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# Environmental assessment of alternative treatment schemes for energy and nutrient recovery from livestock manure

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## ABSTRACT

The application of livestock manure on agricultural land is being restricted due to its significant content of phosphorus (P) and nitrogen (N), leading to eutrophication. At the same time, the growing demand for N and P mineral fertilizers is increasing their production costs and causing the depletion of natural phosphate rock deposits. In the present work, seven technologically feasible treatment schemes for energy (biogas) and nutrient recovery (e.g., struvite precipitation) and/or removal (e.g., partial nitrification/anammox) were evaluated from an environmental perspective. In general, while approaches based solely on energy recovery and use of digestate as fertilizer are commonly limited by community regulations, strategies pursuing the generation of high-quality struvite are not environmentally sound alternatives. In contrast, schemes that include further solid/liquid separation of the digestate improved the environmental profile, and their combination with an additional N-removal stage would lead to the most environmental-friendly framework. However, the preferred scenario was identified to be highly dependent on the particular conditions of each site, integrating environmental, social and economic criteria.

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## 1. Introduction

Livestock manure contains significant contents of nutrients and organic matter that are often used to increase crop yield and improve the chemical and physical properties of soil (Schlegel et al., 2017). However, the current trends in livestock production intensification encounter continuous manure discharge into the environment (Oenema et al., 2007). Organic matter is responsible for acute water pollution incidents and odour problems. Phosphorus (P) and nitrogen (N) contaminate soil and surface waters, which may lead to eutrophication (Menzi et al., 2010). Consequently, European environmental regulations concerning the direct application of manure as fertilizer are increasing (Directive 91/676/EEC, 1991), limiting its use in regions where production is larger than the allowable demand for crop fertilization (Oenema et al., 2007). At the same time, the need of mineral N and P fertilizers and their production cost are steadily growing, jeopardizing the progressively exhausted deposits of phosphate rock (Vaccari, 2009). The combination of these two factors makes

the recovery of nutrients from animal manure crucial to protect the environment and to ensure a renewable source of fertilizers.

Anaerobic digestion (AD) or co-digestion (AcoD) is a widely implemented technology to remove organic matter from manure, which has the advantage of producing methane (CH<sub>4</sub>) as renewable energy (Nasir et al., 2012). Despite the fact that AD does not remove nutrients, it enhances their solubilisation (Tao et al., 2016); therefore, digestate post-treatment is required to either remove or recover nutrients. Several technologies have been tested to fulfil this goal; some of them are still on an experimental level, for example, the recovery of ammonia (NH<sub>3</sub>) from swine manure through gas-permeable membranes (García-González and Vanotti, 2015), while others are developed enough to be implemented at industrial scale. Most digestate post-treatment processes start with physical solid/liquid separation. Centrifugation is the most applied option leading to an organic P-rich solid stream and a liquid N-rich centrate (Burton, 2007; Møller et al., 2007). However, although the solid fraction is often used as fertilizer, its composition is variable, and it is impossible to separate phosphorus from organic matter. Membrane filtration is used to concentrate nutrients in lower volume streams. Additionally, it provides pathogen-free irrigation water and accomplishes limited N elimination but efficient P removal (Masse et al., 2007). When mainly P recovery is targeted, the precipitation of struvite (MgNH<sub>4</sub>PO<sub>4</sub>·6H<sub>2</sub>O) can be considered as slow-release fertilizer (Karakashev et al., 2008). However, an

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efficient solid/liquid separation step should always precede the struvite crystallizer to avoid interference of suspended solids with struvite precipitation. Conversely, if N recovery is pursued, ammonium sulphate production from ammonia stripping and subsequent scrubbing in sulphuric acid is the most applied technology (Bonmati and Flotats, 2003). But, when N recovery is not the final aim or when it is not complete, this nutrient is largely removed as  $N_2$  through the conventional nitrification/denitrification process. Innovative processes, such as nitrification/denitrification (Nit/Denit) and partial nitrification/anammox (PNA) can be performed if the biodegradable organic matter content is low, allowing energy savings and minimizing sludge production (Daigger, 2014). The above-mentioned technologies can be combined (Karakashev et al., 2008) to address regional requirements (mainly regarding N-, P- vulnerabilities, or both), but the environmental implications related to each treatment configuration should also be assessed to guarantee a sustainable scheme. To do so, Life Cycle Assessment (LCA) methodology is considered as a useful and widely applied tool to evaluate the environmental performance of a product or service by identifying critical stages in a supply chain where impacts could be reduced. In this sense, several LCA studies related to the AD of animal manure and the use of digestate for fertilization purposes have been published (Bacchetti et al., 2016). However, only a few go beyond those conventional strategies, focusing attention on the potential environmental advantages of nutrient recovery from digestate (Pardo et al., 2017; Rehl and Müller, 2011). In this context, the primary goal of this work was to study the environmental performance of seven technologically feasible treatment schemes aiming at energy and nutrient recovery from a mixture of livestock (pig and cow) manures.

## 2. Materials and methods

### 2.1. Technologies and treatment schemes

Seven technologies (anaerobic digestion, acidification, centrifugation, membrane ultrafiltration, struvite precipitation, nitritation-denitrification and High Rate Activated Sludge (HRAS) + PNA) were considered and tested in the operation of the ManureEcoMine pilot plant in Spain, with the exception of the biological N-removal processes. The precise description of the pilot plant can be found in Pintucci et al. (2017) and consisted of a 3 m<sup>3</sup> anaerobic digester, an ammonia stripping column, a decanter centrifuge (MD-60, Lemitec GmbH), an ultrafiltration system (UF F0701 Pilot skid, Likuid Nanotek) made of ceramic membranes with 100 nm pore size and a total filtrating surface of 1.08 m<sup>2</sup> and a 100 L struvite reactor. The seven technologies chosen were combined into seven treatment schemes (scenarios, Fig. 1).

The input mixture was identical for all scenarios, and it was composed of 52% cow manure, 43% pig manure and 5% segregates (w/w). Both manures were provided by a farm located in Folgueroles (Catalonia, Spain), while segregates is a sugar-rich product derived from the cleaning process in the food industry. The average physicochemical characteristics of the feeding mixture are shown in Table 1. A total of 100,000-ton feeding mixture per year, a representative value for medium-sized agro-industrial digesters (Bacchetti et al., 2016), were considered as the basis for comparison, resulting in a mass-based functional unit (FU) of 274 ton d<sup>-1</sup>.

Anaerobic stabilization was considered in all scenarios to reduce the environmental hazard of direct manure application, while at the same time allowing biogas production for energy needs (Nasir et al., 2012). The base scenario only included AD. In scenario 1, a centrifuge was added after AD to recover chemical oxygen demand (COD), total phosphorus (TP) and part of total Kjeldahl Nitrogen (TKN) in the solid fraction, while centrate was

treated through nitrification via nitrite pathway (nitritation) followed by denitrification (Regmi et al., 2014) to remove N. The novelty of scenario 2 was the production of struvite from the centrate prior to N removal. Struvite crystallization was supposed to be feasible even in the presence of suspended solids in the centrate (Capdevielle et al., 2015; Tao et al., 2016), as in the pilot plant trials struvite was detected in the precipitate when the abovementioned influent was used. In scenario 3, an ultrafiltration unit was implemented after the centrifuge to improve struvite precipitation, and, at the same time, to facilitate the testing of biological N-removal not only through nitritation/denitrification (option a), but also through partial nitritation/anammox (option b) (Lackner et al., 2014). The high concentration of biodegradable organic matter in the permeate required a HRAS process before the anammox step to diminish the incoming Biochemical Oxygen Demand (BOD) concentration (Jenni et al., 2014). The only difference between scenarios 3 and 4 was the acidification step with the addition of sulphuric acid to the digestate before centrifugation aiming at increasing P solubility, thus augmenting its availability for struvite precipitation (Daumer et al., 2010).

### 2.2. Inventory data: Mass balances

Since it is not possible to test at pilot scale all feasible scenarios, steady-state simulation is a powerful tool to generate detailed inventory data of a process integrating the experimental and literature data of each single unit. This methodological approach was applied to perform solid, organic matter, N and P mass balances in the seven scenarios. The hypotheses taken to set the mass balances are summarized in Table 2 and the average removal efficiencies of total solids (TS), COD, TKN and TP in each unit are listed in Table S1. The performance of the decanter centrifuge depended on the pH of the digestate (Table S1), as observed in other studies (Daumer et al., 2010). Concerning BOD, no removal occurred beyond that of water partition (Burton, 2007). Also the removal efficiencies in the filtration unit depended on the pH value of the digestate (Narong and James, 2006). All soluble COD in the permeate was assumed as BOD. Pure sulphuric acid was dosed to the digestate before the centrifuge in scenario 4 to lower pH up to values around 4.5–5, thus avoiding P precipitation in the solid/liquid separation step (Daumer et al., 2010). As a consequence, the buffering capacity was mostly removed. The struvite crystallizer was designed according to Tarragó et al. (2016). As during the pilot plant operation struvite presence in the precipitate was detected but not quantified, the performance of this unit was assessed using the Visual Minteq 3.1 software, a chemical equilibrium model which, once the main ions present in an aqueous solution at a given pH and temperature are determined, can predict their chemical speciation and the solubility and/or precipitation of solid phases. Biological nitrogen removal was not conducted in the pilot plant, and thus, all data for mass balances come from literature (Table 2). Both nitritation/denitrification unit and HRAS+PNA units worked as a sequencing batch reactor (SBR). In case of organic matter deficiency in the nitritation/denitrification, BOD supplementation (80% glycerol solution) was considered. If the buffering capacity was not sufficient, NaHCO<sub>3</sub> dosage was assumed. In case that P was limiting, it was simulated to be supplied in form of phosphoric acid. Organic N was supposed not to be consumed during this biological process. In the HRAS+PNA combination, the first compartment was employed to lower the organic matter concentration in contact with anammox biomass (Daigger, 2014). In case that P was limiting, the dosage of phosphoric acid was again considered. The nitrate produced during the anammox process was assumed to be consumed by the small portion of heterotrophic

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