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Nutrient recovery from anaerobically digested chicken slurry via struvite: Performance optimization and interactions with heavy metals and pathogens

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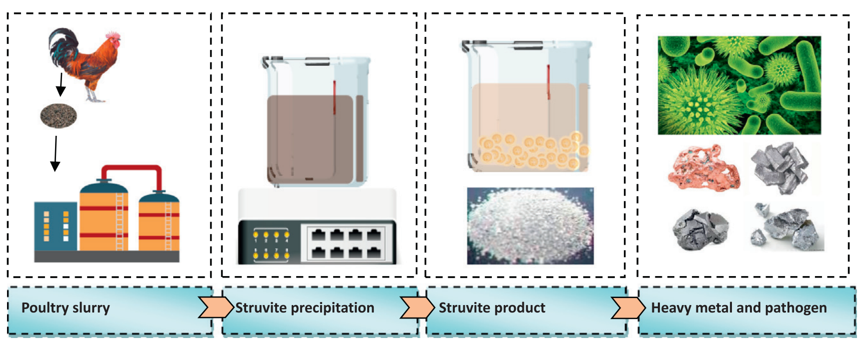
HIGHLIGHTS

- Nutrients recovery performance was optimized by using poultry slurry.
- The count of total coliform and *E. coli* reduced during struvite precipitation.
- Struvite precipitate was free of pathogen and with traces of heavy metals.
- Struvite precipitation recovered nutrients along with pathogens reduction.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to assess the potential of struvite precipitation to recover nutrients from anaerobically-processed poultry slurry and struvite's interactions with heavy metals (Zn, Cu, Pb, Cr, and Ni) and pathogens (total coliforms and *Escherichia coli*). The impacts of pH, Mg, N, and P molar proportion, reaction time, and mixing rate and duration were explored to determine the optimal conditions for nutrient recovery through struvite precipitation. A pH range of 9.5 to 10.5, was ideal for P and N removal and recovery, with a molar ratio of 1:1:1 for Mg:N:P. A mixing rate of 150 rpm for 10 min could allow nutrient recovery with little loss (3.32%) of NH_3 through volatilization, and also achieve an optimal struvite crystal size (50–60 μm). The results of X-ray diffractometry and scanning electron microscopy confirmed that the precipitates generated at pH 9 and 10 were orthorhombic struvite. Moreover, along with the recovery of nutrients, 40, 45, 66, 30, and 20% of Zn, Cu, Pb, Cr, and Ni, respectively, and 70% total coliforms and *E. coli* were removed by struvite precipitation from poultry slurry. This was observed despite that the levels of contaminants (heavy metals) detected in struvite were well below the permissible limits and free of pathogens. Consequently, it was inferred that the struvite quality was reasonable by virtue of its heavy metal and pathogen content, and therefore appropriate for application in the field. Similarly, struvite precipitation has multiple benefits as it can effectively recover nutrients as well as reducing pathogenic populations.

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1. Introduction

The global demand for meat has resulted in the rapid, intensive industrialization of animal breeding. The poultry industry has developed enormously in recent years, as it produces both meat and eggs. Poultry meat generation expanded by 5.3% every year from 2000 to 2011, while the beef (2%) and pork (3.4%) industries developed more slowly (FAO, 2014). Due to the rapid development of the industry, large amounts of chicken manure are generated which have caused environmental issues, such as air, water, and soil contamination, due to poor regulation and administration of manure deposition and disposal (Tilman et al., 2011).

Anaerobic digestion of chicken manure is beneficial as it generates biogas and a digestate. The digestate is rich in macro- (N, P, and K) and micronutrients (Zn, Cu, Fe, Mn, and Mo) (De La Fuente et al., 2013; Riseberg, 2015). However, digestate from biogas plants is continuously deposited onto nearby agrarian land, often exceeding the carrying capacity of the soil. This practice has caused the soil to become over-fertilized, thus discharging lethal runoff and causing nutrient leaching. Appropriate management of digestate is required to control nutrient content and to reduce runoff and leaching into aquatic systems (Tampio et al., 2016). Furthermore, in some plants where there is no nearby arable land, digestate is either discharged directly into water or deposited onto the same land consistently, which causes a variety of environmental and human health problems (Win et al., 2016).

Nutrients are valuable for agriculture, so recycling and recovery are becoming increasingly imperative to minimize nutrient losses and environmental concerns their release into aquatic systems (Wang et al., 2012; Kumar et al., 2013). Several technologies have recently been developed to recover nutrients, specifically N and P, from waste effluent (Mehta et al., 2015). These technologies include the osmotic membrane bioreactor (Qiu and Ting, 2014), electrodialysis/crystallization (Tran et al., 2014), amorphous calcium silicate hydrate adsorption (Ogata et al., 2016), biosorption (Kilpimaa et al., 2015), and struvite crystallization (Guiza et al., 2015; Huang et al., 2016a). Of these, struvite precipitation has attracted interest as it is a mature technology that generates a concentrated, saleable slow-release fertilizer (Rahman et al., 2014).

Nutrient recovery from poultry slurry via struvite has been investigated (Yilmazel and Demirer, 2013; Yetilmezsoy et al., 2013), but most previous studies focused on nutrient recovery and the potential usage of struvite as a slow-release fertilizer. In addition to nutrients, the digestate from poultry slurry contains heavy metals as they are not degraded during by anaerobic digestion (Zhang and Jahng, 2012). Similarly, a variety of pathogenic bacteria are also present in manure, and, although most can be killed during anaerobic digestion, some can remain in digestate after treatment (Viancelli et al., 2013; Fongaro et al., 2014). The fates of residual heavy metals and pathogens during nutrient recovery by struvite precipitation have not been well-studied. However, this would influence struvite quality, considering its contaminant content, and its potential use on land.

In this study, the ability of struvite precipitation to improve the efficiency of nutrient recovery from anaerobically-digested chicken slurry, was optimized considering several influential factors. During this process, struvite's interactions with heavy metals (Zn, Cu, Pb, Ni, Cd, and Cr) and pathogens (total coliforms and *Escherichia coli*) were also evaluated. The quality of struvite was evaluated based on its content of the aforementioned contaminants.

2. Materials and methods

2.1. Experimental materials

The anaerobically-digested poultry slurry used in the study was collected from the Deqing Yuan Biogas Plant, located in the suburbs of Beijing, China. The biogas plant operates under mesophilic conditions (37 °C) with a hydraulic retention time (HRT) of approximately 28 days. To eliminate influences of suspended solids, the anaerobically digested chicken slurry was centrifuged prior to experimentation. The characteristics of the filtered poultry slurry are presented in Table 1.

2.2. Batch experiments

A series of batch experiments were conducted for to separately evaluate the influence of different parameters on struvite, including pH, reaction time, molar ratio, and mixing rate and duration, for aqueous and poultry slurry. Standard Mg^{+2}, NH_4^+, and PO_4^{-3}solutions were prepared from analytical-grade $MgCl_2 \cdot 6H_2O$, NH_4Cl, and NaH_2PO_4 with purity ≥99%, respectively. $MgCl_2 \cdot 6H_2O$ was selected as the magnesium source as it is highly soluble. The solutions mixed at equimolar concentrations, and pH was then increased from 8.0 to 11.0, at 0.5 increments using 2 M NaOH and HCl. The solutions were stirred for 10 min and then allowed to react for 30 min. The mixtures were left for another 30 min to encourage the struvite to precipitate. Supernatant samples were collected and filtered through 0.45 μm for residual nutrients ($PO_4^{-3}$and NH_4^+) and heavy metal analysis. The samples taken for pathogen analysis were not filtered and used as such. The precipitates were filtered through a 0.45 μm membrane and dried at 40 °C for 48 h. The effects of other parameters, including reaction time (1 to 90 min), molar ratio (1:1:3, 1:1:2, 1:1:1 Mg:N:P, respectively), and mixing rate (150, 300, 450 rpm) and durations (5, 10, 20, 30 min) were determined at pH 9, which is favorable for struvite precipitation (Ryu et al., 2008; Gunay et al., 2008). All experiments were conducted at room temperature. Batch studies were also conducted using synthetic wastewater ($MgCl2 \cdot 6H2O$, NH4Cl, and NaH_2PO_4) as a control to allow the response of precipitation to various parameters to be compared with poultry slurry. To estimate the population sizes of total coliforms and *E. coli*, 10 mL of supernatant was collected after 0.5, 1, 3, 6, and 24 h of struvite precipitation in anaerobically-digested poultry slurry at pH 9 and 10. To measure heavy metal content (Zn, Cu, Pb, Cd, Cr, and Ni), poultry slurry samples were collected before and after struvite precipitation. Visual MINTEQ model version 3.1 was used to assess the distribution of various N and P species at different reaction pH levels so the optimum pH for maximum struvite precipitation could be determined. All experiments were conducted in triplicate.

2.3. Analytical methods

Methods of the American Public Health Association Standard (APHA, 2012) were followed to determine soluble chemical oxygen demand, NH_4^+, and PO_4^{-3} in the filtrate samples. The concentration of PO_4^{-3} in the supernatant was determined following the ascorbic acid method on a UV spectrophotometer, while that of NH_4^+ was analyzed following the phenate method. An inductivity coupled plasma mass spectrometer (ICP-MS-X2, Thermal Fisher., and USA) was used to analyze metals ions, including Zn, Cu, Ni, Cr, Pb, and Cd. The contents of heavy metal were

Table 1
Chemical composition of poultry wastewater.

Parameter	Symbol	Unit	Value
pH	–	–	8.23 ± 0.06
Soluble chemical oxygen demand	CODs	mg L 1	4500–6000
Total solids	TS	mg L 1	182 ± 13
Ammonical nitrogen	NH_4^+-N	mg L 1	4500–5000
Orthophosphate	PO_4^{3}-P	mg L 1	200–220
Potassium	K	mg L 1	2585
Calcium	Ca	mg L 1	35.5
Magnesium	Mg	mg L 1	13.7
Zinc	Zn	mg L 1	2.91
Copper	Cu	mg L 1	1.69
Lead	Pb	mg L 1	0.65
Cadmium	Cd	mg L 1	<0.01
Chromium	Cr	mg L 1	0.063
Nickel	Ni	mg L 1	0.55