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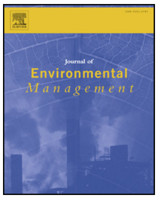

Review

Formation of struvite from agricultural wastewaters and its reuse on farmlands: Status and hindrances to closing the nutrient loop

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ABSTRACT

To meet the needs of a fast growing global population, agriculture and livestock production have been intensified, resulting in environmental pollution, climate change, and soil health declining. Closing the nutrient circular loop is one of the most important sustainability factors that affect these issues. Apart from being a serious environmental issue, the discharge of N and P via agricultural wastewater is also a major factor that disturbs nutrient cycling in agriculture. In this study, the performance, in terms of recovery, of N and P (individually, as well as simultaneously) from agricultural wastewaters via struvite has been comparatively summarized. Details on the hindrances to nutrient recovery through struvite formation from agricultural effluents, along with strategies to overcome these hindrances, are provided. In addition, various strategies for recovery performance intensification and operational cost reduction are comprehensively discussed. This work will provide scientists and engineers with a better idea on how to solve the bottlenecks of this technique and integrate it successfully into their treatment systems, which will ultimately help close the nutrient loop in agriculture.

1. Introduction

The manifestations of rapid population growth, urbanization, improved standards of living, and concurrent intensification of socioeconomic activities on overall environmental health are well recognized and acknowledged (Cordell et al., 2009; Clarke, 2013). Global cereal production has doubled in the past 40 years, mainly from the increased yields resulting from greater inputs of fertilizer, water, pesticides, and so on. This has increased the global per capita food supply and alleviate hunger in poverty-stricken areas (Alexandratos and Bruinsma, 2012). During this process, however, the increase in nitrogenous fertilizer application and exhaustion of the limited reserves of rock phosphate have been quite considerable. At present, the annual fertilizer consumption of rock phosphate is reported to be over one million tons, while the use of N fertilizer could be three times as much (Rahman et al., 2011). Moreover, it is estimated that within 100 years, mined P rocks will be completely exhausted (Cordell and White, 2014”).

Global cycling of these nutrients has been altered, owing to their widespread use in intensive agriculture, in ways that can contribute to severe environmental issues. Predominantly, nutrients can escape from farm fields to the surrounding soils, air, and waterways, when applied in excess of the plants' needs (Deng et al., 2006). Hence, the notion of a closed-loop nutrient cycle provides a simple, persuasive, and elegant approach for realizing efficient natural resource management-improved human well-being, and long-term food security (Maurer et al., 2006). The closing of the nutrient loop includes a wide range of ongoing efforts to make sure that nutrients are applied at times and places that align with the requirements of the plants. It also includes efforts to recover nutrients in usable forms from waste effluents and recycle them into cropping systems (Yorgey, 2014). The logic is that by recovering nutrients from waste effluents, a more “closed” system for sustainable agricultural development can be created.

For the recovery of nutrients (N and P), several techniques have been developed in the last five decades, including biological uptake, physical adsorption, and chemical precipitation (Tran et al., 2014; Güiza et al., 2015). Living organisms, such as microbes and plants, can be used to recover N and P as essential elements through uptake mechanisms. However, this process is highly dependent on the growth of these living organisms, which is often influenced by seasonal fluctuations (Cai et al., 2013; Pérez et al., 2015). For adsorption processes, N and P compounds in wastewater can be adsorbed onto the surface of

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adsorbents that are made of porous materials with a large surface area. This technique is often the last step in the wastewater treatment process and is suitable for waters with low pollutant contents. Compared to biological uptake and physical adsorption processes discussed above, chemical precipitation might be more effective, due to its high recovery efficiency. Therefore, it would be more economical to use this method for agricultural wastewaters with high contents of N and P.

Struvite formation is one promising option that can be used to sustain the nutrient loop in agriculture, as it simultaneously recovers N and P from waste effluents. Furthermore, the precipitated struvite ($MgNH_4PO_4{\cdot}6H_2O$) is in the form of stable orthorhombic crystals and can potentially be used as a slow-release fertilizer. Compared with traditional chemical fertilizers, struvite can equal crop production, but has fewer negative effects in runoffs into downstream water bodies (Liu et al., 2011; Dalecha et al., 2012). However, the use of struvite as a fertilizer still represents a challenge because of poor market development, high operating costs and lower crystal sizes. The potential of struvite for nutrient recovery from various wastewaters has been studied extensively, and some review papers have been published accordingly. However, those review papers did not specifically target agricultural wastewater (Kumar and Pal, 2015; Darwish et al., 2016; Kataki et al., 2016a, 2016b). Theoretically, this technology can be used to close the nutrient loop in agriculture, however, its efficiency will vary with type of agriculture wastewater because of the variability in the physical and chemical characteristics of different wastewaters. Therefore, the literature lacks a comprehensive review specifically targeting the status of, and hindrances to, nutrient recovery from agricultural effluents via struvite formation. Moreover, agriculture is a low-profit industry, hence technology developed to close the nutrient loop should be economical (Ravallion et al., 2007). Furthermore, information about the specific characteristics of agricultural wastewaters and ways to improve struvite formation efficiency and reduce costs is needed but not well reported.

In this review, the variability in the chemical composition of various agricultural wastewaters is compiled to assess their suitability for maximum nutrient recovery via struvite. Then, the performance in terms of recovery of N and P, individually as well as simultaneously, from agricultural wastewaters is comparatively summarized. Moreover, a detailed discussion on the hindrances to nutrient recovery from agricultural effluents through struvite formation is presented, as well as on the strategies to overcome those hindrances. The potential of struvite as a fertilizer for improving the growth and production of different crops is addressed. Most importantly, various strategies extracted from the latest publications on recovery performance intensification and operational cost reductions are comprehensively discussed. This will enable scientists and engineers to have a better idea on how to solve the bottlenecks of this technique and to integrate it successfully into their treatment systems, which ultimately will help in maintaining the nutrient loop in agriculture.

2. Sources and characterization of agricultural wastewater

Agricultural wastewater is generated from a variety of farm activities, including animal feeding operations and the processing of agricultural products. Sources of agricultural wastewaters include, but are not limited to, animal breeding discharge, agricultural food processing wastewater, leachate from the composting of biomass or manure, digested effluents, slaughterhouse wastewaters, horse washing waters, barnyard and feedlot runoff, and egg washing and processing effluent. Additionally, runoff from cropland results in sedimentation and fertilizer and pesticide discharges in water streams (Table 1). Discharge of agriculture-based wastewaters not only results in water pollution but also leads to the loss of essential nutrients (N and P). Therefore, recovery of these nutrients from such wastewaters could be a suitable option for closing the nutrient loop. However, some wastewaters, such as agricultural field runoff, effluents from the beverage and brewery industries, and horse washing waters, are not suitable for recovery because of their very low nutrient contents.

Livestock breeding effluent, from dairy, beef, swine, and poultry operations, as well as the anaerobic digestates of manure, are optimal for recovery, as they are rich in nutrients (Table 2). The chemical composition of agricultural wastes (manures) and effluents varies among daily operations and seasonally within the same operation (Bernet and Béline, 2009). Generally, multiple factors, including animal feed, animal age, local climate, bedding material, manure collection, storage, and handling are responsible for the variability in the chemical composition of agricultural wastes.

3. Struvite and its recovery performance from agricultural wastewaters

3.1. Struvite formation

Struvite is a crystalline compound, formed with equal molar concentrations of magnesium, ammonium, and phosphate, combined with six water molecules ($MgNH_4PO_4{\cdot}6H_2O$), as depicted in Equation (1).

$$Mg^{2+} + NH_4^{+} + PO_4^{3-} + 6\ H_2O \rightarrow MgNH_4PO_4{\cdot}6H_2O \quad (1)$$

Its molecular weight is 245.43 g mol^{-1}, and its solubility varies from sparingly soluble to readily soluble in alkaline and acidic conditions, respectively (Chirmuley, 1994). Its solubility value in water is 0.018 g cL^{-1} at 25 °C, while the solubility value increases from 0.033 g cL^{-1} to 0.178 g cL^{-1} at 25 °C as the concentration of HCl in solution increases from 0.001 M to 0.01 M (Le Corre et al., 2009). The crystallization process occurs across a wide range of alkaline conditions. Struvite might be described as a soft mineral due to its low specific gravity (1.7 g cm^{-3}) and orthorhombic structure (Lee et al., 2009). It can occur as an elongated structure, a tight aggregate of fine crystals, star-like particles, or coarse crystals in white, yellowish or brownish-white colors (Rahman et al., 2011; Kozik et al., 2011; Hutnik et al., 2013; Matynia et al., 2013). The size of struvite crystals can vary from 15 μm to 3.5 mm in length depending upon the production conditions (Adnan et al., 2003; Zhang et al., 2009). The chemical composition of struvite contains around 13%, 6% and 10% of P, N and Mg, respectively Ueno and Fujii (2001).

The development of struvite crystals takes place in two phases: crystal birth or nucleation, and crystal growth. Factors such as the initial crystal state of the compound, liquid-solid equilibrium thermodynamics, mass transfer between the liquid and solid phases, and reaction kinetics (Jones, 2002; Ohlinger et al., 1999) control the process of struvite formation. Nucleation begins with the formation of a crystal embryo from the combination of ions in solution (Mullin, 1992). Depending upon the supersaturation level, one of several mechanisms (homogenous primary nucleation, heterogeneous primary nucleation, surface secondary nucleation, etc.) could allow struvite to nucleate.

Homogenous primary nucleation requires the highest degree of supersaturation as nuclei apparition takes place in a supersaturated solution. Heterogeneous primary nucleation requires a lower degree of supersaturation, and nucleation takes place on a foreign surface, such as dust particles or parts of the crystallizer. Surface secondary nucleation (true nucleation) requires suspended particles of the same species as the solid being crystallized. The new surface nuclei are then detached either by particle shock or fluid shear forces (Regy et al., 2002).

After nucleation, struvite crystal growth begins, and the crystal embryos grow into noticeable crystals. Mass transfer and surface transfer or agitation methods control the growth rate of struvite crystals. The transport of solutes from a solution to the crystal surface by diffusion, convection, or a combination of the two are referred to as mass transfer, while the incorporation of materials into the crystal lattice through surface integration mechanisms is called surface transfer or agitation. Various physiological parameters like solution pH,