



## Review

# Membrane-based processes for wastewater nutrient recovery: Technology, challenges, and future direction



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## ABSTRACT

Wastewater nutrient recovery holds promise for more sustainable water and agricultural industries. We critically review three emerging membrane processes – forward osmosis (FO), membrane distillation (MD) and electrodialysis (ED) – that can advance wastewater nutrient recovery. Challenges associated with wastewater nutrient recovery were identified. The advantages and challenges of applying FO, MD, and ED technologies to wastewater nutrient recovery are discussed, and directions for future research and development are identified. Emphasis is given to exploration of the unique mass transfer properties of these membrane processes in the context of wastewater nutrient recovery. We highlight that hybridising these membrane processes with existing nutrient precipitation process will lead to better management of and more diverse pathways for near complete nutrient recovery in wastewater treatment facilities.

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## 1. Introduction

We face a major grand challenge in the twenty-first century: sustainably meeting food demands while simultaneously reducing agriculture's environmental harm (Foley et al., 2011; West et al., 2014). This challenge is being exemplified as an annual increase of 4% in fertiliser demand to feed additional 2.3 billion people by 2050, thereby requiring a sustained supply of fertilisers (Elser and Bennett, 2011).

Current fertiliser production heavily relies on the consumption of non-renewable energy and finite mineral resources. For example, the generation of ammonia from air in the Haber-Bosch process requires 35–50 MJ per kg nitrogen in the form of fossil fuel for energy supply (Desloover et al., 2012), which accounts for 2% of the world energy use. Phosphorus mining leads to a huge amount of gypsum by-products that are contaminated with heavy metals and radioactive elements (Ashley et al., 2011). More alarming, the forecasted phosphorus production peak is approaching in 2030, with an accelerated depletion of minable phosphorus rock (Elser and Bennett, 2011).

The use of fertiliser to meet food demand also carries a heavy burden for wastewater treatment processes. Once through production and application of fertilisers results in major nutrients (nitrogen and phosphorus) being primarily found in wastewater. It is estimated that 30% of nitrogen and 16% of phosphorus in fertilisers ends up in wastewater (Rahman et al., 2014; Verstraete et al., 2009). Consequently, wastewater treatment facilities consume up to 4% electrical energy in the United States (Energy, 2006; EPA and Water, 2006), more than 77% of which is used for activated sludge aeration for nitrification (McCarty et al., 2011; Svardal and Kroiss, 2011). The removal of nitrogen from wastewater requires substantial energy, 45 MJ per kg nitrogen, only to release it back as gaseous nitrogen into the atmosphere. This energy-intense nutrient removal also contributes to greenhouse gas emission of 0.9 kg CO<sub>2</sub> per cubic litre of treated wastewater (Hall et al., 2011; Rothausen and Conway, 2011). The large energy and environmental footprint of nutrient removal from wastewater, in turn, aggravates the sustainability of fertiliser production for food security. As a result, wastewater nutrient recovery is anticipated to become a promising strategy to sustain fertiliser and food production, and at the same time, potentially bring benefits to wastewater treatment facilities (Grant et al., 2012; Guest et al., 2009; Verstraete et al., 2009).

High-rejection membrane processes, such as nanofiltration (NF)

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and reverse osmosis (RO), have demonstrated huge potential in wastewater nutrient recovery. For example, RO was applied for urine concentration in a source-separation toilet system, achieving a concentration factor of five and high rejection of ammonium, phosphate and potassium (Maurer et al., 2006). NF separation also exhibited medium to high rejection of a range of nutrients, such as urea (Pronk et al., 2006b), ammonium, phosphate and potassium (Blöcher et al., 2012; Niewersch et al., 2014). Despite the potential of NF and RO processes in wastewater nutrient recovery, current pressure-driven membrane processes are not without limitations. NF and RO processes are prone to membrane fouling in wastewater nutrient recovery where the feed streams are challenging and difficult to treat, such as urine and digested sludge. Fouling of NF and RO membranes impairs membrane performance and shortens membrane lifetime, thereby restraining productivity in nutrient recovery. Hence, there is a critical need for robust separation processes for nutrient recovery from challenging wastewater streams.

We critically review membrane processes that enable the reclamation of nutrients from wastewater and illustrate the challenges for membrane processes in wastewater nutrient recovery. Emerging membrane processes — forward osmosis (FO), membrane distillation (MD), and electrodialysis (ED) — are discussed and evaluated based on their applications, nutrient recovery potential, and process limitations. Unique challenges associated with the agricultural application of recovered nutrients are also elucidated.

## 2. Existing technology illustrates challenges for wastewater nutrient recovery

Struvite ( $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$ ) precipitation is widely accepted as the most promising technology in wastewater nutrient recovery (de-Bashan and Bashan, 2004). Struvite is a slow-release fertiliser, applicable to crops in soils with relatively low pH value. In the process of nutrient recovery via struvite precipitation, an alkaline solution is obtained either by addition of basic solution or aeration stripping of  $\text{CO}_2$ , and followed by the introduction of magnesium salts for struvite precipitation. Previous studies have demonstrated nutrient recovery via struvite precipitation from various nutrient-rich streams, such as wastewater (Gerardo et al., 2013; Ichihashi and Hirooka, 2012), anaerobically digested sludge (Battistoni et al., 2005; Lahav et al., 2013; Marti et al., 2008; Pastor et al., 2010; Quintana et al., 2003), and urine (Ronteltap et al., 2010; Trigger et al., 2012). Despite the struvite precipitation reaching commercial implementation for nutrient recovery, there remains two critical challenges in wastewater nutrient recovery via struvite precipitation.

The efficiency of nutrient recovery via struvite precipitation is limited by the phosphorus concentration in wastewater. The driving force and kinetics for struvite precipitation are significantly influenced by the phosphorus concentration. Extensive experimental results showed that effective struvite precipitation could only be achieved when the phosphorus concentration was above 100 mg/L (Fig. 1A) (Çelen et al., 2007; Guadie et al., 2014; Jaffer et al., 2002; Liu et al., 2011; Münch and Barr, 2001; Pastor et al., 2008, 2010; Ronteltap et al., 2010; Song et al., 2011). Low phosphorus concentration resulted in either low (<40%) struvite recovery or a longer precipitation reaction time, which substantially impaired the economic feasibility of nutrient recovery via struvite precipitation. The demand for high phosphorus concentration is challenging for wastewater where typical phosphorus concentrations for wastewater influent and digested sludge supernatant were 6 and 56 mg/L, respectively (Jaffer et al., 2002; Münch and Barr, 2001). As a result, it is desirable to enrich nutrients in the waste stream prior to struvite precipitation, thereby significantly

enhancing the struvite precipitation potential and efficiency.

Struvite precipitation for nutrient recovery is also challenged by the presence of toxic heavy metal ions and emerging organic contaminants in wastewater (Pronk et al., 2006b), which substantially compromises struvite purity and safe agricultural application. For example, a close examination of recovered struvite crystals revealed the presence of toxic heavy metals in struvite, with arsenic concentration up to 570 mg/kg (Fig. 1B) (Lin et al., 2013; Ma and Rouff, 2012; Pizzol et al., 2014; Rouff, 2012; Rouff and Juarez, 2014). The presence of such contaminants in struvite fertiliser is strictly regulated and excessive amounts can result in the fertiliser being banned from agricultural application.

Alternative nutrient recovery approaches with better selectivity should be considered to improve the nutrient product quality. For example, instead of struvite precipitation, ammonium can be recovered under alkaline condition by membrane distillation as 10% ammonia solution (Bonmati and Flotats, 2003; Jorgensen and Weatherley, 2003); and phosphorus can be fractionated as phosphoric acid by electrodialysis (Wang et al., 2013; Zhang et al., 2013a). These nutrient recovery technologies targeting specific nutrient ions demonstrated better selectivity and resulted in nutrient products with higher quality.

## 3. Emerging membrane processes advance wastewater nutrient recovery

The challenges of higher nutrient enrichment and membrane selectivity discussed above (Section 2) open opportunities for emerging membrane processes to advance wastewater nutrient recovery. Forward osmosis (FO), membrane distillation (MD) and electrodialysis (ED) are three membrane-based processes that are best suited to overcome the challenges in wastewater nutrient recovery, and could potentially represent a paradigm shift in wastewater nutrient management (Table 1). As described herein, these technologies can achieve high concentration factor for struvite precipitation, their selectivity is conducive to the fraction of valuable nutrient substances in various formats, and their energy requirements and associated costs are competitive with more conventional, pressure-driven membrane processes. A process overview of the three technologies is presented below and the advantages and disadvantages of each for wastewater nutrient recovery are discussed.

### 3.1. Forward osmosis

Forward osmosis (FO) could substantially enhance wastewater nutrient recovery via struvite precipitation by its unique mass transfer properties: lack of hydraulic pressure and the occurrence of reverse draw solute flux. In FO, a semipermeable membrane is placed between two solutions of different concentrations: a concentrated draw solution and a more dilute feed solution. Instead of hydraulic pressure, FO employs an osmotic pressure difference to drive the permeation of water across the membrane. As a result, FO has demonstrated a lower fouling propensity and higher fouling reversibility in comparison with pressure-driven RO membrane filtration (Lee et al., 2010; Mi and Elimelech, 2010). Consequently, FO enables concentration of a range of challenging, nutrient-rich streams, achieving high enrichment factors for streams (Table 1), such as anaerobically digested sludge (Holloway et al., 2007), activated sludge (Achilli et al., 2009; Cornelissen et al., 2008) and raw sewage (Cath et al., 2005; Xie et al., 2013, 2014a; Xue et al., 2015).

Reverse draw solute diffusion, an inherent phenomenon commonly considered detrimental to FO (Boo et al., 2012; Xie et al., 2014b), can be beneficial by elevating struvite precipitation

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