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Methods for phosphorus recovery from waste water: A review

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Abstract

The commercial source of phosphorus is phosphate rock whose reserves are estimated to be depleted within 150 years at the current rate of use. Wastewater is used as an alternative source of phosphorus, which was initially recovered mainly by chemical precipitation. Nowadays, other methods have been researched and developed. They include biological, sewage sludge, wetland plants and wastewater irrigation. Chemical precipitation produces round pellets that can be used directly as fertilizer but the method is associated with high operating costs. Biological method is much more favorable than precipitation due to lower sludge production and chemical usage. However the system is less stable and flexible compared to chemical precipitation. Sewage sludge has high phosphorus recovery efficiency of 90-95% but it has an adverse effect on human health and the environment due to contaminants such as heavy metals and pathogens. Wastewater irrigation and wetland plants methods are potentially the simplest and low-cost methods of phosphorus recovery. However, poor wastewater irrigation management can affect human health as well as crops. Wetland plants must be routinely harvested to prevent phosphorus that has been incorporated into plant tissue to be returned to the water by decomposition processes. The choice of a method is more likely dependent on factors such as its efficiency, strength of wastewater, legislation, population served and economics of the particular method.

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Introduction

Phosphorus (P) is an element fundamental to all living things. It is vital for food production since it is one of the three nutrients (nitrogen, potassium and phosphorus) used in commercial fertilizers (Renee, 2013). The commercial source of P is Phosphate Rock (PR), the collective name given to natural calcium phosphates of various forms (Driver *et al.,* 1999). Deposits of PR occur throughout the earth's crust. However, high grade rocks are found in Morocco, United States, South Africa, Russia and China. Throughout the years, PR deposits have been heavily exploited and are in risk of being rapidly depleted (Driver *et al.*, 1999). As a result, P is becoming scarce as a mined resource, making the search for alternative sources a matter of urgency (Anirudhan *et al.*, 2006; Zhang *et al.*, 2012).

Wastewater is an important alternative source of P (Oluwafeyikemi, 2013). It contains high amounts of P derived from domestic and industrial sources that can be recovered to provide a reliable source of the nutrient. According to Cornel and Schaum (2009), phosphorus recovery is defined as a process that can (i) convert P into either plant available form for reuse as fertilizer or a raw material for P industry, and (ii) separate valuable P from harmful substances. In this study, the term 'phosphorus recovery' is used according to the above definition.

Development of technologies for P recovery began in 1950s in response to the issue of water pollution in the form of eutrophication and the need to minimize levels of P entering surface waters. It has however become a matter of interest in recent years due to the following reasons. Firstly, the imminent depletion of finite PR reserves in earth's crust. Researchers have predicted that PR reserves cannot last for more than 150 years (Loganathan *et al.*, 2014; Tyagi and Lo, 2013). As a result, more attention is being given to the recovery of P from wastewater. Another reason is the increasingly restriction of the direct use of sludge in agricultural soils in developed countries (Nguyen, 2015). This has in turn contributed to the desire to develop proper technology for recovering P from sludge. Moreover, P recovery is believed to create revenue by converting waste into commercial products (Tyagi and Lo, 2013). Last but not least, the need for compliance with regulatory obligations (Oluwafeyikemi, 2013). In general, these are some of the major factors that have played a role in the considerable advancement of the technologies for recovery of P from wastewater.

Currently, there are a number of established technologies employed to recover P from wastewater. This paper is therefore a literature review of the methods and techniques employed in P recovery technologies, emphasizing on their efficiency as well as strength and weaknesses.

Phosphorus recovery methods

Phosphorus recovery cycle first begins with accumulation or removal of P from wastewater and ends with the use of the captured nutrient directly as fertilizer/soil amendment or as raw material for P industry. Initially, P from wastewater was recovered by using chemical precipitation, which according to Morse *et al.* (1998) is the leading method. Nowadays different methods and technologies have been researched upon and developed, including biological and sewage sludge. Other simple and cost effective methods such as wastewater irrigation and wetland plants have also become well established, as further discussed in the following sections.

Chemical method via Precipitation

Precipitation in wastewater can occur naturally or by addition of chemicals. Chemical precipitation involves addition of metals ions that are able to form precipitates with inorganic phosphates dissolved in wastewater, forming an insoluble metal phosphate that is settled out by sedimentation. The most suitable and hence common metal ions used are Calcium (Ca^{2+}), aluminium (Al^{3+}) and ferric iron (Fe^{3+}), added as chlorides or sulphates (Morse *et al.*, 1998). Other coagulants such as natural and synthetic organic polymers and prehydrolyzed metal salts (e.g. polyaluminum chloride and polyiron chloride) are also used.

However, they generally have a relatively higher cost (Mehta *et al.,* 2015). The chemicals can be dosed prior to primary settling, during secondary treatment or as part of a tertiary treatment process. According to Morse *et al.* (1998), adding chemicals in tertiary treatment produces a high-quality effluent but the approach is not generally favoured because of high chemical costs and the creation of an additional chemical tertiary sludge.

The most promising compound for recovery from wastewater plants is magnesium ammonium phosphate hexahydrate ($MgNH_4$ PO_4. $6H_2O$) commonly known as struvite, which precipitates spontaneously in some wastewater processes where high concentrations of soluble phosphorus and ammonium are present (de-Bashan and Bashan, 2004; Williams, 1999)). Additional essential conditions are low concentration of suspended solids and a pH above 7.5. Precipitation of struvite requires that its components are available simultaneously in wastewater in the molecular ratio 1(Mg^{2+}): 1(NH_4^+): 1(PO_4^{3-}). Most wastewaters normally tend to be rich in ammonium but deficient in magnesium (Chimenos *et al.*, 2003; Munch and Barr, 2001). Therefore, supplementation of magnesium is required that also helps to increase solution pH. Sodium hydroxide (NaOH) can also be used to elevate pH (Stratful *et al.,* 2001). Conditions for struvite precipitation can be duplicated and exploited in a practical engineering process to economically extract it from wastewater in commercial quantities. This can be achieved by precipitating struvite in a dedicated reactor instead of allowing spontaneous formation (Munch and Barr, 2001; Stratful *et al.*, 2001). Precipitation typically produces P bound as a metal salt within the wasted sludge that has potential value to be used in agriculture as fertilizer.

Precipitation is a very attractive method for P recycling. It can accumulate 80-90% of soluble P in wastewater (Le Corre *et al.*, 2009). Struvite (magnesium ammonium phosphate) and hydroxyapatite (calcium phosphate) have been the most popular products, normally recovered as round pellets with low water quantity and high purity (Giesen, 1999). Struvite is an effective slow-release fertilizer, providing both nitrogen and phosphorus. Calcium phosphate is more or less the same as mined PR. It is therefore possible to use it as a direct substitute of PR in industrial production of P-fertilizer. Calcium phosphate can be mixed together with other nutrients or applied directly on agricultural fields as a slow-release fertilizer (Tchobanoglous *et al.*, 2014). The chemical coagulants can also remove organic matter, pathogens, viruses and other inorganic species such as arsenic and fluoride. As reported by Mehta *et al.* (2015), ease of operation and flexibility to changing conditions are the additional benefits of recovering P by precipitation. However, chemical methods are associated with high operating costs, increased salinity in the effluent (mainly as chloride or sulfate) and increased sludge production (up to 35%) (De-Haas *et al.,* 2000). Additional sludge that is produced can be challenging, especially if the method selected is lime application during primary treatment (Tchobanoglous *et al.*, 2003).

Use of alum after secondary treatment is predicted to produce much less sludge but the increase could still be problematic. Calcium increases chemical and sludge handling costs significantly more than aluminium and iron (Tchobanoglous *et al.*, 2014). Other weaknesses of this method are the addition of heavy metals present in raw coagulant and inhibitory effects on biological processes such as anaerobic digestion following the coagulation process. Sludge produced from chemical accumulation techniques, particularly with aluminum and iron coagulation, is agronomically less useful due to low bio-availability of the strongly bound P. Consequently, if this technique is to be applied as part of an overall nutrient recovery strategy, a subsequent release step is essential to improve bio-availability of the bound nutrients.

Biological method

Biological technologies makes use of the action of microorganisms to accumulate P from wastewater into their biomass allowing for this P to be removed and directly applied (as sludge) to agricultural land or solubilized and subsequently recovered as a mineral product e.g. struvite (Oluwafeyikemi, 2013). These species are known as Phosphate Accumulating Organisms (PAOs) (e.g. *Acinetobacter* sp.). During aerobic conditions, PAOs store phosphate as Polyphosphate (PolyP); using up Poly-β-hydroxybutyrate (PHB) as organic carbon. This makes PAOs less dependent on organic nutrients in wastewater thereby providing a competitive advantage.

The process of improving storage capacity of P as polyphosphate by PAOs biomass in activated sludge is known as Enhanced Biological Phosphorus Removal (EBPR). EBPR uses the fact that some organisms are able to take up more P than they require for cellular growth often called "luxury uptake". There exist various types of PAOs that are able to take up P in excess amounts as energy-rich polyphosphates whose P content can reach up to 20-30% of dry weight, while for common heterotrophic bacteria P content can be about 2% (Tchobanoglous *et al.*, 2014). So the basis of EBPR is to maximize the fraction of PAOs in the wastewater to ensure optimal P accumulation and recovery. Biological methods can recover P as (i) biomass, where P is concentrated in sludge and can be recycled for uses in agriculture or recovered after incineration, and (ii) mineral form, such as recovery of struvite as commercial fertilizer or for other industrial applications.

As far as recovery of P is concerned, biological method via EBPR is much more favorable than precipitation. Under favorable conditions the method can accumulate around 90% of P in sludge. Phosphorus is accumulated in the activated sludge and is relatively easy to recover compared to chemical method. Due to lack of chemical usage, biological methods have lower sludge production, lower chemical costs and sludge contains fewer metals (Morse *et al.*, 1998). Biological methods have higher value for agriculture as P is not bound to metals and therefore more available to crops. Nevertheless, biological methods have various shortcomings.

The EBPR process is very dependent on the wastewater characteristics and is less stable and flexible compared to chemical precipitation (Tchobanoglous *et al.*, 2014). It normally involves more complex configuration and operating regimes (Biswas, 2008) and high energy consumption (Ning *et al.*, 2008; Peleka and Deliyanni, 2009). Biological recovery methods require the addition of readily bio-degradable organic carbon, which make these processes costly (Nieminen, 2010). The functional micro-organisms (PAOs) are sensitive to variation of temperature and feed concentrations (Onyango *et al.*, 2007). Factors such as fluctuations of temperature, high rainfall and nutrient limitations can cause EBPR processes to experience upsets, deterioration of performance and even failure, thus requiring installation of backup systems (Oehmen *et al.*, 2007). Furthermore, these processes cannot accumulate trace levels of phosphorus (Sengupta and Pandit, 2011).

Wetland plants

Wetlands, either constructed or natural, offer a cheaper and low-cost alternative technology for recovery of P. One characteristic of wetland ecosystems is dominance of typical plant vegetation adapted to flooded or saturated conditions (Martin, 2006). These plants are known as wetland plants or macrophytes. They play a major role in P uptake from wastewater primarily through their root systems while some uptake occurs through immersed stems and leaves. There are different types of macrophytes i.e. free-floating, submerged, emergent and floating-leaved. Free-floating plants grow on the surface of wastewater and have the highest capacity for P accumulation since their roots are kept suspended in wastewater column rather than being rooted in sediments. As wetland plants are very productive, considerable amount of P is bound in the above ground biomass which can then be harvested to recover the nutrient. Once harvested, P can be recovered by using plant biomass as animal feed or fertilizer.

Wetland plants method offers several benefits. As pointed out earlier, P-rich biomass derived from wetland plants can be used as animal feed or fertilizer.

It can also be used as raw materials for nutrient release processing or as feedstock for bio-fuels production. However, one of the biggest concerns about this method is harvesting of the plants. Wetland plants must be routinely harvested to prevent P that has been incorporated into plant tissues to be returned to the water by decomposition processes. For maximum P recovery, harvesting is required at a time when high amount of P is accumulated in the above ground biomass of the plants. Currently, only limited information about peak P uptake of different wetland plant species is available. Therefore, there is a need for further research in that particular field. Furthermore, the area required by plants to recover nutrients is dependent on nutrient content and areal biomass productivity. According to Mehta *et al.* (2015), more studies are required in the field of plant biotechnology to improve nutrient uptake while minimizing biomass yields and footprint, so that it is more comparable with other biologically based nutrient uptake systems.

Sewage sludge methods

Historically, sludge has been used directly as fertilizer. The application of sewage sludge to agricultural land has been for a long time considered as the best P recovery option because of its high fertilizer value (Cornel and Schaum, 2009). Apart from direct use as fertilizer, P can be recovered after sewage sludge has been dried, ashed or otherwise subjected to chemical methods. Incineration and drying reduces volume of sludge requiring treatment and increases P concentrations in ash.

The latter is then applied directly on agricultural fields as fertilizer or soil amendment. However, the most common recovery methods involve leaching sludge or ash with dilute acid (hydrochloric acid/sulfuric acid) or alkaline solutions. Extraction of P is followed by a precipitation reaction using Ca and Mg (which are the most preferred salts) and Fe (Rittmann *et al.* 2011). A good example of this method is the BioCon process that incinerates sludge or dries under heat, leaches the residue with sulfuric acid before separating a range of by-products (Fe, heavy metals) using ion exchange resins (Levlin *et al.*, 2002).

Sewage sludge incorporates 90-95% of P in wastewater, resulting into most recovery processes having a potential to recover 80-95% of total P (Balmér, 2004; Cornel and Schaum, 2009). Recovering P by applying sludge to soil as P fertilizer has been done for many years and is likely to continue into the distant future. Studies have found that sewage sludge has equal or better performance as agricultural amendments when compared with commercial fertilizers and provides numerous additional soil conditioning benefits that inorganic fertilizers do not (Park, 2011; Pritchard *et al.*, 2010). It is also a simple and cost effective method of recovering P. Although there are benefits to soil fertility from regular application of sludge, there are also concerns regarding the potential odour and possible sludge contaminants. Sewage sludge carries significantly higher loads of heavy metals than other organic and mineral fertilizers (Nieminen, 2010). It also carries endocrine disruptors, Persistent Bioaccumulative Toxic (PBT) chemicals and pharmaceuticals which have an adverse effect on human health (Nieminen, 2010). An important drawback of directly using sludge as fertilizer is that the bio-availability and solubility of P is not well defined. Phosphorus release from organic species to "available" species will take an often unknown time frame. The presence of P in organic and unknown organic-P species may result in significant losses from agricultural land as runoff or leaching (Phillips, 2002). In addition, the use of sludge as fertilizer is seasonal, creating a need to store the material during the time they are not in use.

In order to recover P from ash, sewage sludge must be mono-incinerated without bark, solid waste or any other added material since co-incinerated sludge (together with other wastes) often lower the P concentrations and increase the contaminant levels. Incineration has an advantage of destroying organic pollutants such as Polychlorinated biphenyl (PCB), Dichloro-diphenyl-trichloroethane (DDT), dioxins and Persistent Organic Pollutant (POP) (Nieminen, 2010). Also, sewage sludge ashes have a small volume and are easy to transport making them an attractive source of P. In incineration process, P concentrates and absorbs in ash, simplifying P recovery processes. Sewage sludge ashes can potentially be used directly as fertilizer if the heavy metal content satisfies fertilizer legislation. However, the bio-availability of P in ashes is normally low compared to commercial fertilizer. Thermo-chemical technologies have been developed to primarily reduce the heavy metal content and increase bio-availability of P in sewage sludge ashes. Studies have shown that thermo-chemical processes are very effective in heavy metal removal from ashes produced by municipal solid waste incinerators (Jakob *et al.*, 1995; Lutz, 2002).

Wastewater irrigation

Wastewater is used to irrigate in many forms. It can be used as treated (reclaimed water) or untreated (raw wastewater). It can be applied directly to crops or indirectly after discharge and dilution with water from rivers or reservoirs (Blanca, 2006). Wastewater contains P and other macronutrients such as nitrogen and potassium and micronutrients such as magnesium and calcium that are needed for crop growth. Therefore, the major objectives of wastewater irrigation are to provide a reliable source of water and valuable nutrients such as P to crops. As pointed out by Finley (2008), wastewater use can supplement or even replace commercial fertilizer inputs.

In developed countries where environmental standards are adhered to, much of the wastewater is treated prior to use for irrigation of crops, while in developing countries both treated and untreated wastewater are used (Nyomora, 2015). According to WHO (2006), careful planning and management are needed for the positive aspects of wastewater irrigation to be achieved. This is because growth and development of crops depend on the quality of wastewater used.

Wastewater irrigation is potentially the simplest and low-cost option for P recovery from wastewater. Apart from P, wastewater irrigation can also recover other valuable nutrients (calcium, magnesium, potassium, nitrogen and organic matter) important to crops and soil health. Wastewater irrigation also benefits the environment because it allows these valuable nutrients to be diverted from waste stream and recycled instead of being released into watercourses where they could become significant pollutants (Finley, 2008). Wastewater irrigation is also emerging as a form of climate change adaptation because it provides a consistent source of water in arid/semiarid areas (Trinh *et al.*, 2013).

Wastewater that is treated properly prior to irrigation has least impacts on human health and environment compared to untreated wastewater (Mahoo *et al.*, 2018). Untreated/poorly treated wastewater often contains a large range of contaminants from municipal, agricultural and industrial sources. The contaminants include for example excreta-related pathogens, heavy metals, skin irritants and toxic chemicals. These contaminants pose health risks to farmers and agricultural workers, their families, communities living in proximity to wastewater irrigation as well as consumers of the irrigated crops (Dickin *et al.*, 2016; Qadir *et al.*, 2007). Wastewater exposure has been linked to viral, bacterial and protozoan diseases such as salmonellosis, shigellosis, cholera, giardiasis, amoebiasis, hepatitis A, viral enteritis and other diarrhoeal diseases. Due to these widespread health risks, The World Health Organization (WHO) developed wastewater irrigation guidelines to ensure that contaminant levels in wastewater are below limits that are harmful to human health (WHO, 2006). This, among others, aimed at ensuring that P recovery potential of wastewater irrigation is exploited while minimizing associated public health risks. Nevertheless, in most developing countries wastewater irrigation is still generally practiced with raw or poorly treated wastewater, not only for its fertilizing properties but also because it is the only way to earn a living (Blanca, 2006).

Conclusion

A variety of methods for phosphorus recovery from wastewater have been developed. Some are simple and cost effective such as wastewater irrigation and wetland plants while others are complex and expensive e.g. chemical methods.

All methods reviewed have advantages and disadvantages as well as a variation in P recovery efficiency.

The choice of a method is more likely dependent on, among others, factors such as its efficiency, strength of wastewater, legislation, population served and economics of the method. As a result, the most appropriate way of recovering P may differ from region to region since national context and drivers are different. In poor countries, methods such as poorly treated/raw wastewater irrigation and direct application of sewage sludge on agricultural fields are expected to continue regardless of the adverse effects on environment and human health.

On the other hand, in developed countries more complex and costly methods with high efficiency may be employed. It can be concluded that, sustainable use of P cannot be achieved by P recovery from wastewater alone. There is a need for taking a holistic approach on the phosphorus cycle as a whole, improving use efficiency in agriculture, mining and industries.

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