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SYSTEMATIC MAPPING OF PHOSPHORUS RECOVERY FROM INDUSTRIAL WASTEWATER

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Abstract

Phosphorus is an essential nutrient used in fertilizers and food production but excessive levels in hydrological bodies of water can cause environmental issues. In a geochemical cycle, phosphorus is considered non-renewable, which by itself would be sufficient justification for its recovery from industrial wastewater. The objective of the present work was to conduct a literature review and present a panorama of current wastewater recovery methods. The review focused on the specific conditions of each technique, possible uses of the recovered element and major worldwide locations where this subject was studied. To this end, a systematic mapping technique was used with previously established empirical criteria. Publications from 3 scientific databases were scanned, resulting in 132,551 initial results and, through a rigorous filtering process, 81 studies were selected for review. In the selected studies, physicochemical recovery techniques were the most cited. An increase in studies was observed in 2016, when the number of publications doubled with respect to the preceding year and onwards. Worldwide, this subject is studied the most in Europe and Asia.

Keywords: industrial wastewater, phosphorus, recovery, systematic mapping
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Introduction

Every element or chemical compound necessary for the development of a living organism is considered a nutrient. As such, phosphorus (P) is considered one of the main essential nutrients for life. It is a vital component of nucleic acids, a fundamental chemical in energy-producing adenosine triphosphate (ATP) molecules and is found in phospholipids of cellular membranes (Quevedo and Paganini, 2011). Phosphorus is also used in the manufacturing of fertilizers for food production (Atienza-Martínez *et al.*, 2014).

Phosphorous has 2 known biogenic cycles: a short and a long cycle. The short cycle, also known as the biological cycle, happens when a part of the phosphorus atoms are recycled by the soil and decomposers, considering phosphorous as a renewable resource. In contrast, the long cycle, also known as the geochemical cycle happens when another part of phosphorus atoms is sedimented and incorporated into the rocks, therefore this cycle considers phosphorous a non-renewable resource. It is in this context that recovery from industrial wastewater is justifiable since excessive phosphorous levels can cause negative environmental consequences, such as eutrophication of the receiving hydrological body (Correll, 1998).

Shortfalls of natural deposits of phosphate minerals are projected in the future and current known deposits are heterogeneously distributed: the largest deposits are found in Morocco, United States, China, South Africa and Jordan (Nakakubo *et al.*, 2012; Kataki *et al.*, 2016). In the other hand, other studies estimate extractable phosphate minerals reserves to last between decades and hundreds of years (EFMA, 2000). Approximately 82% of all extracted phosphorous is used in agriculture (Sørensen *et al.*, 2015; Cordell *et al.*, 2009), which makes it a well-known strategic nutrient for food production and whose demand increases alongside population growth. There is no substitute nutrient to phosphorous in nature, which makes recovery techniques particularly desirable (Adam *et al.*, 2009). The European Union is highly dependent on phosphorous imports and is subjected to market fluctuations. For example, in 2008 the market price of phosphate minerals rose 800% (Schröder *et al.*, 2010). While the lifetime of current phosphorous reserves may be debatable, yields have been known to decline. This combination of yield, availability, geopolitical distribution of reserves and elevated market cost are sufficient driving factors for the research and development of new recovery techniques (Sørensen *et al.*, 2015; Biswas *et al.*, 2009).

Phosphorous recovery processes vary between physicochemical and biological methods. Physicochemical methods usually incorporate phosphorous in particulate suspensions for filtering while biological methods make use of accumulating micro-organisms to produce phosphates (Faria, 2006). This work reviews the most commonly used recovery techniques applied specifically to industrial wastewater. Additionally, data regarding the volume of publications classified by geopolitical region and suggestions for further research in the area are also presented. This task is accomplished through a systematic mapping methodology, which allowed the filtering of prime results according to main reference works. This allows the construction of a general view of the subject and even identifies possible shortfalls and areas in need of further work (Petersen *et al.*, 2015).

This work is organized in 7 sections. Section I consists of the introduction. Section II presents other related studies which followed similar mapping techniques. Section III describes the methodology used in this work and presents the mapped studies organized according to the survey questions and scientific databases. Section IV presents the finds obtained with respect to each search criteria and analyses the mapping. Section V deals with validation of the methodology. Section VI presents a discussion of the mapped studies and Section VII contains the conclusions and suggestions for further work.

Related studies

Systematic mapping yielded 81studies presented in more detail in Section VI. Among these studies, Cieslik and Konieczka (2017) and Roy (2017) were of particular interest since they are directly related to the present work. The study conducted by Cie and Konieczka in 2017, presented a review of phosphorous recovery techniques applied along multiple wastewater treatment stages under the perspective of “no waste generation.” In addition to liquid wastewater recovery, Cie and Konieczka also examined recovery from sludge ash and outlined analytical methodologies to determine the chemical composition of the ash. The study of Roy (2017), presented a survey of the global phosphorous cycle, its recovery and recycling techniques.

The review focused in recycling systems based on ecological technologies with attention to the general human impact of the phosphorous cycle. Taken together, Cie and Konieczka (2017) and Roy (2017) presented different but equally effective methods of phosphorous removal or recovery and were relevant to the present work. However, while both studies focused in a more general vision of phosphorous recovery from both industrial and non-industrial wastewater, the present work focused exclusively in industrial wastewater recovery. While Cie and Konieczka (2017), Roy (2017) and other review works can be found, few systemic mapping studies have been conducted on this specific subject. Consequently, the present work is a step forward and presents a more comprehensive review capable of directing further research topics.

Methodology

Systematic mapping was used in order to obtain more expressive results with fewer divergences when compared to other review studies, which should also yielded a higher degree of reliability (Petersen *et al.*, 2015). In this systematic mapping, besides reaching a final conclusion on the topic at hand, further data were obtained regarding the frequency of publications, geopolitical origin of the studies, media format of publication and the different techniques used. Figure 1 presents the main stages of this methodology which serve as a reference guide for the remainder of this work.

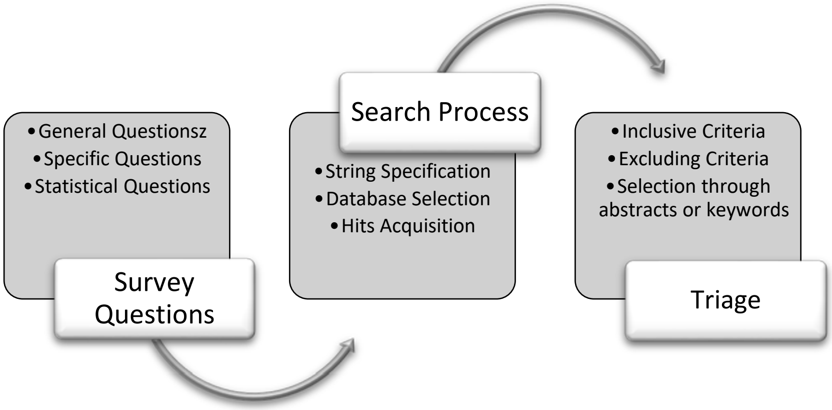

Figure 1. Stages of Systematic Mapping.

Survey Questions

Survey questions were defined as general questions (GQ), specific questions (SPQ) and statistical questions (STQ) as shown in Table 1. General questions were aimed at current recovery techniques and possible applications of recovered phosphorous. Specific questions were used to identify the scope of application of each technique with respect to the type of industrial wastewater, in this case liquid or sludge. Statistical questions were used to determine the geopolitical source of the studies and frequency of publication within the last 10 years.

Table 1. Survey Questions

Type	Detailed Question
	General Question
GQ1	What is the method of phosphorous recovery?
GQ2	What are the applications of the recovered phosphorous?
	Specific Question
SPQ1	What are the methodologies used for phosphorous recovery from liquid wastewater/sludge?
	Statistical Question
STQ1	Where were the results published?
STQ2	How many publications per year?

Search Process

The search process was split in 3 parts: search string specification, database selection with attention to each particular search algorithm and acquisition of results that match the criteria. For this stage, the methodology of (Petersen *et al.*, 2015) was applied. For the first part, search terms and their more relevant synonyms were identified. Primary search terms were defined as "phosphorous" and "recovery" while the secondary search term was "application." Thus, mapping would include any situation in which both "phosphorous" and "recovery" would be related to "application." In order to ensure that the hits would include the largest number of studies, synonyms of secondary terms were also included, as shown in Table 2.

Table 2. Search string terms and synonyms

Terms	Synonyms
phosphorous	*"phosphorus"* OR *"phosphor"* OR *"phosphate"*
Recovery	*"recovery" OR "recuperation"* OR *"retrieval"*
Application	*"application" OR " adhibition "* OR *"applied"*

The resulting search string based on the terms and synonyms of Table 2 was defined as:
(("phosphorus" OR "phosphor" OR "phosphate") AND ("recovery" OR "recuperation" OR "retrieval") AND ("application" OR "adhibition" OR "applied"))

Once the search string has been defined, 3 relevant databases were selected for the search: Science Direct, SciELO e EBSCOhost. The databases were selected based on their relevance in the areas of chemistry and agriculture in order to adhere to the desired emphasis on physicochemical or biological recovery of phosphorous and its application in food production. Each database has its own search algorithm so that incorporating the search string required different formulations. In the case of Science Direct, the "advanced search" resource was used and the search string was sought throughout the entire body of text of each publication. In the case of SciELO, the "advanced search" feature was also used with the search string applied to all indexes. In the case of EBSCOhost, the search string was inserted in a built-in advanced search template with a Boolean search option selected.

Triage

Triage was applied to keep the more relevant results, eliminate eventual dissonances in the search results and restrict the timeframe of publication. To this end, a series of including and excluding criteria were applied as auxiliary tools to filter out the results. The including criteria (IC) were:

- IC1: study is published in a journal;
- IC2: study must be a full paper;
- IC3: study must be related to the field of engineering or life science.

Studies that did not conform to all three including criteria were eliminated and the remaining studies subjected to a series of excluding criteria (EC):

- EC1: study was published before 2008;
- EC2: study was published as thesis/dissertations, magazines or as a congress paper;
- EC3: study is not related to industrial wastewater.

After excluding studies non-relevant to the scope of this work, an additional filter was applied with respect to the title and keywords. Further reading of the abstract and full text removed other non-relevant studies and for the last step, duplicate studies were excluded. The entire process is shown schematically in Figure 2. Figure 2 shows the filtering actions, remaining number of studies and % of excluded studies after each step. It can be seen that mapping started with over 132 thousand studies which were whittled down to 81.

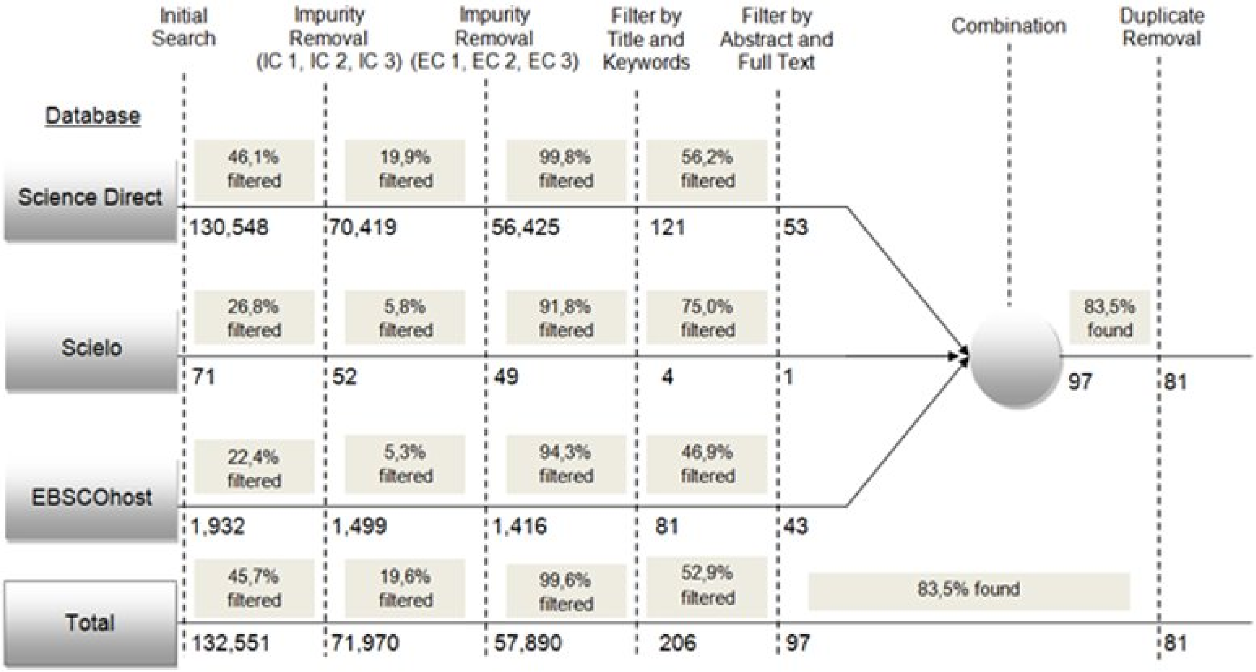

Figure 2. Filtering steps of the systematic mapping method.

The largest numbers of studies were obtained from Science Direct and EBSCOhost. After triage, a proportional comparison between final and initial results indicates that EBSCOhost provided the most qualitative results. In terms of raw numbers, Science Direct provided the largest number of studies post-filtering. However, these were the result of pre-filtering a number of studies 100 times larger than the ones obtained from EBSCOhost. Scielo, on the other hand, provided the least number of studies and a single one remained post-filtering. This result was understandable as Scielo was the database which least conformed to the search criteria.

Table 3 shows the 81 studies resulted from the filtering process identified with a Reference Number.

Table 3. Identification of the filtered articles

Reference Number	Reference Identification	Reference Number	Reference Identification
[1]	(Kataki *et al.,* 2016a)	[42]	(Liu *et al.,* 2017)
[2]	(Hirota *et al.,* 2010)	[43]	(He *et al.,* 2017)
[3]	(Sturm *et al.,* 2010)	[44]	(Cie and Konieczka, 2017)
[4]	(Tan and Lagerkvist, 2011)	[45]	(Roy, 2017)
[5]	(Fischer *et al.,* 2011)	[46]	(Furuya *et al.,* 2017)
[6]	(Lu *et al.,* 2012)	[47]	(Huang *et al.,* 2017a)
[7]	(Yuan *et al.,* 2012)	[48]	(Huang *et al.,* 2017b)
[8]	(Cusick and Logan, 2012)	[49]	(Marshall *et al.,* 2017)
[9]	(Kodera *et al.,* 2013)	[50]	(Lei *et al.,* 2017)
[10]	(Guedes *et al.,* 2014)	[51]	(Melia *et al.,* 2017)
[11]	(Tsuji and Fujii, 2014)	[52]	(Chang *et al.,* 2017)
[12]	(Niewersch *et al.,* 2014)	[53]	(Peng *et al.,* 2018)
[13]	(Atienza-Martínez *et al.,* 2014)	[54]	(Ma *et al.,* 2018)
[14]	(Gifford *et al.,* 2015)	[55]	(Kim *et al.,* 2018)
[15]	(Schütte *et al.,* 2015)	[56]	(Luo *et al.,* 2018)
[16]	(Shalaby *et al.,* 2015)	[57]	(Nir *et al.,* 2018)
[17]	(Huang *et al.,* 2015a)	[58]	(Shashvatt *et al.,* 2018)
[18]	(Fang *et al.,* 2015)	[59]	(Ottosen *et al.,* 2018)
[19]	(Huang *et al.,* 2015b)	[60]	(Adam *et al.,* 2018)
[20]	(Yan *et al.,* 2015)	[61]	(Xu *et al.,* 2018)
[21]	(Wang *et al.,* 2016)	[62]	(Almatouq and Babatunde, 2018)
[22]	(Tian *et al.,* 2016)	[63]	(Yu *et al.,* 2018)
[23]	(Havukainen *et al.,* 2016)	[64]	(Haddad *et al.,* 2018)
[24]	(Shepherd *et al.,* 2016)	[65]	(Li *et al.,* 2018a)
[25]	(Kataki *et al.,* 2016b)	[66]	(Sørensen *et al.,* 2018)

Reference Number	Reference Identification	Reference Number	Reference Identification
[26]	(Tarayre *et al.,* 2016)	[67]	(Yan *et al.,* 2018)
[27]	(Kim *et al.,* 2016)	[68]	(Fang *et al.,* 2018)
[28]	(Ye *et al.,* 2016)	[69]	(Gorazda *et al.,* 2018)
[29]	(Hermassi *et al.,* 2016)	[70]	(Yan *et al.,* 2018)
[30]	(Huang *et al.* 2016)	[71]	(Santos and Pires, 2018)
[31]	(Valverde-pérez *et al.,* 2016)	[72]	(Li *et al.,* 2018b)
[32]	(Lu *et al.,* 2016)	[73]	(Lei *et al.,* 2018)
[33]	(Guedes *et al.,* 2016)	[74]	(Wan *et al.,* 2018)
[34]	(Lee *et al.,* 2016)	[75]	(Costa *et al.,* 2019)
[35]	(Dai *et al.,* 2016)	[76]	(Li *et al.,* 2019a)
[36]	(Almatouq and Babatunde, 2017)	[77]	(Li *et al.,* 2019b)
[37]	(Remmen *et al.,* 2017)	[78]	(Salehi *et al.,* 2019)
[38]	(Tian *et al.,* 2017)	[79]	(Tian and Zhang, 2019)
[39]	(Wu *et al.,* 2017)	[80]	(Zhao *et al.,* 2019)
[40]	(Li *et al.,* 2017)	[81]	(Sempiterno, 2017)
[41]	(Córdova *et al.,* 2017)		

**Tile of articles related to similar Reference Identifications:*

(Kataki et al., 2016a) - Phosphorus recovery as struvite: recent concerns for use of seed, alternative Mg source, nitrogen conservation and fertilizer potential

(Kataki et al., 2016b) - Phosphorus recovery as struvite from farm , municipal and industrial waste : Feedstock suitability , methods and pre-treatments

(Huang et al., 2015a) - Recovery of phosphate and ammonia nitrogen from the anaerobic digestion supernatant of activated sludge by chemical precipitation

(Huang et al., 2015b) - Crystallization and precipitation of phosphate from swine wastewater by magnesium metal corrosion

(Huang et al., 2017a) - Phosphate recovery from swine wastewater using plant ash in chemical crystallization

(Huang et al., 2017b) - A pilot-scale investigation on the recovery of zinc and phosphate from phosphating wastewater by step precipitation and crystallization

(Li et al., 2018a) - Recovery of phosphate and dissolved organic matter from aqueous solution using a novel CaO-MgO hybrid carbon composite and its feasibility in phosphorus recycling

(Li et al., 2018b) - Ultrafast selective capture of phosphorus from sewage by 3D Fe_3O_4*@ZnO via weak magnetic field enhanced adsorption*

(Li et al., 2019a) - Phosphorous recovery through struvite crystallization: Challenges for future design

(Li et al., 2019b) - Effects of pH , soluble organic materials , and hydraulic loading rates on orthophosphate recovery from organic wastes using ion exchange.

Results

The 81 studies obtained through systematic mapping were found to answer the survey questions. However, not all studies were able to reply to all 5 survey questions simultaneously and fewer studies were able to reply to the general and specific questions.

GQ1: What is the method of phosphrous recovery?

Several methods of phosphorous recovery from urban and industrial wastewater have been developed. Some of these methods include chemical precipitation, adsorption, crystallization, ionic membrane exchange and electrochemical separation while others are exclusively biological (Peng *et al.*, 2018). These methods are not mutually exclusive and can be applied in combination with each other along the entire recovery process.

The studies were classified with respect to the recovered phosphorous compound in Table 4. Studies that appear in more than one row contained comparison between multiple distinct recovery methods. Studies with a more general outlook of recovery, be it psychochemical or biological with no specific detailing of the methods, are not listed in Table 4. Consequently the "% of total" values shown in Table 4 are with respect to the total number of studies contained in the table instead of the 81 results from the systematic mapping.

Table 4. Phosphorous recovery compound

Recovery Compound	Number of Studies	% of total	Reference Numbers
Struvite ($NH_4MgPO_4·6H_2O$)	24	39.3%	[1], [5], [7], [8], [17], [19], [21],[25], [30], [32], [34], [36], [44], [47], [51], [53], [56], [58], [61], [62], [66], [67], [76], [80]
Ca-P	15	24.6%	[6], [11], [18], [29], [35], [40], [49], [50], [51], [53], [54], [57], [58], [68], [73]
Fe-P / Al-P	5	8.2%	[11], [27], [51], [72], [73]
Poly-P	17	27.9%	[2], [7], [9], [14], [22], [26], [28], [31], [32], [38], [39], [51], [52], [60], [63], [70], [78]

In the studies presented, phosphorous was released through biochemical or thermochemical processes while recovery occurred usually through precipitation as struvite. Table 4 shows that struvite is the most common recovery compound obtained, followed by polyphosphates (Poly-P) and calcium phosphates (Ca-P). The more uncommon recovery compounds were ferrous phosphates (Fe-P) and aluminum phosphates (Al-P). Struvite crystals were particularly useful since they could be used as a slow-release fertilizer or raw material for chemical plants (Li *et al.*, 2019a). Despite its advantages, the reported elevated cost of large-scale struvite recovery operations prevented its widespread application as an alternative fertilizer (Kataki *et al.*, 2016a). However, this recovery technique could be coupled with other wastewater treatment processes to yield additional nutrients (Li *et al.*, 2019a).

The advantages of struvite recovery outweight its disadvantages, and in view of the possibility of using it in conjunction with other tecniques led to the development of several methods which aim to be more economical and ecologically viable. Some authors researched, in example, coupling alternate extraction of magnesium (Mg) or its oxidized metal (MgO) (Huang *et al.*, 2015b), low-cost bacterial mineralization of struvite (Luo *et al.*, 2018; Zhao *et al.*, 2019) and even optimization through hybrid microwave processes (Wang *et al.*, 2016).

Phosphorous could also be recovered through electrochemical precipitation of struvite crystals in cathode-chamber microbial fuel cells, with the added benefit of electricity generation (Almatouq and Babatunde, 2018) or hydrogen gas (Cusick and Logan, 2012). Another coupled electrochemical process allowed for the simultaneous extraction of ammoniacal nitrogen and recovery of struvite (Huang *et al.* 2016).

More recent work focused in crystallization through membranes, which allowed phosphorous recovery from liquid flows and allowed direct use as fertilizer (Sørensen *et al.*, 2018). Heating and cooling could also be used in a pre-treatment stage since struvite crystallization ran parallel to adsorption, composting and anaerobic digestion (Li *et al.*, 2019a). Vegetable ash could also be used as an alkaline reagent in struvite crystallization (Huang *et al.*, 2017a) which, coupled with air stripping and crystal precipitation, indicated a phosphorous recovery potential close to 100% (Huang *et al.*, 2015a).

An alternative to struvite precipitation was the crystallization of calcium phosphate compounds (Ca-P) such as hydroxyapatite. Selective precipitation of calcium phosphate was achieved through different release methods such as nano-filtration (Nir *et al.*, 2018). This technique had lower operational costs and lower chemical additives when compared to chemical precipitation. Further two-step extraction from residual sludge ash could increase calcium phosphate concentrations significantly and reduce in 50% metal contamination (Fang *et al.*, 2018). However, the use of calcium phosphates as fertilizer was limited to acidic soils (Zhao *et al.*, 2019). Despite the benefits of lower cost and use of chemical additives, Ca-P-crystallization is very restricted when related to fertilizer application and its use is directly linked to the desired end application. If the final product could be used in acid soils, this technique has more advantages than recovery in the form of struvite.

Ferrous and aluminum phosphate were more restricted in use and consequently appeared less frequently in the results (Melia *et al.*, 2017). Systems which used activated iron, more commonly in the form of zero-valence iron, were considered favorable for phosphorous recovery with elevated economic efficiency (Li *et al.*, 2018a). In addition, nanotubes of ferrous oxides already allowed for the separation of iron particles in conjunction to phosphorous recovery from wastewater (Kim *et al.*, 2016).

Biological removal of phosphorous included mechanisms that accumulate bacterial polyphosphates (Poly-P) through advanced enhanced biological phosphorous (EBPR) removal techniques. EBPR made use of a biofilm of polyphosphate accumulating organisms (PAOs) (Yu *et al.*, 2018) to concentrate phosphorous in the biomass (Yuan *et al.*, 2012). Sequestered phosphorous could be released from the biofilm through an alternating anaerobic/aerobic biofilter fed by additional sources of carbon in its aerobic phase (Tian *et al.*, 2016). Conventional EBPR methods could also be coupled with other nutrient recovery techniques such as SIPER, which was known to minimize residual sludge while recovering phosphorous (Yan *et al.*, 2015). A disadvantage of this process was the potential excess formation of granular sludge. However, systems that coupled nitrification/denitrification with phosphorous removal were developed which reduce granular sludge generation (Lu *et al.*, 2016).

Techniques involving membranes and osmotic membrane bioreactors (OMBR) were also widely studied (Yan *et al.*, 2018) and integrated with reverse osmosis and membrane distillation techniques (Guedes *et al.*,2014). Phosphorous could be simultaneously removed and recovered by selective membranes appropriate to specific sources: nano filtration for diluted effluents (Nir *et al.*, 2018), calcium phosphate adsorption for effluents from anaerobic membrane bioreactors (Furuya *et al.*, 2017) or zeolite adsorption activated by powdered calcium (Hermassi *et al.*, 2016).

GQ2: What are the applications of the recovered phosphorous?

Phosphorous could be used in the production of several chemicals of aggregate value (Hirota *et al.*, 2010) and as a fundamental component of fertilizers in current food production (Peng *et al.*, 2018). Since it is essential for modern agricultural production, demand for phosphate-based fertilizers can be projected to rise drastically alongside population growth. This worldwide context was responsible for 89.6% of the surveyed studies citing phosphate-based fertilizers as one of the applications of recovered phosphorous. Depending on the composition of the recovered phosphorous compound, it could be used in acidic (Fang *et al.*, 2015) or alkaline soils (He *et al.*, 2017).

Table 5 shows the different applications of recovered phosphorous, the number of studies, total percentage with respect to the studies in the table and the reference numbers.The granular form of struvite obtained from precipitation and crystallization was usually used as a slow-release fertilizer (Shalaby *et al.*, 2015). Crystal formation could be improved with both natural seeds and synthetic materials. Other crystallized compounds, such as calcium phosphate, were determined to be efficient only in acidic soils while struvite could be applied to either acidic or alkaline soils (Peng *et al.*, 2018), making struvite at the outset a more interesting alternative. An alternative process developed in Finland used a thermochemical treatment to extract phosphate from sludge ash from residual manure.

Table 5. Applications of recovered phosphorous

Application	Number of Studies	% of Total	Reference Numbers
Phosphate-based fertilizers	24	88.9%	[1], [2], [11], [16], [18], [20], [23], [24], [25], [32], [34], [42], [43], [47], [54], [65], [66], [67], [69], [71], [74], [75], [77], [81]
Agricultural soil correction	1	3.7%	[64]
Raw material for agregate value chemicals	2	7.4%	[2], [72]

The resulting phosphate presented a phosphorous pentoxide (P2O5) potential more than sufficient for the agricultural fertilizer demand of the entire country (Havukainen *et al.*, 2016). Many wastewaters contained both phosphorous and nitrogen which could also be recycled through algae cultures and reused (Santos and Pires, 2018), this process also has the benefit of reducing the overall chemical fertilizers use. Depending of their psychochemical characteristics, these compounds could be applied directly as fertilizer (Adam *et al.*, 2018; Sørensen *et al.*, 2018).

However, it was not always possible to use recovered phosphates as a raw material for fertilizers. Some specific waste, however, such as ash from residual sludge was more susceptible to containing heavy metal concentrations (Guedes *et al.*,2014), and had limited application in agriculture (Li *et al.*, 2017). Therefore, the viability dependes on the levels of non-volatile inorganic elements, heavy metal concentration and mobility potential.

SPQ1: What are the methodology used for phosphorous recovery from liquid wastewater/sludge?
The selected method for phosphorous recovery had to account for the characteristics of the source wastewater/sludge. For example, a recovery method suitable for liquid wastewater may not have been able to cope with a thick sludge. In the case of liquid wastewater, the recommended technique was crystallization (Peng *et al.*, 2018), which had the advantages of high recovery levels and low environmental risks. As presented in the replies to GQ1, crystallization could occur in the form of struvite or calcium phosphates. In the case of calcium-rich wastewater, calcium phosphate recovery was promoted with applications of modified or unmodified biocoal (Fang *et al.*, 2015; Marshall *et al.*, 2017) while adsorptives were used to recover phosphorous as hydroxyapatite (Tsuji and Fujii, 2014; Ma *et al.*, 2018).

However, this process had high costs. Nano filtration through membranes was being studied as a more economic method to achieve selective calcium phosphate precipitation (Luo *et al.*, 2018). Struvite recovery from aqueous solutions through precipitation (Shalaby *et al.*, 2015) and electrochemical decomposition were well documented, which had the added benefit of simultaneous phosphorous recovery and ammoniacal nitrogen removal (Huang *et al.* 2016). Bacterial mineralization could also be used to reduce costs of struvite recovery (Luo *et al.*, 2018) and chemically-induced crystallization could further increase yields in biological phosphorous removal (Lu *et al.*, 2012).

Biological removal from liquid wastewater was achieved through alternating anaerobic/aerobic biofilters (Tian *et al.*, 2016; Tian *et al.*, 2017) and polyphosphate-accumulating organisms (PAOs). This method could also be applied to sludge, with a PAO-enriched biofilm recovering nutrients without requiring the removal of excess sludge from wastewater (Yuan *et al.*, 2012; Kodera *et al.*, 2013). The alternating anaerobic/aerobic reactor allowed simultaneous phosphorous removal and denitrification (Lu *et al.*, 2016) and if the residue sludge was pre-treated with acid or alkali products, anaerobic fermentation further increased phosphorous release (Wu *et al.*, 2017).

Thermal conversion of sludge was conducted through combustion, co-combustion, pyrolysis or gasification. These techniques showed potential (Gorazda *et al.*, 2018) since sludge ash had elevated phosphorous recovery thermochemical potential (Havukainen *et al.*, 2016). In the case of biomass ash, electrodyalisis separation of phosphorous could be performed in parallel with heavy metal removal (Guedes *et al.*,2014), which increased the viability of the technique. Selective leaching of phosphorous could also be performed in the solid residue from gasification (Gorazda *et al.*, 2018). In order to obtain a phosphorous extract with a higher degree of purity, it was recommended that ashes be treated in 2 stages (Fang *et al.*, 2018).

STQ1: Where were the results published?

Table 6 groups the studies according to the continent in which they were published. Some studies were conducted in cooperation between researchers of different continents and consequently appear in more than one row. Table 6 shows that 45 publications originated from Asia while 33 were from Europe. The least number of publications were from Africa and South America, with 3 and 2, respectively.

Figure 3 presents the % of publications and number of studies analyzed distributed with respect to each continent. Publications originated from Asia and Europe accounted for about 75% of the selected studies. According to a 2018 U.S. Geological Survey (USGS) report, regarding mining and reserves of phosphate minerals, China was the largest world producer of phosphorous despite having reserves 15 times smaller than Morocco and West Sahara, which contained the largest world reserve. This high level of extraction could explain the elevated number of studies being conducted in Asia. Europe, on the other hand, lacked phosphorous reserves and at the time relied on imported phosphates (Schröder *et al.*, 2010). This also explained the large number of publications from Europe and their focus on new recovery methods.

Table 6. Location where studies were published

Continent	Number of Studies	Reference Numbers
Africa	3	[16], [64], [79]
North America	11	[8], [12], [14], [22], [27], [38], [45], [58], [65], [73], [77]
South America	2	[26], [75]
Asia	45	[1], [2], [6], [9], [11], [14], [17], [18], [19], [20], [21], [22], [25], [27], [28], [30], [32], [34], [35], [38], [39], [40], [41], [42], [43], [46], [47], [48], [52], [53], [54], [55], [56], [61], [65], [67], [68], [70], [72], [73], [74], [76], [77], [79], [80]
Europe	33	[1], [3], [4], [5], [10], [13], [15], [23], [24], [25], [26], [27], [29], [31], [32], [33], [36], [37], [42], [43], [44], [50], [51], [57], [59], [60], [62], [64], [66], [68], [69], [71], [81]
Oceania	8	[7], [28], [49], [61], [63], [70], [76], [78]

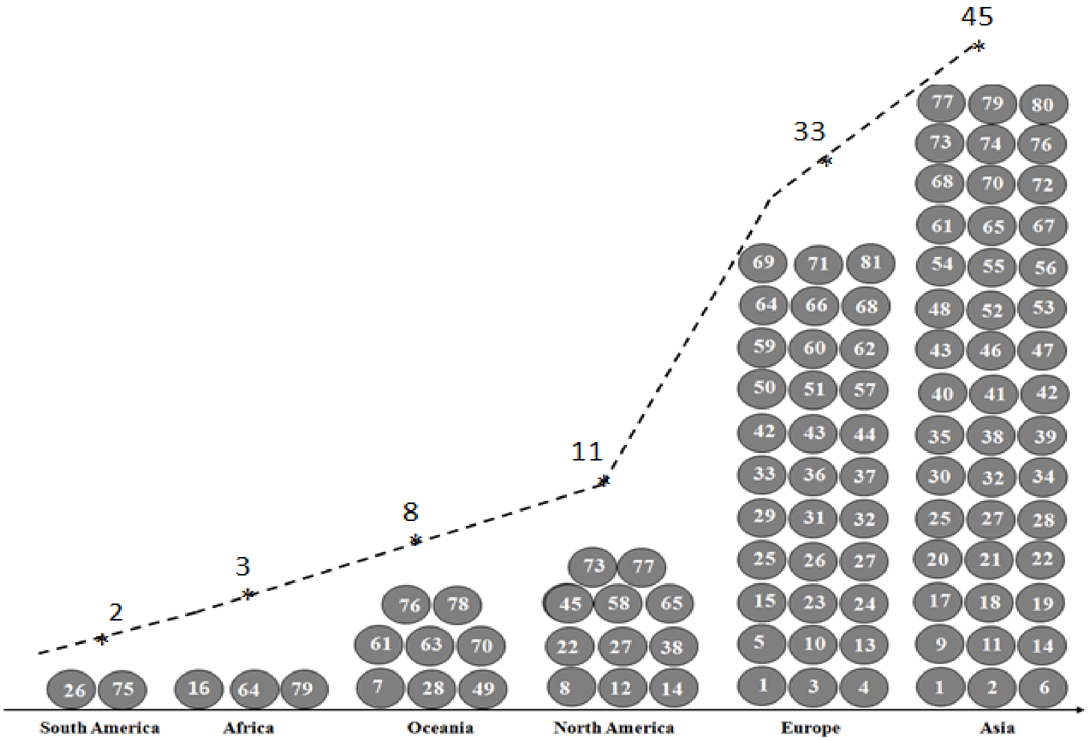

Figure 3. Publications and % distribution by continent.

STQ2: How many publications per year?

In order to detail the number of publications per year, Figure 4 shows the number of studies published according to year except for 2019. The studies were further broken down according to their database of origin. Figure 4 shows a rising trend in the number of publications per year. Between 2010 and 2018, the number of publications increased tenfold. This increasing in studies regarding phosphorous recovery in the past few years reflected the importance of this topic and the investment in the development of new techniques that were both more cost-effective and ecologically-sound.

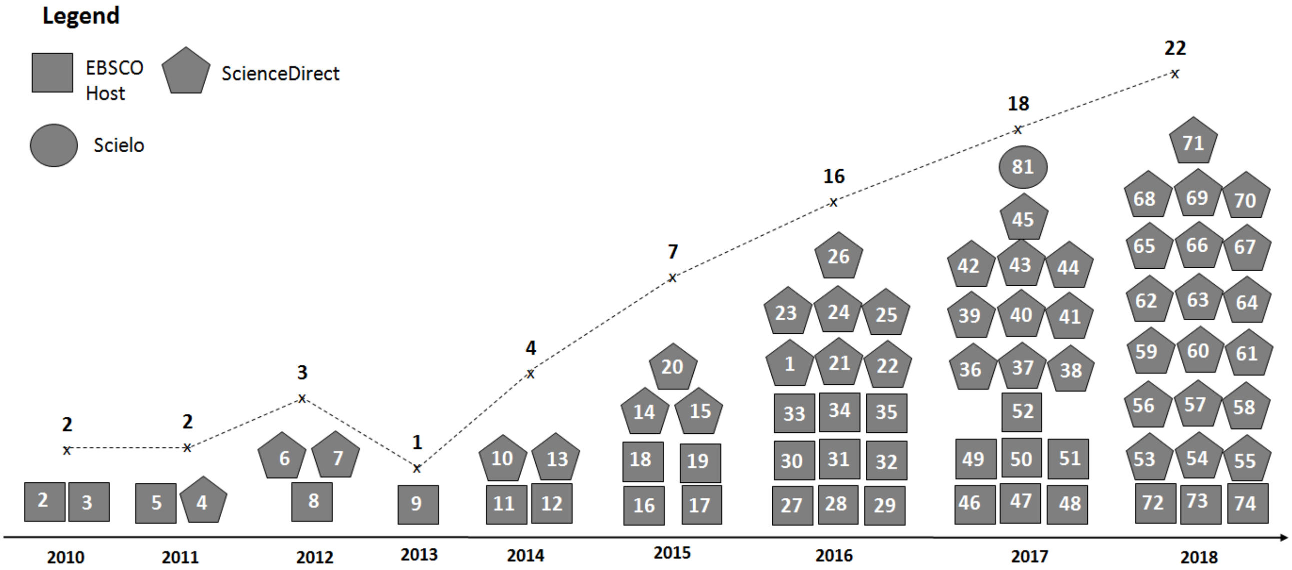

Figure 4. Yearly Publications Originating Databases from 2010 to 2018.

Validation of results

Systemic mapping, as a tool for literature review, is not immune to factors that may affect the reliability of results. These factors can arise in the mapping process due to decisions by the authors or in the filtering process which may exclude relevant studies as well as include others that are less relevant. To reduce the possibility of negative impacts, the filtering process was based in a previous technique (Petersen *et al.*, 2015), mapping followed accepted revision studies (Dias *et al.*, 2018; Bischoff *et al.*, 2019) and additional tools such as Mendeley reference manager were used. The three selected databases were considered to be the most comprehensive with the respect to the topic at hand and were able to supply the largest number of relevant results. The terms used in the search strings shown on Table 2 were carefully selected based on previous surveys and the synonyms were specially selected to increase the possibility of hits and reduce the accidental exclusion of relevant studies.

Discussion

Crystallization was the most studied phosphorous recovery method, wich can be explained due to its high yield and good cost-benefit relation. In the surveyed studies, crystallization largely recovers phosphorous as struvite and calcium phosphate,wich was also observed by Cieslik and Konieczka (2017), showing that this technique remains an important trend in the development of new methods for phosphorus recovery. This can be explained by the fact that these compounds have the widest range of applications at low production costs, with high-quality phosphoric minerals. Precipitation as ferrous or aluminum salts were efficient but limited in their application, since they tended to leave behind residues with limited biological destination and could produce other forms of contamination (Melia *et al.*, 2017).

Struvite recovery was the most studied technique. This was a direct result of its range of applications as a raw material for other aggregate value products, slow-release fertilizer and versatility of use in either acidic or alkali soils. This result is also in line with whats was observed by Cieslik and Konieczka (2017) in their review, where is stated another advantage of this technique: the low environmental riks, due to its poorly soluble feature.

Its importance was clear as phosphate fertilizer produced from recovered struvite, cited in 89.6% of the studies surveyed, showing that, although there has been research focused on other applications, the main application found for recovered phosphorus is in agriculture. However, issues remained and a viable large-scale method of struvite recovery had yet to be developed, since pipelines can clog due to spontaneous precipitacion. Roy (2017) brings up an important point to be analyzed: for this technique to be favorable, it is necessary that the amount of P recovered compensates for the intensive chemical and energetic operation involved.

Recovery in the form of calcium phosphate was an alternative to struvite, as identified in 23.8% of studies surveyed but its application as a fertilizer was limited to acidic soils (Schütte *et al.*, 2015). Biological removal and recovery methods were considered “cleaner” and were highlighted in recent studies, characterized as a trend for new recovery methods, but still without major industrial applications. Biological processes were more attractive to the agricultural sector, yielded less residual sludge and made use of less chemical products when compared to psychochemical processes (Tarayre *et al.*, 2016).

The largest number of publications originated from Europe and Asia, motivated by the lack of phosphorous reserves and improving production methods, respectively. Asian publications were

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driven by China being the leading world producer of phosphorous. There was also a growing trend in the number of publications in recent years, which attested to the relevance of phosphorous recovery as a research topic and the importance of systematic mapping studies such as the current work. Data from this study indicated that interest in alternative methods of phosphorous recovery should remain relevant and future increases in the number of studies could be expected.

In view of the barriers encountered by the methodologies developed so far, future studies on the subject should account for the demands of better cost-benefit relation, large scale viability and potential applications mostly connected to the field of agriculture.

Conclusions

This study presented a literature review of phosphorous recovery from industrial wastewater conducted through systematic mapping. The objective of gaining better understanding of the topic was achieved by condensing and characterizing a large volume of information. The study identified the different forms of recovery, their range of applicability and applications. Statistical distributions with respect to geopolitical origin and frequency of publication were also analyzed.

It could be concluded that phosphorous recovery occurred mostly as struvite crystals.Despite sharing the same objective, the studies were not repetitive since they analyzed particular details of each method. Biological recovery had the potential of lowering costs and could yield an effective method with less reliance in chemical products.

The main application of recovery phosphorous was in agriculture, as raw material for either fertilizer or additives. While the use in the production of chemicals with higher aggregate value was discussed, most studies focused solely in fertilizer production.

The systematic mapping conducted indicated that phosphorous recovery from industrial wastewater was a growing field. There was an increasing trend in research and new methodologies and processes should arise in the future, with a corresponding increase in the number of publications.

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