in the agricultural sector is sometimes done inefficiently. This inefficient use of P ends up impacting environmental quality, promoting the eutrophication of water resources that drastically affects the aquatic biota of coastal regions, rivers, and lakes worldwide [8]. The accumulation of nutrients such as P in aquatic environments favors the proliferation of algae, leading to the formation of hypoxic dead zones and compromising the local environmental sustainability [9,10]. According to the United States Environmental Protection Agency (US EPA), the country has about 40% of streams or rivers damaged due to anthropogenic inputs of P and nitrogen [11]. In China, all 109 lakes under monitoring had some level of eutrophication, according to the "Report on the State of the Ecology and Environment in China 2017" [12]. Malone and Newton [13] highlighted that in Western Europe about 65% of the coastal waters of the Atlantic Ocean suffer some eutrophication while for the coastal region of the United States the amount reaches 78% of commitment. Bennett et al. [14] estimate that between 1 to 3.1 Tg yr^{-1} of P is accumulated in freshwater sediments worldwide.

In this sense, P recovery and recycling second-generation, that is, that which comes from waste becomes a fundamental element for sustainable management given the current resource's conditions, related to environmental contamination and supply risks [15]. Agriculture can recycle the P-rich effluents produced in the food chain, directing it to the production systems and concomitantly reducing supply risks and ensuring an economically P stable value [16,17]. This posture is possible since the world P market has undergone a significant change since 2008, with product prices rising by 800% in 14 months, contributing to incentivizing the search for alternative solutions [18]. In practice, in developed and developing countries, the P recovery is still a challenge and requires greater attention.

Although the routes for P recovery have been widely studied in the last few years, few works bring a broad analysis about the problem, considering spatial (geographical), temporal, and environmental issues for around the globe, including treatment alternatives at the generation source, avoiding costs with wastes transportation. Besides, P sources pollution and regulations for controlling the impact, and solutions are not commonly discussed in the literature. Thus, this review aims to evaluate and compile the methods used for second-generation P recovery from agricultural effluents by studying theoretical concepts and mechanisms related to recent results regarding physical, chemical, and biological methods.

2. Animal Wastes

Among the agricultural sectors, livestock production stands out, with growing yearly in the last 60 years, becoming more technologically advanced and industrialized due to the high demand for food boosted by the population growth and consumption patterns [19,20].

Adoption of animal confinement is a management strategy to meet maximizing production goals while intensifying the importation of nutrients to a small geographical area, but affecting the land's absorption capacity of nutrients [19,21]. As a result, the residue's beneficial use as organic fertilizer and soil amendments ends up generating an environmental liability, requiring their treatment, which is indispensable for the viability of agricultural activities [22].

Table 1 shows different characteristics particular to animal wastes. There is significant variability of these material's characteristics due to many aspects that can change animal waste composition, such as diet composition, environmental conditions, growth phases, among others [23,24]. However, N and P stand out among the nutrients present in animal wastes due to their high concentrations [1].

Table 1. Composition of different animal wastes (DM = Dry matter).

Reference	Animal Waste	TP ($g\ kg^{-1}$)	DM (%)	Organic C ($g\ kg^{-1}$)	Total N ($g\ kg^{-1}$)	C/N Ratio
Handin and Eriksson [25]	Horse manure	1.5	23.5	7.9	0.34	23.2
Szögi et al. [26]	Fresh swine manure	18.2	21.3	-	37.8	-
Lemming et al. [27]	Cattle manure	1.7	26	432	21	21
Li et al. [28]	Chicken manure	3.0	24.26	-	12	-
Li et al. [28]	Dairy manure	0.6	17.91	-	3.6	-
Grigatti et al. [29]	Bovine slurry	8.0	4.25	488.1	32.6	15
Grigatti et al. [29]	Swine slurry	33.0	5.16	394	46.5	8.5
Grigatti et al. [29]	Anaerobic digestate from bovine slurry and energy crops	6.2	5.05	515	43	12
Kunz et al. [30]	Swine manure	0.46 [a]	-	14.59 [a]	1.52 [a]	9.6

[a] $g\ L^{-1}$.

The possibility of recovering these nutrients draws attention to the sustainability in the productive chain and promotes food safety for future challenges regarding the resource's availability, with P being one of the critical elements in this perspective. Added to this, the environmental damage caused by the practice of indiscriminate land disposal has affected the environment all over the globe.

3. Global Challenges versus Local Solutions: Case Studies

A myriad of problems caused by P accumulation in the environment has been reported in the scientific literature for many years worldwide. However, only in the last decade, these problems aroused attention, and the growing demand of society for renewable sources of raw materials acts as a catalyst for recovery initiatives [31]. In the case of P, besides the exhaustion of natural deposits from where it is extracted, leading to an increase in price, the accumulation of the element in the soil and water of densely populated regions with tremendous industrial and agriculture activities, has caused great environmental and economic problems. In these regions, the nutritional need of crop plants for this limiting nutrient is overcome by the supplementation of chemical fertilizers and residues containing P, which is often made indiscriminately and unintentionally.

In short, the exhaustion of deposits (economic problem) and the accumulation of P (environmental problem) are the main drivers to incentivize the P's recovery from various wastes. Several studies worldwide reported methodologies and recommendations to predict how the soil is and when it will exhaust its capacity to retain P [32]. However, even in the regions where the contribution is made intentionally, studies that indicate which areas need P supplementation are not consistently wide and constant over the years. This mismanagement is the main reason for excess concentrations of P and its consequent environmental damage [33–35].

Phosphorus loads in an agricultural watershed depend not only on the soil use characteristics of this basin but also on the type and intensity of rainfall in the region, especially rainfall events soon after fertilizer application. Therefore, it is vital to improve soil, nutrient, and water management during the first planting [36]. However, not all storms will carry P to the water bodies, and seasonal P limits must be created.

Zhang et al. [37] proposed a model to evaluate separate Agricultural Non-point Source (AGNPS) models for three diverse small agricultural watersheds in southern Ontario, Canada. The model was calibrated for runoff volume, sediment yield, and total P and executed for representative storms with increasing return periods (2-year through 100-year). The results showed that a spring storm with return times ranging from 4.8 to 14.9 years tends to generate P loads in water bodies above the environmental limit for the region. In all watersheds, summer storms with return times of up to 100 years did not reach the P's seasonal tolerance. The researchers also reported that the critical areas were evenly distributed across all the basins studied.

These diffuse sources of agricultural P are related to several eutrophication cases from Lake Tai in China to Lake Erie in North America to the Baltic Sea in Northern Europe to Rotorua lakes in New Zealand, generating several environmental and economic damages [35]. In the USA, it is estimated that the eutrophication of freshwater bodies costs USD 2.2 billion per year in services spent [38]. In comparison, the costs of remedying the problems caused by eutrophication for England and Wales are estimated to be around USD 160 million per year [39].

Over the last three decades, several cases reported on the situations of productive and densely populated areas around the globe from the perspective of environmental problems caused by P accumulation [40–43].

Behrendt and Boekhold [40] estimated that the accumulation of P surplus from the balance of agricultural soils in industrialized countries of Europe in the 1950s to 1980s was already between 800 and 1500 kg ha^{-1}, although this distribution was not uniform. In areas with intensive cattle-raising, P accumulation was more than an order of magnitude of the average of the surrounding soils. By 1993, about half of the Dutch sandy soils (approximately 300,000 ha) were already considered saturated with P.

Jia et al. [41] reported that peri-urban intensive livestock farming's rapid growth has led to excessive P loads in some regions of China. The researchers assessed the potential for the distribution of P from manure on arable land in the Beijing area, excluding soils with a slope above 25% and land designated as aquatic buffer zones. The results showed that manure generation in 2011 resulted in P's surpluses, with an average input rate on arable land of 53 kg P ha^{-1}. On the outskirts of Beijing, the authors estimated that 11% of arable land was unsuitable for continuous applications of P-containing waste due to the high levels already contained in the soil, which exceeded the environmental risk limits for P. Under traditional management, soils used for growing vegetables and orchards would be saturated before 2030, just like the soil with grain cultivation near the environmental threshold also around 2030. Researchers have suggested that increasing the proportion of manure applied to cereal fields could reduce the annual surplus of P, which would alleviate the accumulation of P in orchards and fields used for growing vegetables. Balanced P management strategies could keep P levels in the soil much lower, i.e., 24, 22, and 27 mg kg^{-1} in soil used for growing cereals, orchards, and vegetables, respectively. Exporting surplus manure out of Beijing's peri-urban regions was considered one of the main strategies to minimize the environmental risk of P [41].

Sharma et al. [42] reported increased P levels of Northeast United States soils. Historically, potato planting is considered a significant contributor to P contamination in water due to its high sensitivity to P and low absorption (25–30 kg ha^{-1}). Although agronomic recommendations in the region are at a rate of 56 kg ha^{-1} P, producers tend to apply fertilizers at a rate of 182 kg ha^{-1}, to compensate for eventual leaching losses. Another reason for P's excessive application is its fixation to soil, which is known to make it unavailable for crops in the short term. Phosphorus application in non-reactive sites, natural soil variability and pH changes over the decades have also been reported as other possible reasons for the increase in P levels in the soil, resulting in its erosion into water bodies.

However, there are studies aimed at evaluating the management and use of P produced and recovered globally. Weikard and Seyhan [43] have studied the effects of P recycling in developed countries on global rock phosphate extraction and imports from developing countries. The authors advocate that developed countries should explore new technologies for P recycling and build a resource extraction model for a competitive fertilizer market. This model reflects that most developed countries have soils saturated with P. In contrast, soils in many developing countries are deficient in P. Such countries differ in the demand and recycling options that should be adopted and encouraged by their governments.

It was found that recycling P in developed countries would extend the life of the resource globally and increase the share of the resource in developing countries, providing greater food security [43].

In a study conducted by Powers et al. [44], they analyzed the subnational, national, and global P recycling potentials, emphasizing densely populated areas and waste-rich cultivated areas. Populated cultivated areas were less abundant globally than manure-rich cultivated areas, reflecting the greater segregation between crops and people, especially in the Americas. In countries such as India, China, Southeast Asia, Europe, and parts of Africa, disproportionately large portions of sub-national areas with P recycling potential were identified. In conclusion, the abundance of potential areas for P recycling in nations that depend on imported fertilizers may help to develop local sources of P and maintain agricultural independence [44].

Hanserud et al. [45], in their case study for Norway, already pointed out that p values from animal manure and sludge have the theoretical potential to meet the all phosphate fertilizer needs of all Norwegian crops, assessed from both a short and long term perspective. Animal wastes generally contain large amounts of P and therefore represent promising sources for P recovery, both economically and environmentally [46]. These factors bring attention to the public sector about the emergency of political incentives for P-recovery.

4. Environmental Legislation: Global Trends

In recent years, effluent discharge limits to receiving waters imposed by regulatory agencies from several countries are becoming more stringent, owing to increased awareness and growing concerns regarding pollution and water resource degradation [47]. Since P release due to anthropogenic activity promotes eutrophication in aquatic ecosystems, it has detrimental consequences for aquatic life and the domestic and industrial water supply [48–50].

The Brazilian legislation that governs conditions and standards for managing the effluents' discharge into receiving water bodies is CONAMA Resolution number 430/2011. However, it does not provide specific discharge limits for P. Some states have specific laws that establish maximum concentrations for release into water bodies, such as the states of Santa Catarina (4 mg L^{-1}), Rio Grande do Sul (1–4 mg L^{-1}), and Rio de Janeiro (1 mg L^{-1}) [51–54].

To protect the quality of Europe's water resources, the European Union (EU) has adopted several norms and laws in environmental protection, which have imposed a set of quality standards with clear objectives for the protection of human health and the environment [55]. The effluent discharge limits in Stockholm (Sweden) restrict the concentrations of total P in treated wastewater to less than 0.3 mg L^{-1}. However, these requirements are projected to decrease to 0.2 mg L^{-1} in the next few years, with the operational effluent target set to 0.15 mg L^{-1} [47].

In the United States, the total P requirements established according to the National Pollutant Discharge Elimination System varies between 0.1 to 1 mg L^{-1}, and the most restrictive requirement found was 0.05 mg L^{-1} [56,57].

A significant focus of the Federal Water Pollution Control Act has been to improve surface water quality through control of point source P inputs from wastewater treatment plants to waterbodies. Bashar et al. 2018 [58], in a review of US EPA's Discharge Monitoring

Report, showed that Massachusetts (0.1 mg L^{-1}), New Hampshire (0.2 mg L^{-1}), Michigan (0.3 mg L^{-1}), and Maryland (0.3 mg L^{-1}) have the lowest total P (TP) permit limits for one or more Publicly Owned Treatment Works (POTWs).

While P is one of the most pollutant elements in water, it is also an irreplaceable and non-renewable resource that supports global food security [59]. Phosphorus has been classified as a critical raw material in the EU because its production worldwide is concentrated in few countries. This concentration is in many cases composed of low substitutability and low recycling rates like Morocco, for instance, that controls 75% of the remaining world's P reserves [60,61]. Studies estimate that the recovery of P from wastewater would satisfy 15 to 20% of its global demand [59].

Despite the already consolidated legislation regarding the limits for effluents' discharge, the current scenario has encouraged countries to legislate on the recovery of nutrients and establish discharge limits. The communication "Towards a circular economy: A zero waste program for Europe" lists specific waste challenges that are related to significant loss of resources or environmental impacts [62].

Countries such as Austria, Germany, and Switzerland have now made P recovery mandatory from municipal sewage sludge and other activities, with future perspectives for expansion and goals to be achieved [48,63]. According to Jupp et al. [15], P recovery's difficulties are that the recovered material is considered a waste, and of difficult transportation and commercialization. Therefore, the legislation still needs to evolve to change the scope from waste to high value-added products, considering the recovered P as a second-generation product.

The idea of closing the cycle and changing the way of thinking from legislating to control pollution to recovering a valuable resource is one of the main challenges to be put into practice to change the current reality and promote the recovery of P from waste. Making the scope more comprehensive by including agricultural flows and not only sanitary waste also need attention in these legislations. Encouraging the use of recovered P should be a strategy to be adopted by countries. In this sense, the technologies for P recovery deserve attention to help meet these goals towards the closure of cycles and sustainability of production chains [3,15].

5. Phosphorus Recovery Processes

There are several technologies aimed at the recovery and/or removal of P from waste. Chemical precipitation, thermal treatment, sorption and/or ion exchange, membrane-based separation by nanofiltration, electroflocculation, electrocoagulation; and some biological processes, such as enhanced biological P removal(EBPR). Examples of these technologies include one or a combination of physical, chemical and biological processes [64–66]. Among the available processes, precipitation with magnesium or calcium and the crystallization of struvite (magnesium ammonium phosphate) or hydroxyl apatite are the most applied methods in animal wastewater treatment systems due to the relatively high concentration of calcium, $NH4^+$ and PO_4^{3-} already present in these effluents. Consequently, depending on the stochiometric combination of Mg, NH_4^+ and PO_4^{3-} or Ca, PO_4^{3-} can be crystallized either struvite or hydroxylapatite (Figure 1). Common to the precipitation processes, the P will be recovered in solid form and the purity of the recovered P material will be dependent on the wastewater type and characteristics [61,65,67].

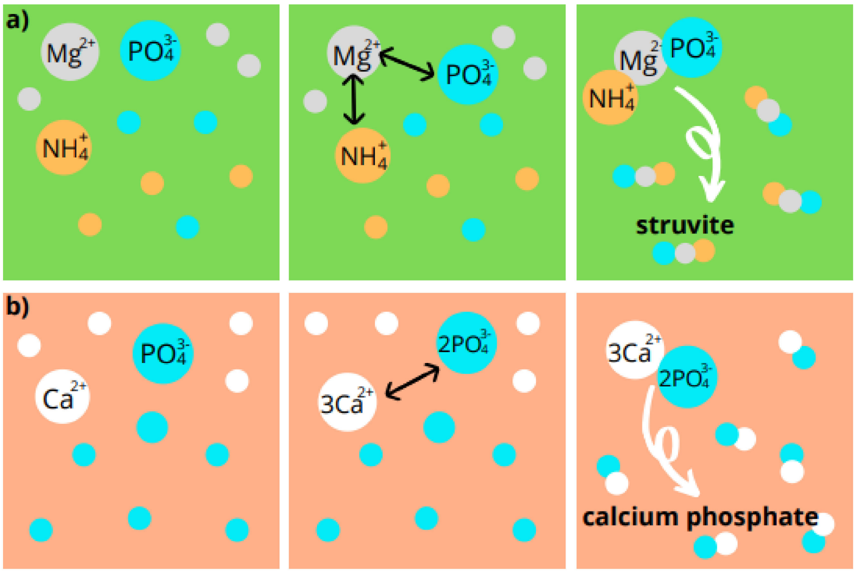

Figure 1. Illustration of the chemical precipitation process for P recovery. (**a**) struvite; (**b**) calcium phosphate.

5.1. Chemical Precipitation

The P removal by calcium compounds is an easy and low-cost alternative to be implemented in treatment plants, based on the application of hydrated lime as a calcium source to precipitate it. On such, the soluble phosphate present in the digestates is converted to an insoluble form, as calcium phosphate ($Ca_3(PO_4)_2$), hydroxyapatite (HAP; $Ca_5(PO_4)_3OH$)) and/or brushite (DCPD; $CaHPO_4$), which are crystalline equivalents [68–70].

The different chemical species of calcium phosphates that the P chemical precipitation can obtain are dependent on the process reaction conditions, such as pH, temperature, supersaturation level and Ca/P molar ratio. According to the literature, this kind of process's efficacy can reach up to 90% in wastewater treatment plants with the addition of hydrated lime [$Ca(OH)_2$] to reach a pH of at least 8.5. Among the advantages of the chemical precipitation, the generated sludge can be used as P-fertilizer, which is microbiologically safe, since the high pH ($\geq$8.5) also has the ability to inactivate pathogens [70–72]. Equation (1) represents the chemical reaction of PO_4^{3-} with the addition of $Ca(OH)_2$ [73].

$$3\ Ca(OH)_2 + 2\ PO_4^{3-} \rightarrow Ca_3(PO_4)_2 + 6\ (OH)^- \tag{1}$$

According to the stoichiometry described by Equation (1), the Ca/P ratio required P chemical precipitation is 1.5 (3 mols of Ca^{2+} per two mols of PO_4^{3-}). However, other ionic P forms ($H_2PO_4^-$ and HPO_4^{2-}) allow different Ca/P ratios for the chemical precipitation process. Additionally, the process can undergo pH instability, which can lead to the formation of several compounds with different solubilities (pK_s). The increasing order of solubility for the most common compounds is: sintered hydroxyapatite [$Ca_{10}(PO_4)_6(OH)_2$] (pK_s = 116.8); octacalcium phosphate [$Ca_8(HPO_4)_2(PO_4)_4.5H_2O$] ($pK_s$ = 96.6); tricalcium phosphate [$Ca_3(PO_4)_2$] (pK_s = 28.9); dicalcium phosphate anhydrous [$CaHPO_4$] (pK_s = 6.90); dicalcium phosphate dihydrate [$CaHPO_4H_2O$] (pK_s = 6.59) and amorphous calcium phosphate [$Ca_xH_y(PO_4)_{zn}H_2O$] (solubility cannot be measured), which are formed with Ca/P ratios between 1.3 to 2 [74,75].

Referring to the applicability of the chemical precipitation on animal wastewaters treatment, according to Suzuki et al. (2005) [67], this is an excellent alternative to P removal from swine manure digestates due to the high concentration of PO_4^{3-} and the presence of Ca (~210 mg L^{-1}) would reduce the amount of added hydrated lime. However, the swine manure also has high concentrations of ammoniacal nitrogen and alkalinity, impairing P precipitation because these species also react with hydrated lime, as well as the presence of organic matter in swine manure co-precipitates with hydrated lime and the P accumulated in the produced sludge can show high concentration of impurities [71,76]. In addition, alkaline metal ions (potassium and sodium), transition metals (iron, copper, and zinc), among others, generate phosphate or hydroxide salts, and at the alkaline conditions compete directly for phosphate ions, co-precipitating and reducing product purity. Although aluminum and iron are used for P precipitation, the toxicity of aluminum to plants and the low bioavailability of iron-bound P make them not recommended for P recovery [77]. Therefore, pretreatment of swine manure is essential to reduce organic matter, alkalinity and nitrogen compounds that may interfere with the process of P precipitation with calcium [76].

In this sense, Suzin et al. [76] compared the efficiency of P removal from different swine wastewater stages (after anaerobic digestion and biological nitrogen removal), using hydrated lime for the chemical precipitation of P . As expected, better efficiency was obtained with the effluents with reduced organic matter, alkalinity and ammoniacal nitrogen (after nitrogen removal process), since this effluent also showed Ca/P close to stoichiometry.

Fernandes et al. [71] evaluated the chemical treatment for P removal from swine wastewater using $Ca(OH)_2$. They found that the higher the pH (8.5 to 10.5), the greater the removal P efficiency obtained (90 to 98%). According to their results, 92% of total P was present as soluble P, and 75% of the soluble P was recovered as phosphate in the sludge. The sludge obtained by the chemical separation contained considerable amounts of amorphous calcium phosphate according to X-ray diffraction spectroscopy results, indicating good potential for fertilizer use.

Among the chemical precipitation technologies, Quick Wash (QW) is a process developed for the recovery of P from solid animal waste in a concentrated solid form, having as a differential the acid extraction of P before the precipitation phase. QW is patented (US Patent 8.673.046) and is based on P's chemical recovery through a combination of acid, hydrated lime, and organic polyelectrolytes [76]. The QW process consists of three stages: (1) extraction, which occurs through acid hydrolysis with a pH ranging from 3.0 to 5.0. The QW produces a liquid extract containing low suspended solids content and a soluble P in this stage; (2) after the extraction of P from the manure solids, the second stage comprises P recovery by precipitation with the addition of hydrated lime to reach a pH in the range of 9.0–11.0, thus obtaining a P precipitate product rich in P and Ca. In the third stage, the addition of an organic anionic polymer facilitates precipitation, increasing P's concentration in the product, having value as a fertilizer rich in P [52].

Szögi et al. [77] evaluated the dredged sludge of a swine manure anaerobic lagoon through P recovery's QW process. They verified that at pH 10.0, the recovered P corresponded to 79% of the total P present in the sludge, being the precipitated product identified as amorphous calcium phosphate with low heavy metal content, an advantage of this technology, besides the low operating costs which makes it an attractive technology for P recovery from anaerobic lagoon dredged sludge.

5.2. Crystallization of Struvite

In wastewater treatment systems, the crystallization of struvite ($MgNH_4PO_4.6H_2O$) as an alternative process to P recovery occurs in the presence of Mg^{2+}, $NH_4{}^{+}$ and $PO_4{}^{3-}$, being the treatment costs of wastewater mainly linked to the cost of magnesium chemical costs (contributes up to 75% of overall treatment cost) since this element is present in low concentration and must be added to satisfy the reaction stoichiometry for struvite precipitation. The mineral struvite is a white crystalline substance composed of magnesium, ammonium, and phosphate, and according to pH and ion concentration, it can precipitate in an aqueous solution. It shows low solubility in water, reducing the runoff capacity and improving the plant uptake, when applied as fertilizer [78,79].

To precipitate the P as struvite, magnesium, ammonium and phosphate (100–200 mg L^{-1}) are required at equi-molar concentrations (1:1:1) at alkaline conditions (pH ~7.5) and adequate mixing [80]. At such ideal conditions, from each 100 m^3 of wastewater, around 1 kg of struvite can be recovered as a crystallized material [81]. Struvite forms according to Equation (2) [82,83].

$$Mg^{2+} + NH_4^{+} + H_nPO_4^{n-3} + 6H_2O \rightarrow MgNH_4PO_4 \cdot 6(H_2O) + nH^{+} \quad (n = 0, 1 \text{ or } 2) \qquad (2)$$

The advantages of this P recovery route are linked to the commercial properties of the struvite, which can be used to produce fertilizers, food additives, chemical agents, structural products and can also be applied as an adsorbent for ammonia removal. Compared to other routes the wastewater treatment system's costs are also reduced since the sludge volume generated is reduced around 49% compared to biological removal [80,81,84].

Due to the presence of some inhibitory species in the effluents of wastewater treatment systems, such as Fe, Ca, and Al, along with the inherent heterogeneity and instability of the characteristics of these, sometimes a pretreatment step before the struvite crystallization is necessary to maximize the P recovery yield (i.e., anaerobic digestion, electrocoagulation, chelating and microwave treatment). According to the effluent concentrations of P and NH_4^+, the supplementation of such compounds can also be necessary for effluents from the dye, textile, tanning and fertilizer industry. However, urine and effluents from cattle, swine and poultry wastes usually rule out the required supplementation of P (NaH_2PO_4, KH_2PO_4, H_3PO_4) and NH_4 (NH_4Cl) due to the better balance between the contents of the three components of struvite [80].

As mentioned, the crystallization of P as struvite occurs in the presence of stoichiometric amounts of magnesium. Despite the advantages and high purity and reactivity, the use of commercial magnesium salts ($MgCl_2$, $MgSO_4$ or MgO) represents onerous costs to the wastewater treatment process. The costs with this chemical reagent can contribute up to 75% of overall production costs but others cited some low-costs magnesium source alternatives to struvite crystallization [79,80].

The use of low-costs magnesium sources can minimize the overall struvite crystallization costs by around 18–81%. These renewable alternative materials vary according to the availability, reactivity, and solubility of the Mg sources. According to the solubility, classified as high-solubility Mg sources (i.e., mainly compound by $MgCl_2$, $MgSO_4$) or low-solubility (i.e., mainly composed by MgO, $Mg(OH)_2$ and $MgCO_3$), seawater, brucite, magnesite, struvite pyrolysate, wood ash, and desalinated reject water are some potential low-costs magnesium sources industrial byproducts for struvite crystallization [80,85,86].

Wang et al. [85] tested commercial $MgCl_2$, $MgSO_4$, MgO, $Mg(OH)_2$, and low-solubility bittern as Mg sources to recover struvite from swine wastewater. With operation conditions of pH 8.5 and an Mg/P ratio of 1.5, the authors did not find differences in P removal and product quality between the different Mg sources. Such results proved the feasibility of using alternative Mg sources to reduce the process costs.

In addition to the properties of high magnesium content in ashes from biomass, the ashes' alkaline properties provide a cost-effective alternative for the struvite crystallization process. Huang et al. [86] evaluated the use of straw ashes as an Mg source for struvite crystallization and P recovery from swine wastewater. Under controlled pH conditions, they obtained a struvite material with properties comparable with those obtained using commercial Mg.

As already mentioned above, the pH is a crucial factor in struvite production. The pH directly affects the efficiency of the process since it influences the reaction's solubility and supersaturation. The P recovery and struvite crystallization increase as the pH increases in a range from 7 to 11.5 [82]. According to Li et al. [87], a pH over 9.0 promotes efficiency of over 90% in the process.

Moulessehoul et al. [82] studied the process of struvite crystallization over a wide pH range (8.5, 8.7, 9.6, 10.5, and 11.8), using a synthetic solution with equimolar proportions of ions Mg^{2+} ($MgCl_2$), NH_4^+ and PO_4^{3-}, at 25 °C. They obtained the best crystallization conditions at pH 8.5, with concomitantly higher P removal and better quality of the precipitate obtained as struvite crystals.

Shaddel et al. [79] evaluated the use of seawater as an Mg source (1276 mg L^{-1} Mg^{2+}) and tested different pH conditions (7.5, 7.7 and 7.9) for treating biological waste sludge from a wastewater treatment plant, including P removal by struvite crystallization. The seawater performed as an adequate magnesium source, and the pH was the critical factor for struvite crystals formation. The removal efficiency and struvite crystal growth rate increased according to the increased pH.

Accordingly, the suitable pH range for efficient P removal and high struvite purity is mainly dependent on the type of wastewater in question, which shows different concentrations of magnesium, P, potassium, ammoniacal nitrogen, sodium, and other compounds; and the magnesium source, which can be commercial or alternative. Thus, some gaps and challenges should be scrutinized before applying the struvite crystallization technology in wastewater treatment pilot plants, to avoid compromising the process's feasibility and economy [87,88].

Another route of precipitation gaining scientific interest is the K-struvite or magnesium potassium phosphate hexahydrate ($MgKPO_4 \cdot 6H_2O$). The process to obtain K-struvite is analogous to the process of formation of struvite, having the NH_4^+ replaced by potassium (K). The K-struvite has physical and chemical properties similar to struvite, and is considered an excellent fertilizer source since the K and P are essential nutrients [83].

K-struvite precipitation has as ideal conditions highest at a pH around 10.0 (within a pH range of 9.0–11.0) and a molar ratio of Mg:K:P of 2:1:2. that ensures removal efficiencies of 77% of P and 98% of K from wastewater. [83,89,90]. Tarragó et al. [91] found that the temperature also had an influence on the precipitation of K-struvite from animal manure, being the temperature of 38 °C the most favorable for the formation of crystals. The presence of other ions such as Ca^{2+}, NH_4^+, and Na^+, can affect the formation of K-struvite crystals due to the low competitive capacity of K in comparison to these ions. The low concentration of Mg and K in the residues can be a limiting factor for the process to meet the ideal molar ratio for precipitation and crystal formation [92].

Urine is one of the main wastes being studied for the precipitation of K-struvite. Huang et al. [89], Xu et al. [92], Wilsenach et al. [93], investigated the precipitation of K-struvite from urine because urine has high P concentrations that allow the process to be simpler and more economically efficient. Nevertheless, the removal or recovery of ammonia from urine is one of the essential factors for the technology's success since it can interact with Mg and P to form K-struvite [93]. With this, K-struvite can be a promising solution as a fertilizer source of P and K, recovered from waste, and ensuring the stocks of these mineral resources.

5.3. Newberyite Recovery

An alternative approach to struvite precipitation is the production of newberyite ($MgHPO_4.3H_2O$). In this New-P process [94], P recovery is combined with ammonia recovery using gas-permeable membranes (Figure 2). In a first step, the ammonia and carbonate alkalinity are removed from digested livestock wastewater using low-rate aeration and a GPM manifold. The N removal is done with low-rate aeration in the reactors that naturally increases the pH of the liquid and facilitates subsequent P precipitation without the need for alkali chemical supplementation. In a second step, the P is removed using magnesium chloride ($MgCl_2$). After ammonia recovery, the effluent is low in ammonia, low in carbonate alkalinity, and has a higher pH. In turn, these conditions improve the precipitation of phosphate minerals of high-grade. The combined New-P process [94] applied to digested swine manure provided 97–98% N recovery in one product and quantitative (ca 100%) P recovery in another product. The preliminary removal of the soluble ammonia destroys carbonate alkalinity. As a result, the phosphates produced contain high P_2O_5 grade (37–46%) and low N, similar to the composition of the biomineral newberyite [94,95]. Vanotti et al. [95] reported high-grade phosphates produced from anaerobically digested swine manure using this process having high P and Mg content and low concentration of N: approximately 46% P_2O_5 (20% P) and approximately 17% Mg, and 1.8% N (Ca and

K content were generally low, 0.4% and 1.7%, respectively). The resulting molar ratio of the P product was 1.0:1.1:0.2:0.0:0.1 for P:Mg:N:Ca:K, respectively, like the composition of the biomineral newberyite ($MgHPO_4.3H_2O$) found in guano deposits, which has approximately 18% P and 14% Mg and 1:1 P:Mg molar ratio.

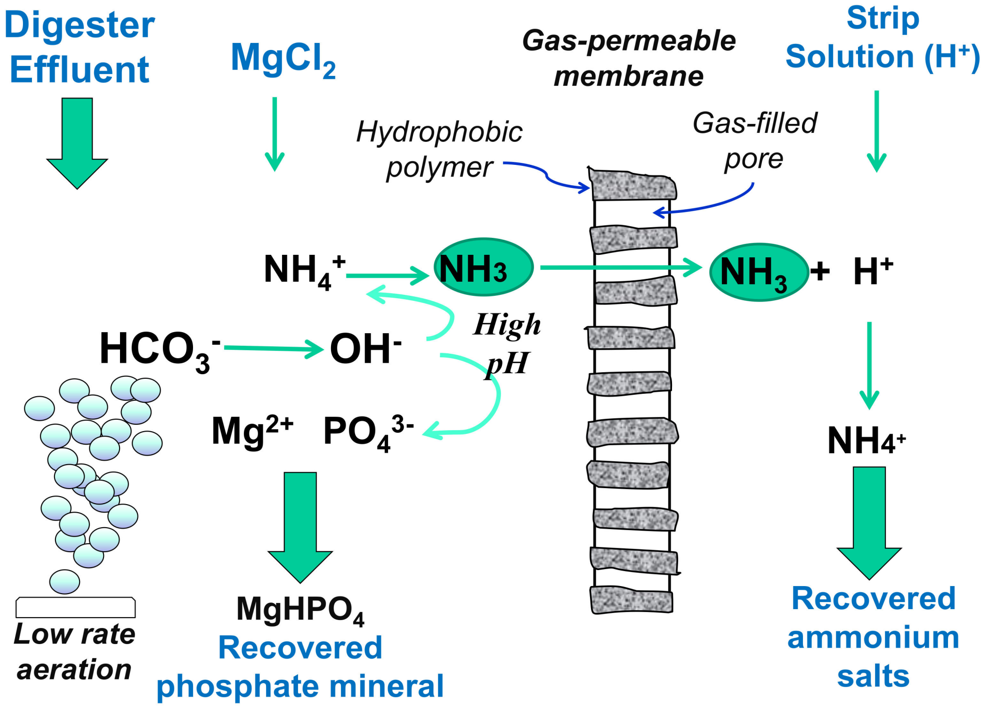

Figure 2. Schematic diagram of recovery of Newberyite using Mg precipitation in a—P process [94].

5.4. Thermal Treatment

The thermal treatment is a P recovery alternative route applied for the treatment of sludge, wastewaters and digestates from agro-industrial treatment plants; and sludges from other P recovery routes (i.e., sludge generated from chemical precipitation process) [96,97]. Thermal treatments include combustion, incineration, pyrolysis, gasification, and hydrothermal carbonization processes (Table 2). These technologies for P recovery reduce organic pollutants and pathogens, enabling the reuse of energy accumulated in wastewater or sludge and generate other products with lower weight and final volume, which can be easily stored and transported [98,99].

Table 2. Different P-rich byproducts obtained by animal waste thermal processes.

Reference	Residue	Thermal Process	Temperature (°C)	Product	N	P	K
					$g\ kg^{-1}$		
Leng et al. [100]	Poultry litter	Combustion	850	Ash	-	97.8	148
Bergfeldt et al. [101]	Poultry litter	Combustion	815	Ash	-	73.0	89.2
Więckol-Ryk et al. [102]	Chicken manure	Combustion	500–900	Ash	-	125.6	130.0
Lang et al. [103]	Swine manure	Hydrothermal carbonization	180–220	Hydrochar	20.2	20.7	-
Song et al. [104]	Swine manure	Hydrothermal carbonization	160–240	Hydrochars	28.5	26.0	8.0
Song et al. [105]	Swine manure	Hydrothermal carbonization	140–220	Hydrochars	44.8	18.8	1.7
Nest et al. [22]	Swine manure	Pyrolysis	500	Biochar	17.0	1.2	67.1
Novak et al. [106]	Poultry litter	Pyrolysis	500	Biochar	40	31.5	69.4
Liang et al. [107]	Dairy manure	Pyrolysis	450	Biochar	-	25.2	-

After the thermochemical process, the P remains in the mineral phase, it can be in the form of ash, for combustion processes; already in the case of other thermal processes in absence of oxygen such as pyrolysis, the P will be in the charcoal type of compounds, called biochar, or hydrochar, in the case of hydrothermal carbonization. The ashes contain up to 20–25% of P_2O_5, as well as K, Si, Mg, Al, and heavy metals, in concentrations according to the waste stream. In relation to biochar or hydrochar, this presents high efficiencies of P recovery from waste (>70%), with concentrations of P_2O_5 that vary according to the waste [104,108].

The recovered P in some of the byproducts from thermal treatment is readily available as plant nutrients and in use as fertilizer. Otherwise, P is extracted and recovered following various protocols using acids (i.e., citric, oxalic, H_2SO_4, HCl, HNO_3 and H_3PO_4), alkalis (i.e., NaOH, KOH), supercritical fluid extraction, and wet oxidation. These recovering techniques can render P_2O_5 concentrations around 78% in the recovered P byproducts [108–110].

Adam et al. [108] evaluated the thermochemical treatment of sewage sludge for P recovery. After the thermal treatment, they observed the P's high bioavailability, being the ashes suitable to be used as a fertilizer source.

Meng et al. [111] proposed low-temperature combustion as a recovery technique to enrich the P content in sludge ash. Such ash was derived from the dewatered sewage sludge thermal treatment for P recovery by combustion. After the treatment of the ashes, the bioavailable P content increased 2.9 times (45.6%). This alternative had higher P yields in recovered P than combustion, pyrolysis.

Before thermal processing, it is necessary to dewater the sludge or digestate it to at least 35% moisture, making the process non-self-energy sufficient. Additionally, some of these routes also demand gases during the combustion (i.e., nitrogen for dry pyrolysis), increasing the costs of the P recovery. The application of this kind of process is relatively restricted to the economic feasibility and energy demands of these routes [110,112].

In this sense, hydrothermal carbonization is configured as an emerging technology that can reduce the energy needed and the cost to dry the feedstock prior to thermal treatment. Hydrothermal carbonization is a process that uses high-pressure hot water as the reaction medium, with relatively low temperature (180–350 °C), producing hydrochar, which has been considered a promising product from the energy (as fuel), agronomic (as fertilizer), and chemical (as adsorbent) points of view [113].

5.5. Nanofiltration (NF) Membrane Separation

Nanofiltration (NF) comprehends a pressure-driven filtration that occurs on a membrane-based material. Due to the dense active layer, these membranes contain charged functional groups on their surfaces with different ion selectivity and low pore diameters (<2 nm). Thus, only compounds with low molecular weight are retained on these membranes' surface, like natural organic matter and nutrients [114–116].

During the process, due to the use of hydrostatic pressure, it is possible to separate large particles or high molecular substances of the digestates from the low molecular weight substances, like PO_4^{3-}, which can pass through the membrane and be retained on it. One of the significant highlights of this type of process is linked to the membrane's high selectivity, retaining only the compounds of interest (Figure 3) [61,117].

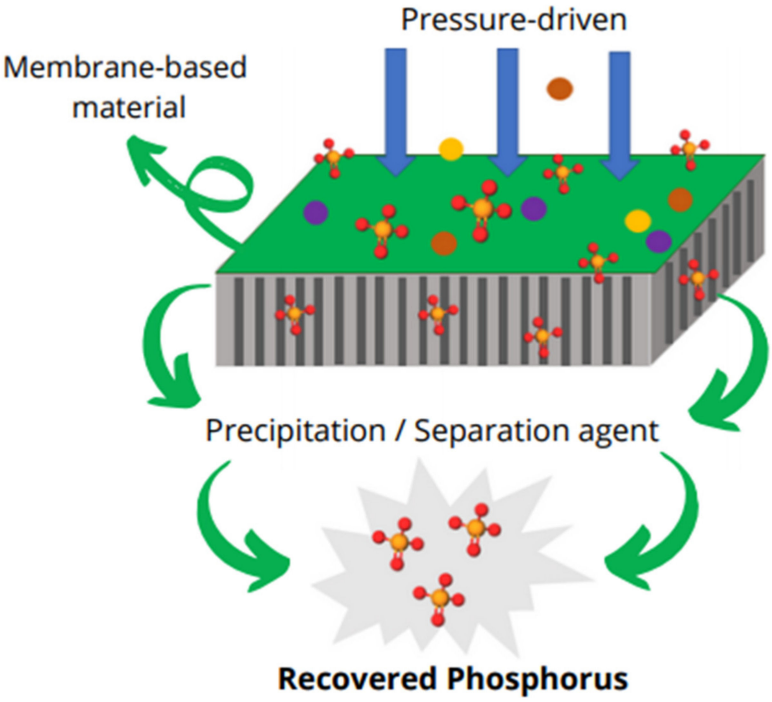

Figure 3. Illustration of membrane-based separation by nanofiltration (NF) for P recovery.

Some studies proved that the NF could promote yields of 89% on P recovery from wastewaters [118] or as high as 97–98% [110]. Such efficiencies depend mainly on the NF membrane surface, which needs to be specifically selective for phosphate (i.e., deposited by polycations and polyanions) [119].

After the retention of the P compounds on the membrane surface by NF, some post-techniques can be applied to precipitate or separate it from the liquid waste, for example, the addition of magnesium oxide to precipitate the phosphate [120], precipitation as calcium phosphate [121], electrodialysis process [114], diafiltration [115]. Besides the need to combine the nanofiltration with another process to remove the recovered P from the membrane surface, the properties of the membrane materials a key to avoid reducing the membrane's permeability, lifetime, lixiviation, fouling and biofouling that may compromise the applicability of such technology to treat animal wastewaters [117,122].

Yue et al. [123] studied the removal of nutrients from swine waste using three different membrane processes. The authors found that nanofiltration associated with reverse osmosis was the technology that showed greater stability and efficiency, with a nutrient recovery of 61.9% for total nitrogen, 56.3% for total phosphorus and 67.0% for potassium. Fouling is the main limiting factor for the use of this technology in animal waste. According to Adam et al. [124], membrane fouling leads to high operational costs for membrane regeneration, coupled with high energy costs due to the high working pressure, which makes the use of the technology a challenge.

5.6. Adsorption by Ion Exchange

Different materials can be considered efficient for PO_4^{3-} removal. According to their surface energy and physical–chemical characteristics, available materials, can be: natural minerals (i.e., mineral, zeolites, rock, shell), industrial byproducts (i.e., fly ash, steel slag and sludge) and synthesized products (i.e., organic polymer adsorbents and biochar from different sources), for example. Among the widely investigated alternatives for P recovery from wastewaters, adsorption by ion-exchange showed relatively low implementation costs than other treatment options. Concurrently, the costs of some of these materials depend on the source and synthesis conditions required to obtain them [125–127].

According to Liu et al. [128], among the many sources that can be used as materials to phosphate adsorption, the preference is for those ones contained by silicate functional groups. Active sites like La^{3+}, Fe^{3+} and Ca^{2+} in these materials also contribute to P recovery. Phosphate selectivity and affinity allow the adsorbent to capture PO_4^{3-} due to the ion competitions in wastewaters. Examples of adsorbents with the previously cited active sites are the ferric oxide nanoparticles, commercial HAIX resins (that contain Fe^{3+}) and La^{3+} oxide embedded anion exchanger (commercial La-201) [129].

Selective adsorbents can be used to achieve PO_4^{3-} removal (by adsorption) and recovery (by desorption). Lian et al. [127] evaluated the synthesis and use of a low-cost Fe-loaded sulfoaluminate cement (FSC) adsorbent for P removal and recovery avoiding water remediation. The authors obtained positive results that prove the potential of the material to treat wastewaters containing PO_4^{3-}, but further studies are necessary for understanding the practical application of FSC.

Using calcium silicate hydrates (CSH), Lee et al. [65] evaluated the possibility of P recovery from sewage sludge, also analyzing the subsequent use of the final product as fertilizer. The CHS promoted high P recovery efficiency and the final product had 43.1% of available phosphate to be used as fertilizer. The applied route highlights the advantages of the adsorbent material used and the obtained product.

Zhang et al. [130] studied the adsorption of P from swine manure onto wheat straw biochar with the addition of hydrated ferric oxide to improve the P adsorption selectivity. The authors also applied a microwave pretreatment to improve the P solubilization in manure, obtaining biochar with 51.71–56.15 mg g^{-1} of adsorbed P.

One of the drawbacks of large-scale and commercial applications of the adsorption technique for wastewater treatment is separating the adsorbent from the adsorbate for efficient desorption, regeneration, and P recovery from the adsorbent material surface. This stage counts for up to 70% of the total costs of adsorption technologies [10].

5.7. Electro-Based Technologies: Coagulation and Flocculation

Phosphorus recovery from wastewaters can be achieved using electrochemical technologies such as electro-coagulation (EC) and electro-flocculation (EF). The process is based on the induction of PO_4^{3-} precipitation as amorphous calcium phosphate, hydroxyapatite, or struvite, using an electrical current. According to reports, these processes are fast, simple, eco-friendly, easily operable, and generate less sludge than the chemical routes [125,126,131,132].

The principle of EC and EF is based on the formation of OH^- ions and H_2 gas at the cathode, in parallel to the dissolution of metal cations at the anode. Different materials can be used as anodes (Fe, Al, Al, Mn, Cu, Zn—alloy or not), which plays a significant role during the P oxidation and precipitation as sludge. Al and Fe are the commonly applied electrodes due to the low price, non-toxic characteristic, and P removal capability (Figure 4) [132,133].

Zhang et al. [134] evaluated the EC in swine manure treatment for P removal, achieving an efficiency of around 81.3%. Emerick et al. [135] also applied the EC process in the treatment of swine slaughterhouse wastewater, using Al and Al/Fe electrodes. The efficiencies in P removal were around 79 and 75% with such an approach, removing color, turbidity, nitrogen, and biochemical oxygen demand. The efficiency and feasibility of the process correlated with some operational parameters such as pH, type of supporting electrotype and current density [136].

Omwene et al. [137] evaluated the EC potential using aluminum and plate hybrid and no single anodes to phosphate removal from domestic wastewater. The phosphate removal efficiency achieved was >99% using the hybrid Al-Fe plate electrodes, a feasible and economical alternative for wastewater treatment. In the same way, Nguyen et al. 2016 [138] investigated EC's use for P removal from municipal wastewaters. They used different voltages, conductivity, and electrolysis operating times to evaluate total P removal and sludge production to optimize the process and lower treatment costs. With the concurrent combination of these factors, the process had the most outstanding efficiency and cost savings.

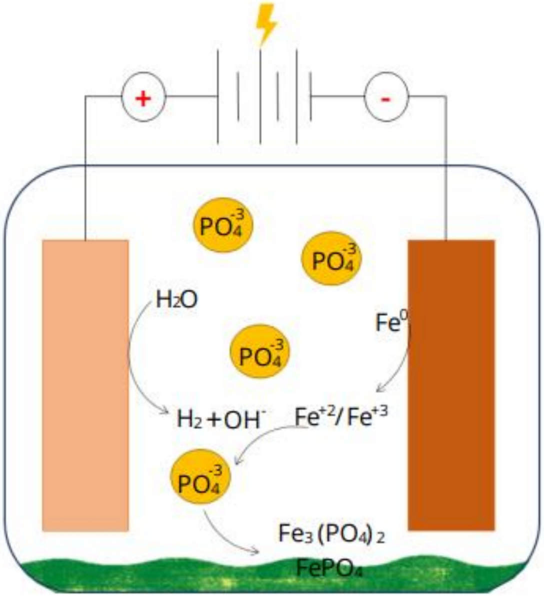

Figure 4. Representation of the electroflocculation/electrocoagulation process with Fe anodes for P recovery.

Mores et al. [139] assessed the effects of operating time, current density, and initial pH for P removal from swine wastewater pretreated by anaerobic digestion in UASB reactor, by the EC process using Al and Fe electrodes. Both electrodes showed the same efficiency for P-removal (96%). However, the operational costs of using the Al ones were considered advantageous in terms of the operation time, and no addition bleach material to the final treated wastewater effluent.

Also, EC is attractive to remove several other pollutants beyond P by combining EC to degrade a wide range of compounds (i.e., organic matter, heavy metals, pathogens, ammoniacal nitrogen, pesticides) to ensure greater efficiency. The EC technologies can combine Fenton, ozonation, photolysis, photocatalysis, photo-Fenton, or electrochemical oxidation processes, among others. The choice to integrate these processes depends on the type of wastewater and treatment goals [136,137].

Among the electro-based alternatives that can maximize the efficiency and applicability of the EC process, combining it with electrooxidation has been widely investigated. The electrooxidation route is based on an anodic reaction using metal oxide electrodes (Ti/PbO_2, Ti/SnO_2, RuO_2-IrO_2/Ti) that forms an intermediate oxidant (Cl_2, HClO or ClO^-). This intermediate reacts with the nutrients, organic matter, pathogens (or the several compounds present in wastewater) for their removal [140,141].

In such a way, Mahvi et al. [142] evaluated the simultaneous removal of ammonia and phosphate from wastewater effluents based on electro-based technologies that match the EC with the electrooxidation process. The process proved to be advantageous for treating effluents with higher concentrations of ammoniacal nitrogen, phosphates, and removed some other pollutants such as organic matter simultaneously. Chackchouck et al. [143] also obtained higher P removal efficiency when combining both processes (87% = EC; 97% = EC + electrooxidation).

5.8. Biological Processes

5.8.1. Phosphorus Recovery by Composting

Composting is another biological route for P recovery from wastewaters. It consists of aerobic waste decomposition through microorganism activity, resulting in stabilized and sanitized material. The product of composting is a valuable fertilizer, rich in nutrients and humified organic matter. In addition, this technology presents advantages in the reduction of volume, humidity and odors, and the final product is solid, which facilitates its transport in comparison to liquid effluents [144,145].

The microbial decomposition of organic matter uses carbon and nitrogen primarily as energy sources rather than P. Thus, the P remains in the final product together with other macronutrients (nitrogen and potassium), micronutrients (copper, zinc, iron, and manganese), heavy metals (lead, cadmium, and chromium), and microbial consortia (protists, fungi, oomycetes, yeast, actinomycetes, and bacteria) [141–143].

According to Wan et al. [144], high P contents in the plant-absorbable P, available P, and citric acid P fractions can be obtained in the final product, depending on the initial waste composition and mineralization process rates. Solid manure, chicken wastes, sewage sludge, and commercial food processing waste are excellent composting sources due to the high C/N ratio, P content, and moisture [17,53].

After the composting process, the P fraction present in digestates and wastewaters in a general shift from organic into more stable fractions. Qin et al. [146] mixed the use of the excess sludge from wastewater treatment plants (rich in organic matter content, nitrogen, and P) with food waste to form a co-composting system. The sludge contributed to enriching the compost in terms of available P content, resulting in a fertilizing amendment product with greater concentrations of P in the HCl-P and NaOH-P fractions.

According to Mieldažys et al. [147], the generation of large quantities of composts could replace the worldwide use of phosphates mineral fertilizer in up to 30%.

5.8.2. Microalgae

Microalgae can also be applied as a biological process to wastewaters treatment since it acts as biocatalysts for nutrients (P and N) and organic matter removal. Some microalgae species (i.e., *Chlamydomonas reinhardtii* and *Pseudokirchneriella subcapitata*) can promote N and P removal higher than 98 and 82%, respectively, from municipal wastewater. Therefore, the increased efficiency in P removal with microalgae's processes allows nutrient recovery and reuse of the harvested algae biomass (Table 3). Many works of research have proved the feasibility of using microalgae in wastewater treatment as an alternative approach for nutrients removal from municipal, agricultural, animal, and industrial wastewaters [148–151].

Table 3. Different P-rich algae biomasses obtained from animal waste.

Reference	**Residue**	**Microalgae**	**Biomass Productivity ($mg\ L^{-1}\ d^{-1}$)**	**Biomass Concentration**	
				TP ($g\ kg^{-1}$)	**TN ($g\ kg^{-1}$)**
Cole et al. [152]	Fish farm wastewater	*Oedogonium* sp.	24–36 [a]	3.4	-
Li et al. [153]	Digestate Chicken manure	*Chlorella* 1067	251	18.75 ± 2.78 [b]	85.11 ± 3.52 [b]
Pandey et al. [154]	Dairy wastewater	*Chlorella vulgaris*	19.6	66.3 [c]	156.4 [c]
Juárez et al [155]	Digestate swine manure	*Scenedesmus obliquus* (39%), *Scenedesmus lagerheimii* (33%), *Scenedesmus opoliensis* (13%), *Scenedesmus magnus* (4%)	-	3.8 [a]	63.8 [a]
Sobhi et al. [156]	Digestate chicken manure	Indigenous microflora "Heterotrophic"	410–3690	32–42 [a]	-

[a] in relation to the Dry weight (DW). [b] in relation to the Total solids (TS). [c] in $mg\ L^{-1}$.

Cultivation of microalgae on wastes provides several advantages over other treatment methods for P recovery (i.e., chemical precipitation, crystallization of struvite, electron-based processes). The effective P assimilation by algae can lead to a reduced cost of P removal from wastewater. The harvested algae biomass has the additional value of its byproducts such as pharmaceuticals, cosmetics, biofuels, fertilizers, and animal feed. Microalgae production reduces greenhouse gas emissions because of no need for fossil fuels and synthetic fertilizers their production [150,151,157–159].

Microalgae efficiently utilize the wastewater's nutrients to increase their mass, metabolic function, and cell growth. These organisms require high amounts of N, P, organic matter, solar energy and CO_2 for protein, nucleic acid, and phospholipid synthesis. According to their molecular formula, around 0.063 g of N and 0.009 g of P are needed to accumulate 1 g of biomass. The N/P ratio requirement can vary according to the microalgae species for each treatment; thus, other researchers consider a range of approximately 6.8–10 as the optimum N/P ratio to P removal [151]. However, the unbalanced N/P can lead to low P removal efficiency by microalgae and consequently, the ratio should be as close as possible to the optimum, to promote microalgae growth and ensure the highest P-removal efficiency [151,159,160].

Scendesmus obliquus proved to be efficient for N, P removal and lipids accumulation from swine wastewater [161]. Ammonia and phosphate from soybean processing wastewater were successfully utilized for biomass productivity increment of *Chlorella pyrenoidosa* ($0.64\ g\ L^{-1}\ d^{-1}$), where, the microalgae promoted N and P removal of up to 89 and 70%, respectively, with high lipid productivity ($0.40\ g\ L^{-1}\ d^{-1}$) [160].

Chlorella vulgaris and *Chlamydomonas reinhardii* were evaluated in P removal from treated leachate. Both organisms were suitable for the removal of N and P, being phosphate stored intracellularly in the microalgae. This intracellular capacity to store P can be suitable for the subsequent agricultural use of the microalgae biomass as fertilizer [162]. *Desmodesmus communis*, *Tetradesmus obliquus* and *Chlorella protothecoides* were studied by Lavrinovics et al. [163] for enhanced P removal from municipal wastewater. All species were suitable for P removal (up to 89%) and to store it as polyphosphate in their cells. The process was highly dependent on the pH, N/P ratio and organic carbon concentration since these affected the metabolic functions and biomass growth.

Li et al. [164] evaluated *Coelastrella* sp. for P removal in swine wastewater under various Zn (II) concentrations. Besides the mentioned effect of pH, N/P ratio, and organic matter concentration, the presence of Zn (II) enhanced P assimilation by *Coelastrella* sp. and removal from swine wastewater by 77.6%. According to Cheng et al. [165], several microalgae species have excellent P removal efficiency, such as *Chlorella Scenedesmus* sp., *Neochloris* sp., *Chlorellaceae* sp. and *Coelastrella* sp. Because several high-value bioproducts obtained from their biomass (e.g., pigments, polysaccharides, lipids, proteins, biofuels, bioelectricity, pharmaceuticals, cosmetics) have a significant commercial interest, the microalgae treatment of swine wastewater is a sustainable approach in the circular economy context [166].

5.8.3. Enhanced Biological Phosphorus Removal

Enhanced biological P removal (EBPR) is attracting daily more attention due to the feasibility of its feature of simultaneous P and N removal. It is an alternative to P removal without chemicals, which operates in anaerobic and aerobic stages in sequence in the wastewater treatment plants. The process consists of the intracellular polyphosphate (poly-P) bioaccumulation by phosphate-accumulating organisms (PAOs) (assimilation and de-assimilation processes) under aerobic and anaerobic conditions. In the anaerobic stage, the PAOs take up volatile fatty acids (VFAs) from the wastewaters, which store as polyhydroxyalkanoates (PHA). Subsequently, it is metabolized at the aerobic stage to provide the phosphate accumulation process's energy by glycogen-accumulating organisms (GAOs). Up to 45–75% of the EBPR costs are related to the aerobic stage's oxygen requirements [167–169].

The most common PAOs organisms are from the genus *Candidatus* Accumulibacter. The GAO organisms include *Candidatus Competibacter*, *Candidatus Contendobacter* cited as the common genus *Defluviicoccus* [170]. However, some algae also are included in the EBPR process [48]. Since GAOs require organic matter for successful P, combining the EBPR with the partial nitritation-anammox (PN/A), which both organisms do not require any organic source to N removal. for simultaneous N and P removal from digestates [171].

Yang et al. [172] proposed the simultaneous EBPR followed by the PN/A to enhance nutrients removal from municipal wastewater. The two SBRs reactors combined: SBR1 for P removal, operating with anaerobic–aerobic conditions (ammonia remained constant); and SBR2 for N removal, operating with aerobic–anoxic conditions. The simultaneous P and N removal resulted in an N removal efficiency higher than 81%, while P and organic matter removal efficiencies were 95% 82%, respectively.

Wang et al. [167] evaluated chemical oxygen demand (COD) simultaneous effect, N and P concentration on the EBPR process, and PAOs and GAOs organisms. The efficiencies achieved were up to 91, 90, and 84% for COD, P and N, respectively. *Candidatus* Accumulibacter was an essential PAO organism involved in the assimilation and de-assimilation processes; *Dechloromonas* had potential as an electron acceptor during the anoxic conditions; and *Thauera* and *Zoogloea* were related to the denitrification routes.

Yuan et al. [171] developed a two-stage process for simultaneous nitrogen and N removal by EBPR-PN, followed by Anammox. The process's feasibility was proved as an alternative route in treating wastes with a low C/N ratio, with removal efficiencies for N and P of 86 and 95%, respectively. The dominant genus related to P removal was found to be *Pseudomonas* (29.04%), related to the PAOs organisms.

Animal waste due to its high concentrations of nutrients and organic matter, can be a challenge for the application of EBPR, especially in terms of competition for substrate with other organisms, however, some studies demonstrate the applicability of this technology in these effluents. Liu et al. [173] studied EBPR, in the removal of P from liquid dairy manure, and obtained good results, with efficiencies that ranged from 56.8 to 73.5% for P removal, demonstrating the applicability of the technology to animal waste.

Since the phosphate-accumulating microorganisms are heterotrophic (GAOs), the process requires enough bioavailable carbon. Thus, the utilization of wastes with considerable levels of organic carbon (i.e., animal wastes) is advantageous. It is also necessary an electron acceptor, which can be nitrate or oxygen, for the PAOs organisms. Besides that, cations such as magnesium or potassium are essential, contributing to phosphate assimilation and dissimilation. In the anaerobic phase, altogether magnesium, potassium, and P are released, while during the aerobic phase these three elemental species are assimilated [174].

6. Technology Comparison, How to Choose Most Appropriate?

As presented, there is a great diversity and under constant development technologies for P recovery. Choose the appropriate process is a challenge, since all the technologies show some advantages and disadvantages. The final product may have different characteristics and recovery rates as a function of the chosen process.

Beyond that, biological processes in general, such as composting, are configured as the most economical alternatives. However, from the environmental point of view, the excessive release of greenhouse gases can limit this technology [175]. Other biological routes, such as the use of algae and EBPR, present disadvantages in terms of the strict control of the process and pre-treatment strategies. Thus, these routes can be expensive and complicated to be applied for agricultural treatment plants [176,177].

Some technologies present high energy costs, which also limit their application. This is the case of membrane-based processes, electron-based technologies, and thermal treatment. Mores et al. [178], studied electrocoagulation for removal of P from swine manure and obtained high efficiency in the removal of the resource, higher than 90%. However, the energy expenditure reached 17.2 kWh L^{-1}, which makes the process onerous from the point of view of recovery and feasibility of the practice for agricultural systems. These technologies present high efficiencies in P recovery, and technological advances are necessary to make these processes feasible. In this sense, Adam et al. [124], demonstrated that dynamic nanofiltration combined with low-pressure reverse osmosis, can be performed with low energy consumption in digested animal waste, the process was optimized reaching the consumption of 11.6 kWh ton^{-1}, which presents opportunities for this sector, which needs advances for the technological development of these processes.

Phosphorus deficiency is the main constraint to agricultural productivity, so the products resulting from the P-recovery should emphasize those that can efficiently replace commercial fertilizers. These allow the closure of the nutrient loop in the agricultural sector. In this sense, P recovered by chemical precipitation, either in the form of calcium phosphate or struvite, stands out. The slow-release capacity of P from struvite, associated with the simultaneous availability of N, makes it a powerful fertilizer, with less risk of losses to the environment due to this low solubility [179].

The energy and input costs of the P recovery technologies mentioned above can be mitigated by combining them with other treatments. In addition to P recovery, energy recovery is an important element for closing the energy cycle of agricultural chains, improving the sustainability and viability of these treatment systems. All these factors mean that the simple choice in terms of economic feasibility is not the only decision parameter. According to Egle et al. [180], the choice of process is difficult and should match the benefits of economic, environmental, and technological spheres; whereas Puchongkawarin et al. [181] complements that the social sphere also must be considered as a deciding factor since the impacts will directly affect society. Sarvajayakesavalu et al. [3], pointed out that the choice of the best process could be a complicated process and nutrient balance, for soil application, and water quality requirement, for water reception bodies must be considered. Thus, to meet all these environmental, social, and economic criteria involved in the decision, life cycle analysis can be a valuable tool to compare these technologies and assist in the selection process.

7. Life Cycle Assessment of the Recovery Systems and the Circular Economy

In the literature, it is possible to find a different range of studies on the sustainability of P recovery by analyzing process life-cycle assessment [182]. Some of these researches report the environmental benefits of P recovery and the associated impacts with this practice, as can be observed in Table 4. Although the comparison of the evaluations is difficult due to the different approaches, functional units, and methodologies of analysis used in each study, it is possible to identify the behavior of P recovery beyond the activity impacts [183].

Table 4. Examples of studies on the environmental feasibility of P recovery using the life cycle approach, with emphasis on climatic or global warming potential (GWP) and eutrophication potential (EP).

Reference	Waste	Recovery Way	Evaluation Methodology	Functional Unit	Impact Category	
					GWP (kg $CO_{2\ eq}$)	EP (kg $PO_{4\ eq}$)
Lijó et al. [184]	Livestock waste	Struvite	ReCiPe Midpoint H	1 t of treated manure	−22	−0.001
Rashid et al. [185]	Municipal wastewater	Struvite	CML-IA	1 m^3 of treated wastewater	7.47×10^{-15}	1.62×10^{-14}
Peters and Rowley [186]	Sludge of municipal wastewater treatment	Biosolids	Carbon Footprint	2 dry tonnes per day of sludge	−490	-
Fang et al. [187]	Municipal wastewater	Algal biomass	ILCD 2011	1 m^3 of treated wastewater	1.2×10^{-2} mPE *	-9.2×10^{-2} mPE *
Nakakubo et al. [188]	Food waste and sewage sludge	Dry granulation	Carbon Footprint	Unit as the processing capacity to provide disposal services for 100,000 people	1000	-
Temizel-Sekeryan et al. [189]	Dairy Manure	Struvite	TRACI 2.1	1 kg of struvite produced	7.02	8.0×10^{-2} [a]
Luo et al. [190]	Swine manure	Compost	CML 2	Annual production of a typical pig farm in Beijing area (1956 LU annually)	5.611×10^{6}	3.41×10^{4}
Struhs et al. [182]	Cattle manure	Biochar	CML-IA	50 metric tons of manure	8642	0.28
Bora et al. [191]	Poultry Litter	Biochar	IMPACT 2002+	1000 kg of fresh or wet poultry litter	657	−4.34

* mPE = presented in milli-person equivalents (mPE), where 1 mPE represents one thousandths of an average European person's annual impact. [a] kg $N_{eq.}$

Even though the P recovery by several routes is favorable in terms of resource security, these technologies' sustainability must be guaranteed. This means these recovery processes should not promote greater impacts than the conventional ones for obtaining P [191]. The implementation of recycling practices plays an important role in reducing resource dependency, promoting the positive circularity of production chains, and inserting wastes as by-products of the recycling processes [182]. In this way, the life-cycle assessment is a robust tool for investigating the impacts that recycling processes can cause in the environment [183].

The studies evaluated different impact groups (Table 4). However, the categories of global warming potential (GWP) and eutrophication potential (EP) are common in most studies. The results for these categories differ in the function of the technology used, the type of waste, the study's scope, among other aspects, which may be optimistic having a net cost that consequently entails a superior environmental impact concerning the reference scenario. When negative, represents benefits in relation to the reduction of the environmental impact [192].

Rashid et al. [185] conducted a life cycle study of the implementation of a P recovery process to an existing treatment system for the treatment of sewage effluents, being the costs associated with adequacy and operation of the system one of the most critical points in the studied scenarios. Despite the environmental gains related to nutrients recovery, mainly in reducing the potential for global warming (7–22%), the process's costs could significantly burden the treatment system (an increase of up to 24%).

The results showed by Pedizzi et al. [193] for the life-cycle assessment of livestock manure treatment by anaerobic digestion showed that the scenario of digested material disposal in the soil, without any additional treatment, was the most favorable in comparison to the chemical P recovery from the digestate. As such, the energy expenditures were also responsible for these values. However, the authors highlight the importance of evaluating the environmental issues beyond the limits of the systems, which in most works is limited to the treatment system, mainly due to the limitations of land disposal that these wastes present, and subsequently environmental risks associated with these land disposal practices. The P recovery allows the export of these materials to more distinct regions where P is scarce and needed to sustain crop production [194,195].

Both works demonstrate that not only the environmental aspects must be considered, but also the economic ones. Many of the existing treatment systems worldwide are not capable or prepared for resource recovery from effluents because of being projected only for the removal of certain compounds. Consequently, they are limited in terms of environmental safety due to the absence of nutrient recovery [185,193]. In some studies, developed by Tonini et al. [196], they verified that the P recovery from different sources promoted gains from an environmental point of view and also social and economic in comparison with the mining process.

Wu et al. [197] verified an environmental gain of 40% within a swine manure composting base scenario. In this scenario, the proposed system's recoveries considered the use of algae biomass for nutrients recovery and replacement of feed for pigs, combined with energy recovery by anaerobic digestion and composting of the solid phase. Lijó et al. [184] noticed a 9-fold reduction for climate change and a 210-fold change for eutrophication. The scenario considered the P recovery by struvite from cattle waste compared with the anaerobic lagoon as the base scenario. Most of the published works attribute that the P recovery systems' main impacts are due to the impacts associated with the operation of the process, such as energy costs and chemical additives. In this sense, Golroudbary et al. [198] emphasize that the problems with the P recovery's actual routes are related to the low efficiency and emergency in improving such technologies. The authors observed that the manure's P was responsible for 82% of the recovered P, considering a worldwide scenario. In the treatment of domestic wastewater, it was responsible for 70% of the greenhouse gases in the P-life cycle, which correspond to 2% of the global recovered P. This study points out the importance of promoting P recovery from animal wastes.

The environmental efficiency of recovery processes improves when more than one nutrient can be recovered, such as P and N. Temizel-Sekeryan et al. [189] simultaneously recovered P and N from bovine manure. They verified the excellent environmental performance to replace conventional fertilizers with the recovered struvite (rich in P and N). Besides that, 78% of the eutrophication potential was also reduced. The production of N-fertilizer (Haber–Bosch) has a high environmental impact. Thus, the technologies that involve N recovery end up being an environmental benefit [195]. Although P-recovery technologies are still a challenge, the chains' circularity is extremely dependent on these recovery processes. It is an essential step towards the sustainability of world food production [199]. Security of resource supply through recovery is a crucial point in productive chains. According to Huygens et al. [200], this recovery has a capacity to meet up to 17–31% of the demand for P-based fertilizers in Europe. It demonstrates the processes' capacity and the necessity for improving these technologies for their environmental and economic viability.

8. Future Perspectives: Technical Implications for Process

The worldwide trend is the waste biorefinery that comprehends the extraction of compounds with high added value from wastes, promoting the circularity of supply chains ensuring the sustainability of these processes [2]. This view from removal to the recovery of the materials is recent, under construction, and underdevelopment worldwide. Consequently, with such, the creation and improvement of new technologies are critical, searching for the optimization of the processes by reducing the consumption of water, energy, chemicals, and transport, with a view in an advanced logic of complete nutrient recovery from the wastes, with low cost and high efficiency [1,182,184]. In this sense, recovery technologies have been gaining prominence in waste biorefinery scenarios. These scenarios show advantages, such as energy and nutrients recovery, products with high added value, high process efficiency, and low cost. However, some of them are also related to the excessive release of gases (i.e., composting and combustion), high energy expenditure (i.e., electrochemical processes), high costs with chemical additives, difficulty in controlling the biological process, among others. All these drawbacks directly influence choosing the appropriate resource recovery route [21].

Resource recovery and export requirements must be combined with the efficiency of their use. Thus, providing the ability to be directly applied to different areas, such as livestock production (as a food supplement), industry (as a biochemical feedstock), or crop production (as plant fertilizer) [2]. Among these practices, recovered-P's agronomic use is an essential point for the feasibility of its recovery. Agriculture is the sector that most consume these resources and the recovered nutrient (such as struvite, sludge, or calcium phosphate). Thus, the P must be plant available so that the replacement of commercial fertilizer could be adequate [201].

According to some research, the P-recovered fertilizer potential is similar to commercial phosphates fertilizer, such as triple superphosphate. Christiansen et al. (2020) [202] verified that the yields of barley biomass fertilized with P-recovered from wastes in different kinds of soils were similar to triple superphosphate. The P apparent recovery varied between 0–30% in such products (struvite, biochar, and ashes), which was very similar to the commercial one, with variations from 26 to 31%.

Nest et al. [22] evaluated the efficiency of P obtained using several techniques for nutrient recovery from animal wastes. They verified efficiencies of 71% for biochar, 79% for compost, 98% for struvite in comparison with triple superphosphate (100%). As such, the fertilizer solubility in water is the determining factor for the efficiency of the use of P by plants. The struvite is in the form of crystals of low solubility, which provides slow phosphate liberation and concomitantly, the plant is more efficient in using P since the low solubility ends up reducing the P fixation to the soil. The other recovery alternatives tend to form a higher amount of soluble compounds, therefore, promoting lower efficiency [203,204].

The viability of these technologies is optimized with the hybridization of the systems, combining recovery with biological, physical, and chemical treatments, thus maximizing the recovery of nutrients and energy. However, a method's choice depends on several characteristics, such as waste composition, availability of resources, purpose, or composition of the product, among others. Government incentives are still incipient and need to advance to encourage the use of these processes and public awareness initiatives for the acceptance that these materials can replace the traditional commercial resource [3].

9. Conclusions

The recovery of nutrients from wastewater has proved highly beneficial, both in environmental and economic terms. Reduction of dependence on inorganic compounds, derived from phosphate rock is one of the main ways to maintain viable human activities in the future. The circular view of the chains has an opportunity to close loops in recovery since technologies have a promising scenario, with gaps for improvement and optimization of processes to maximize efficiency and minimize costs. The choice of the ideal recovery process is relative, according to the type of waste its characteristics and the location where it is being generated. In addition to this, public policies are still necessary to promote the recovery of nutrients, towards a sustainable future reducing the environmental impact caused by P reported around the globe.

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Abbreviations

Phosphorus (P); Dry matter (DM); Nitrogen (N); Total phosphorus (TP); Enhanced biological phosphorus removal(EBPR); Quick Wash (QW); Nanofiltration (NF); Calcium silicate hydrates (CSH); Fe-loaded sulfoaluminate cement (FSC); electro-coagulation (EC); electro-flocculation (EF); poly-phosphate (poly-P); volatile fatty acids (VFAs); polyhydroxyalkanoates (PHA); glycogen-accumulating organisms (GAOs); polyphosphate accumulating organisms (PAOs); nitritation-anammox (PN/A); chemical oxygen demand (COD); global warming potential (GWP); eutrophication potential (EP).

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