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4 **Membrane-based Processes for Wastewater**
5 **Nutrient Recovery: Technology, Challenges, and**
6 **Future Direction**

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25 **ABSTRACT**

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

37 **1. Introduction**

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

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

51 The use of fertiliser to meet food demand also carries a heavy burden for wastewater
52 treatment processes. Once through production and application of fertilisers results in major
53 nutrients (nitrogen and phosphorus) being primarily found in wastewater. It is estimated that
54 30% of nitrogen and 16% of phosphorus in fertilisers ends up in wastewater (Rahman et al.
55 2014, Verstraete et al. 2009). Consequently, wastewater treatment facilities consume up to 4%
56 electrical energy in the United States (Energy 2006, EPA and Water 2006), more than 77% of
57 which is used for activated sludge aeration for nitrification (McCarty et al. 2011, Svardal and
58 Kroiss 2011). The removal of nitrogen from wastewater requires substantial energy, 45 MJ
59 per kg nitrogen, only to release it back as gaseous nitrogen into the atmosphere. This energy-
60 intense nutrient removal also contributes to greenhouse gas emission of 0.9 kg CO₂ per cubic
61 litre of treated wastewater (Hall et al. 2011, Rothausen and Conway 2011). The large energy
62 and environmental footprint of nutrient removal from wastewater, in turn, aggravates the
63 sustainability of fertiliser production for food security. As a result, wastewater nutrient
64 recovery is anticipated to become a promising strategy to sustain fertiliser and food
65 production, and at the same time, potentially bring benefits to wastewater treatment facilities
66 (Grant et al. 2012, Guest et al. 2009, Verstraete et al. 2009).

67 High-rejection membrane processes, such as nanofiltration (NF) and reverse osmosis
68 (RO), have demonstrated huge potential in wastewater nutrient recovery. For example, RO

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

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

86 **2. Existing technology illustrates challenges for wastewater nutrient recovery**

87 Struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) precipitation is widely accepted as the most promising
88 technology in wastewater nutrient recovery (de-Bashan and Bashan 2004). Struvite is a slow-
89 release fertiliser, applicable to crops in soils with relatively low pH value. In the process of
90 nutrient recovery via struvite precipitation, an alkaline solution is obtained either by addition
91 of basic solution or aeration stripping of CO_2 , and followed by the introduction of magnesium
92 salts for struvite precipitation. Previous studies have demonstrated nutrient recovery via
93 struvite precipitation from various nutrient-rich streams, such as wastewater (Gerardo et al.
94 2013, Ichihashi and Hirooka 2012), anaerobically digested sludge (Battistoni et al. 2005,
95 Lahav et al. 2013, Marti et al. 2008, Pastor et al. 2010, Quintana et al. 2003), and urine
96 (Ronteltap et al. 2010, Triger et al. 2012). Despite the struvite precipitation reaching
97 commercial implementation for nutrient recovery, there remains two critical challenges in
98 wastewater nutrient recovery via struvite precipitation.

99 The efficiency of nutrient recovery via struvite precipitation is limited by the
100 phosphorus concentration in wastewater. The driving force and kinetics for struvite

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

113 Struvite precipitation for nutrient recovery is also challenged by the presence of toxic
114 heavy metal ions and emerging organic contaminants in wastewater (Pronk et al. 2006b),
115 which substantially compromises struvite purity and safe agricultural application. For
116 example, a close examination of recovered struvite crystals revealed the presence of toxic
117 heavy metals in struvite, with arsenic concentration up to 570 mg/kg (Figure 1B) (Lin et al.
118 2013, Ma and Rouff 2012, Pizzol et al. 2014, Rouff 2012, Rouff and Juarez 2014). The
119 presence of such contaminants in struvite fertiliser is strictly regulated and excessive amounts
120 can result in the fertiliser being banned from agricultural application.

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

128 **[Figure 1]**

129 **3. Emerging membrane processes advance wastewater nutrient recovery**

130 The challenges of higher nutrient enrichment and membrane selectivity discussed
131 above (Section 2) open opportunities for emerging membrane processes to advance
132 wastewater nutrient recovery. Forward osmosis (FO), membrane distillation (MD) and

133 electro dialysis (ED) are three membrane-based processes that are best suited to overcome the
134 challenges in wastewater nutrient recovery, and could potentially represent a paradigm shift
135 in wastewater nutrient management (Table 1). As described herein, these technologies can
136 achieve high concentration factor for struvite precipitation, their selectivity is conducive to
137 the fraction of valuable nutrient substances in various formats, and their energy requirements
138 and associated costs are competitive with more conventional, pressure-driven membrane
139 processes. A process overview of the three technologies is presented below and the
140 advantages and disadvantages of each for wastewater nutrient recovery are discussed.

141 [Table 1]

142 3.1. Forward osmosis

143 Forward osmosis (FO) could substantially enhance wastewater nutrient recovery via
144 struvite precipitation by its unique mass transfer properties: lack of hydraulic pressure and the
145 occurrence of reverse draw solute flux. In FO, a semipermeable membrane is placed between
146 two solutions of different concentrations: a concentrated draw solution and a more dilute feed
147 solution. Instead of hydraulic pressure, FO employs an osmotic pressure difference to drive
148 the permeation of water across the membrane. As a result, FO has demonstrated a lower
149 fouling propensity and higher fouling reversibility in comparison with pressure-driven RO
150 membrane filtration (Lee et al. 2010, Mi and Elimelech 2010). Consequently, FO enables
151 concentration of a range of challenging, nutrient-rich streams, achieving high enrichment
152 factors for streams (Table 1), such as anaerobically digested sludge (Holloway et al. 2007),
153 activated sludge (Achilli et al. 2009, Cornelissen et al. 2008) and raw sewage (Cath et al.
154 2005, Xie et al. 2013, 2014a, Xue et al. 2015).

155 Reverse draw solute diffusion, an inherent phenomenon commonly considered
156 detrimental to FO (Boo et al. 2012, Xie et al. 2014b), can be beneficial by elevating struvite
157 precipitation potential via supplementing magnesium cation into the feed when magnesium-
158 based draw solution is used (Figure 2). Recent studies demonstrated this proof-of-concept of
159 FO in nutrient recovery (Xie et al. 2013, 2014a). Feed sludge centrate was concentrated by
160 FO driven by $MgCl_2$ draw solution and achieved a concentration factor of five, resulting in a
161 high strength stream comprising ammonium (1210 mg/L), phosphate (615 mg/L), and
162 magnesium from reverse magnesium flux. As a result, the $MgCl_2$ draw solution not only
163 provides the driving force for nutrient enrichment, but also can be incorporated into the
164 nutrient precipitate, which makes beneficial use of lost draw solution. These unique mass

165 transfer properties of FO motivate nutrient recovery from various waste streams such as urine
166 (Gormly and Flynn 2007, Michael et al. 2012, Zhang et al. 2014a), sewage (Ge et al. 2012,
167 Hancock et al. 2013, Phuntsho et al. 2012, Wang et al. 2011a, Xie et al. 2013, Zhang et al.
168 2014b, Zhang et al. 2013b), and sludge (Hau et al. 2014, Holloway et al. 2007, Nguyen et al.
169 2013).

170 [Figure 2]

171 Experimental results from the aforementioned literature were corroborated by
172 mathematical modelling, illustrating promising potential and capacity of the FO process in
173 wastewater nutrient recovery. For instance, Xue et al. (2015) calculated theoretical water
174 recovery of 93% in an FO process using seawater draw solution, thereby achieving ten-fold
175 concentration of ammonium and phosphate in the secondary treated municipal wastewater.
176 This high nutrient enrichment factor also agreed with the solution-diffusion model for FO
177 filtration (Zhang et al. 2014a), yielding 50–80% rejection of ammonium and above 90%
178 rejection of phosphate and potassium.

179 Despite the feasibility of nutrient recovery by FO, membrane performance is
180 constrained by the water permeability – solute selectivity tradeoff (Yip and Elimelech 2011),
181 an intrinsic property of water and solute transport through polymeric membranes. This
182 tradeoff restricts attainment of high water permeability for FO membrane materials without
183 decreasing solute selectivity (Freeman 1999, Geise et al. 2011), which limits the achievement
184 of high nutrient concentration factor. For nutrient recovery, a membrane with high solute
185 selectivity effectively enriches ammonium and phosphate, and hence, yields a high strength
186 nutrient-rich stream. However, lack of sufficient cations, particularly magnesium
187 supplemented into this stream via reverse salt flux, reduces struvite precipitation potential. By
188 contrast, a membrane with high water permeability produces a higher water flux, and the
189 concomitant decline in membrane selectivity simultaneously provides more draw solution
190 cations to the feed due to higher reverse salt flux, while also causing significant loss of
191 nutrient solutes into the draw. Such detrimental effects work against the benefit of a more
192 permeable but less selective membrane to enhance struvite product yield. Therefore, further
193 understanding membrane permeability – selectivity tradeoff is crucial to nutrient recovery by
194 FO process.

195 3.2. Membrane distillation

196 Membrane distillation (MD) is a thermally-driven membrane process that can utilise
197 low-grade heat to drive separation (Alkhudhiri et al. 2012, Alklaibi and Lior 2005). In MD,
198 the aqueous feed stream is separated from the distillate by a hydrophobic, microporous
199 membrane. Liquid is unable to penetrate the membrane pores due to the hydrophobic nature
200 of the membrane, and a difference in the partial vapour pressure drives the transport of water
201 vapour across the membrane pores. Because water is transported through the membrane only
202 in a vapour phase, MD can offer complete rejection of all non-volatile constituents in the feed
203 solution. More importantly, MD could achieve high water recovery because water vapour
204 transport through MD membrane is not significantly influenced by the feed osmotic pressure.

205 Due to this unique transport mechanism, MD processes have been explored for the
206 recovery of valuable components. Based on the volatility and vapour pressure, these
207 components can be concentrated either in the feed stream or permeate streams. For example,
208 non-volatile inorganic nutrient ions, such as potassium and phosphate, can be concentrated in
209 the feed stream to facilitate subsequent nutrient precipitation. Indeed, the MD process
210 achieved a high concentration factor of three for seawater RO brine volume reduction
211 (Martinetti et al. 2009). Similar high enrichment performance could also be observed for
212 mineral acids (Elkina et al. 2013, Tomaszewska et al. 1995, Tomaszewska 2000) and fruit
213 juices (Mohammadi and Bakhteyari 2006). Concentration of sulphuric acid by MD from 16%
214 until 40% was reported with a separation coefficient of above 98% (Tomaszewska and
215 Mientka 2009).

216 Ammonia recovery can be one important application of MD process in wastewater
217 nutrient recovery where ammonia is more volatile than water and can be enriched in the
218 permeate stream of MD processes (du Preez et al. 2005, Zarebska et al. 2014, Zhao et al.
219 2013). Ammonia recovery exemplifies the selectivity of MD membrane process, an approach
220 that is different from aiming for high nutrient concentration factor. MD processes were
221 configured as vacuum MD, gas sweeping MD and direct contact MD for ammonia recovery
222 from varying waste streams, such as urine (Zhao et al. 2013), wastewater (Ahn et al. 2011,
223 Ding et al. 2006, El-Bourawi et al. 2007, Qu et al. 2013, Xie et al. 2009), and swine manure
224 (Thygesen et al. 2014, Zarebska et al. 2014). These MD processes achieved more than 96%
225 ammonia recovery in the form of aqueous solution, which can be conveniently processed as
226 commercial fertiliser. More importantly, in direct contact MD, low concentration of sulphuric
227 acid was used as stripping solution on the permeate side to further enhance the capture of
228 ammonia vapour. The application of acidic stripping solution in the MD permeate stream

229 substantially improves ammonia recovery to 99%, with ammonium sulphate being produced
230 as fertiliser.

231 Varying composition of nutrient-rich waste stream can pose distinctive challenges to
232 MD in nutrient recovery (Meng et al. 2014, Tijing et al. 2015, Van der Bruggen 2013). The
233 volatile organic compounds, such as volatile fatty acids that exert partial vapour pressures
234 comparable to or higher than water are transported across the MD membrane with the water
235 vapour, causing contamination of the permeate stream and jeopardising the quality of
236 recovered ammonia fertiliser. Certain components in wastewater, such as surfactants, can
237 lower the liquid surface tension of the feed solution and cause wetting of the membrane pores.
238 Membrane pore wetting will result in a direct liquid flow from feed through the wetted pores,
239 substantially deteriorating distillate quality. To restore the vapour-liquid interface at the pores,
240 the wetted membrane must be taken out of operation and dried completely, resulting in
241 process downtime and potential membrane degradation. Dissolved organic matters and
242 colloids present in the nutrient-rich waste streams can lead to MD membrane fouling. Fouling
243 clogs membrane pores, which leads to flux decline and pore wetting and imposes additional
244 hindrance to heat and mass transfer, thereby diminishing the MD process productivity in
245 nutrient recovery. Indeed, in ammonia recovery from wastewater by MD process, MD
246 membrane fouling was initiated by adsorption of peptides and proteins on MD membrane
247 surface, and thus reduced Gibbs free energy and hydrophilised the membrane surface, thereby
248 hindering ammonia vapour permeation (Thygesen et al. 2014, Zarebska et al. 2014).

249 **[Figure 3]**

250 Fabrication of MD membranes with special wettability, such as superhydrophobic or
251 omniphobic property, imparts membrane anti-fouling property and mitigates deleterious
252 membrane fouling and wetting, thereby improving the nutrient recovery efficiency of MD in
253 processing challenging waste streams (Liao et al. 2013, 2014, Lin et al. 2014). Increasing the
254 hydrophobicity of an MD membrane leads to higher liquid entry pressure and consequently
255 more resistance to pore wetting. For example, Razmjou et al. (2012) fabricated a
256 superhydrophobic polyvinylidene fluoride (PVDF) MD membrane with TiO₂ nanoparticles
257 providing hierarchical structures with multilevel roughness on the membrane surface. The
258 resultant MD membrane, possessing high liquid entry pressure of 195 kPa, demonstrated a
259 much higher water flux recovery after humic acid fouling in comparison to the pristine PVDF
260 membrane. Another strategy for preventing membrane fouling and wetting is fabrication of
261 MD membrane with omniphobic property that repels both water and low surface tension

262 liquids such as surfactants. Indeed, Lin and coworkers (2014) fabricated an omniphobic MD
263 membrane with silica nanoparticles via surface fluorination and polymer coating, and
264 demonstrated anti-wetting MD membrane performance maintaining water flux and salt
265 rejection, even with the presence of surfactant. The advancement of MD membrane
266 fabrication with special wettability can open up vast opportunities for MD application for
267 beneficial nutrient recovery, particularly ammonia, from challenging waste streams.

268 3.3. Electrodialysis

269 Electrodialysis (ED), which arranges ion-exchange membranes alternately in a direct
270 current field (Xu and Huang 2008), could selectively fraction nutrients from wastewater
271 streams into high quality nutrient products. The direct current field is the driving force in an
272 ED process where cations and anions migrate towards the cathode and anodes, respectively.
273 The ion separation in ED process is achieved by ion-exchange membranes that comprise
274 cation-selective, anion-selective, and bipolar membranes. Cation- and anion-selective
275 membranes are widely used in conventional ED to hinder the passage of co-ions (anions and
276 cations, respectively) by virtue of Donnan repulsion. When bipolar membranes comprising a
277 cation-selective layer and an anion-selective layer are used in an ED process, dissociation of
278 solvent molecules, such as water, into H^+ and OH^- can be realised.

279 The unique ion separation mechanism of ED process provides a selective mechanism
280 for wastewater nutrient recovery. ED process selectively partitioned phosphate from
281 wastewater effluent containing various ions as a concentrated phosphate solution, achieving a
282 concentration factor of up to 7 (Zhang et al. 2012, Zhang et al. 2013a). Similar selective
283 phosphate enrichment by ED process was also observed in urine nutrient recovery, resulting
284 in a purified phosphate concentrate (Escher et al. 2006, Pronk et al. 2006a). Phosphate
285 selectivity in an ED process can be further enhanced by either adjusting the feed stream to the
286 alkaline pH range or increasing current density (Tran et al. 2014, Tran et al. 2015). Better
287 performance was expected based on the ED separation mechanisms where multivalent
288 phosphate migrates more slowly than monovalent ions under the current field (Zhang et al.
289 2012).

290 Nutrient recovery efficiency and product purity could be significantly improved when
291 bipolar membrane was employed in an ED process. The ED process with bipolar membrane
292 integrates solvent (water) and salt dissociation (Bailly 2002); it provides H^+ and OH^- *in situ*
293 without the introduction of salts (Huang and Xu 2006, Huang et al. 2006, Huang et al. 2007).

294 The combination of H^+ and anions in certain chambers leads to production of acid, while the
295 combination of OH^- ions and cations in other chambers leads to production of the
296 corresponding base. As a result, this ED process with bipolar membrane concept could
297 diversify the final products and enhance purity for nutrient recovery. For example, Wang et al.
298 (2013) employed the ED process with bipolar membrane to convert phosphate in sludge
299 supernatant to purified phosphoric acid of 0.075 mol/L, which provided an approach for
300 wastewater nutrient recovery.

301 Despite the high purity and diverse product extracted by ED process, it suffers from
302 membrane fouling during wastewater nutrient recovery. The build-up of fouling layers in ED
303 process increases the cell resistance (current drop), decreases migration yield and ion
304 selectivity, and eventually alters membranes due to irreversible fouling (Mondor et al. 2009).
305 Unlike fouling in RO and FO membranes, the fouling of ion-exchange membrane in ED
306 process is significantly dependent on the charge of the membrane (Wang et al. 2011b).
307 Specifically, more severe fouling was observed in anion-selective membrane when negatively
308 charged humic substance, protein and surfactant were presented (James Watkins and Pfromm
309 1999, Lee et al. 2009, Lindstrand et al. 2000). By contrast, cation-selective membrane could
310 be hampered by calcium-dominated scaling (Ayala-Bribiesca et al. 2006, Bazinet and Araya-
311 Farias 2005). Abating ED membrane fouling could be achieved by periodically reversing the
312 polarity of electrodes, decreasing current density, improving hydraulic conditions in stack
313 compartment by increasing flowrate or gasket with flow pattern, and in-place cleaning with
314 acidic or basic solutions (Lee et al. 2002, Mondor et al. 2009, Ruiz et al. 2007).

315 **[Figure 4]**

316 **4. Path forward**

317 *4.1. 1+1>2*

318 The emerging membrane processes discussed above have demonstrated their capacity
319 to advance wastewater nutrient recovery by either maximising nutrient concentration factors,
320 such as FO and MD, or enhancing nutrient selectivity, for instance MD and ED. Hybrid
321 membrane processes complement each other, thereby maximising overall nutrient recovery
322 efficiency.

323 For example, the requirement for concentrating the diluted draw solution in an FO
324 process opens opportunity for coupling with other membrane processes (e.g., RO or MD) to
325 simultaneously restore the FO driving force and to produce high quality freshwater (Hoover

326 et al. 2011, Xie et al. 2013, 2014a). Integration of FO with other processes could not only re-
327 concentrate diluted draw solution for sustainable process performance, but also complement
328 wastewater nutrient recovery with freshwater production. This concept motivates coupling
329 FO with different membrane processes, such as RO, MD, and ED (Figure 5). For example, an
330 FO-RO hybrid system can achieve high rejections of phosphate and ammonium (99.9% and
331 92%, respectively) from wastewater effluent (Hancock et al. 2013, Holloway et al. 2007) or
332 nutrient-rich sludge (Nguyen et al. 2013). More importantly, this hybrid system also
333 simultaneously produces high quality permeate water. In an FO-MD hybrid system, FO
334 concentrated orthophosphate and ammonium for subsequent phosphorus recovery in the form
335 of struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$), while MD was used to recover the draw solution and extract
336 clean water from the digested sludge centrate (Xie et al. 2013, 2014a). The MD unit in the
337 hybrid system can also be powered by solar energy, thereby reducing the overall operating
338 cost for wastewater nutrient recovery (Qtaishat and Banat 2013). Similarly, ED can also levy
339 solar photovoltaic energy to re-concentrate draw solution in an FO-ED hybrid system (Zhang
340 et al. 2013b), thereby simultaneously concentrating nutrient from the feed waste, and
341 producing freshwater from the draw solution.

342 Coupling FO with a membrane bioreactor (MBR) is also becoming attractive for
343 wastewater nutrient recovery (Holloway et al. 2014, Holloway et al. 2015). This osmotic
344 MBR concept substantially benefits from the high nutrient rejection by the FO membrane in
345 MBR, high concentration factor due to low FO fouling propensity, and supply of magnesium
346 ions to facilitate nutrient precipitation via reverse draw solution diffusion (Figure 3).
347 Recently, Qiu and Ting (2014) applied an osmotic MBR using MgCl_2 draw solution to
348 directly extract phosphorus from wastewater, achieving 95% phosphorus recovery via
349 calcium phosphate precipitation. Subsequently, Qiu and coworkers employed an OMBR with
350 seawater brine draw to achieve 90% phosphorus recovery in the form of amorphous calcium
351 phosphate (Qiu et al. 2015).

352 [Figure 5]

353 Despite the versatility and robustness of hybridized FO system for wastewater
354 nutrient recovery, this technology is not without limitations. One significant hindrance is
355 contaminant accumulation in the draw solution. In the closed-loop FO hybrid system,
356 contaminants that permeate through the FO but not the downstream RO or MD process can
357 accumulate in the draw solution, leading to a build-up of unfavourable contaminants in the
358 draw solution (D'Haese et al. 2013, Shaffer et al. 2012). Significant accumulation of organic

359 foulants in the draw solution was observed in an FO-RO hybrid system (Coday et al. 2015,
360 Hancock et al. 2013). Similarly, this detrimental effect was also found in an FO-MD hybrid
361 system where micropollutant concentrations increased in the draw solution as the cumulative
362 permeate volume increased (Xie et al. 2013, 2014a). Therefore, it is of paramount importance
363 to manage this detrimental contaminant accumulation to ensure system performance and
364 reliability.

365 Struvite precipitation, a key step for phosphorus nutrient recovery, could also benefit
366 from coupling with membrane processes to improve precipitation efficiency. For example,
367 ED process was operated with struvite precipitation reactor in tandem, which enhanced the
368 selective capture of phosphate from the effluent of the struvite reactor (Zhang et al. 2013a).
369 As a result, the phosphate from struvite reactor effluent was further concentrated in ED stack
370 and recirculated into the struvite reactor, thereby improving the overall phosphorus recovery
371 to 97%.

372 Ammonia recovery, which could result in high quality liquid fertiliser, can be
373 substantially advanced by hybrid membrane processes. In an ED-RO hybrid system, the
374 ammonium was fractionated by cation-selective membrane in an ED unit where the
375 ammonium-rich stream from concentrate compartments was further concentrated by RO
376 membrane. This ED-RO hybrid process produced highly concentrated ammonium solution up
377 to 13 g/L (Mondor et al. 2008), which is beneficial in agricultural application. On the other
378 hand, the volatile ammonia could be captured in an ED-MD hybrid process (Ali et al. 2004,
379 Graillon et al. 1996, Udert and Wächter 2012). For instance, ED process with bipolar
380 membranes produced ammonia from ammonium nitrate waste stream via splitting water
381 solvent. The produced ammonia was recovered by stripping under vacuum membrane
382 distillation, achieving an ammonia concentration of 2 mol/L.

383 *4.2. Decentralised or centralised?*

384 Key nutrient concentrations – ammonium and phosphate – decrease along the sewer
385 system from household to a centralised wastewater treatment facilities, with phosphate
386 concentration being 100 times higher from single household in comparison to the
387 concentration at the wastewater treatment plant (1991, Carroll et al. 2006, Chanan and Woods
388 2006, Maurer et al. 2003). This significant variation of nutrient concentration gradient
389 unlocks opportunities for tailoring nutrient recovery approaches with varying membrane

390 processes for both decentralised (e.g., single household) and centralised (e.g., wastewater
391 treatment plant) applications.

392 On-site urine separation and recovery via struvite precipitation is one promising
393 strategy for decentralised, small-scale wastewater nutrient recovery (Larsen et al. 2009). In
394 particular, the urine stream contributes a large proportion of nutrients from household (81%
395 nitrogen, 50% phosphorus, and 55% potassium), but less than 1% of the total volume of
396 municipal wastewater (Karak and Bhattacharyya 2011). More importantly, via nutrient
397 recovery, on-site urine separation can significantly reduce nutrient loadings to the wastewater
398 treatment plants and downstream effluent-receiving water bodies (Ishii and Boyer 2013,
399 Wilsenach and Loosdrecht 2003). As such, on-site nutrient recovery from urine can be more
400 energetically efficient than nutrient removal and recovery in centralised wastewater treatment
401 process, despite low economic efficacy of small-scale system. However, the deployment of
402 urine source separation requires substantial change to existing infrastructure, such as varying
403 flush water from urine diverting toilets (Wilsenach and Van Loosdrecht 2004), proper urine
404 storage for urea hydrolysis (Ishii and Boyer 2015), and precipitation in urine-separating
405 toilets (Udert et al. 2003). In addition, the deployment of on-site nutrient recovery from urine
406 also encounters varying degree of acceptance. For example, fertiliser produced by urine was
407 less accepted by farmers in comparison with the public, where more than 50% farmers have
408 concerns in technical feasibility as well as nutrient product quality (Lienert and Larsen 2010).

409 Emerging membrane processes discussed above also exhibit satisfactory performance
410 in urine separation. For instance, FO process mined macronutrients (nitrogen, phosphate and
411 potassium) from urine after hydrolysis, achieving significant volume reduction, and high
412 rejection of ammonium (50-80%), phosphate and potassium (>90%) (Zhang et al. 2014a). A
413 higher ammonia separation factor from urine could be observed in a vacuum MD process
414 where rejection of ammonia reached 99% (El-Bourawi et al. 2007, Zhao et al. 2013). ED
415 process is also capable of recovering and concentrating nutrient ions from urine contaminated
416 by micropollutants (Pronk et al. 2006a).

417 Wastewater nutrient recovery has been practised in centralised wastewater treatment
418 facility (Cote et al. 2013, Kuzma et al. 2012). However, the benefit of upgrading
419 conventional wastewater treatment process goes beyond economic gains (McConville et al.
420 2014), as re-engineering the existing nutrient recovery process by a suite of membrane
421 processes offers more sustainable sewage management and nutrient cycling.

422 4.3. Energy consumption and bioavailability for recovered nutrients

423 The reviewed three emerging membrane processes for wastewater nutrient recovery
424 could utilise a range of renewable energy to further enhance the process sustainability and
425 substantially reduce the carbon footprint. For instance, via powering the ED process by solar
426 energy, the operating cost for an FO-ED hybrid process was €3.32 to 4.92 per cubic meter
427 treated water (considering the investment for membranes and solar panel) for a small size
428 (Zhang et al. 2013b). In addition, levying waste heat generated by a power plant (Zhou et al.
429 2015) or introducing the biogas produced by the wastewater treatment plant (Qin and He
430 2014) can be an important alternative to drive the MD process with less cost in wastewater
431 nutrient recovery. Furthermore, a life cycle assessment showed that more than 25% reduction
432 of the environmental impact could be achieved when incorporating FO process into
433 traditional seawater desalination or wastewater reclamation process (Hancock et al. 2012).

434 Producing agriculturally applicable fertiliser is the final goal for wastewater nutrient
435 recovery. Apart from the technological aspect of wastewater nutrient recovery, more attention
436 should be also paid to the agronomic efficacy and crop uptake of the fertiliser produced from
437 recovered nutrients (Withers et al. 2014). Phosphorus plant availability of struvite precipitate
438 recovered from waste stream was compared with a well-established, water-soluble fertiliser,
439 triple superphosphate, using pot experiments with isotope ^{32}P -labelled soil, suggesting
440 negligible difference in plant phosphorus nutrition and growth (Achat et al. 2014). However,
441 despite the abundance of phosphorus availability for plant growth, the recovered nutrient
442 fertiliser showed poor nitrogen uptake for plant growth (Ganrot et al. 2007, Matassa et al.
443 2015). In addition, phosphorus that was recovered in the form of amorphous calcium
444 phosphate precipitate, exhibited less water solubility, thereby hindering the crop uptake
445 (Plaza et al. 2007). As a result, wastewater nutrient recovery is a multi-dimensional challenge,
446 with considerable requirements to find a suitable market to distribute recovered nutrient
447 product with proven agronomic efficacy.

448 5. Conclusion

449 Three emerging membrane processes – FO, MD and ED – can advance wastewater
450 nutrient recovery with their unique mass transfer properties. FO, demonstrating low fouling
451 propensity and supplementing magnesium ion via reverse salt flux, is able to maximise
452 nutrient enrichment prior to struvite precipitation. MD, driven by vapour pressure difference,
453 is not only capable of achieving a high concentration factor, but also can recover volatile

454 ammonia as a high quality fertiliser. ED can selectively partition phosphate with an anion-
455 selective membrane, or produce phosphoric acid or ammonia with a bipolar membrane that
456 splits water solvent into proton and hydroxide. In addition, integration of these membrane
457 processes with existing nutrient precipitation processes could substantially improve nutrient
458 recovery efficiency, and diversify the nutrient product that can be extracted, even achieving a
459 near complete wastewater nutrient recovery. For the future, detailed techno-economic
460 analysis of these hybridised membrane-based processes in wastewater nutrient recovery
461 should be performed, such as process energy demand, CO₂ footprint, system robustness,
462 operating costs, product quality and market demands.

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467 **7. References**

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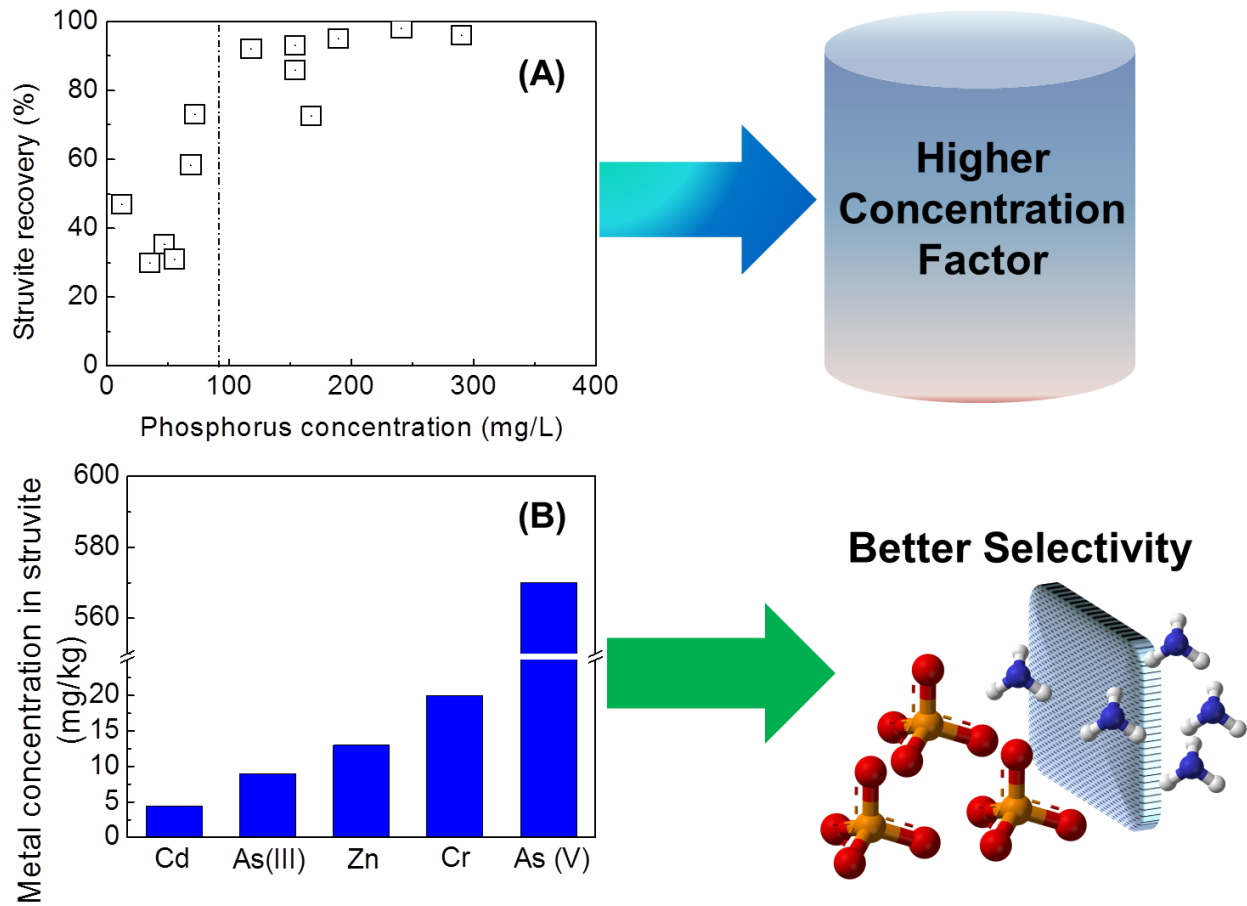
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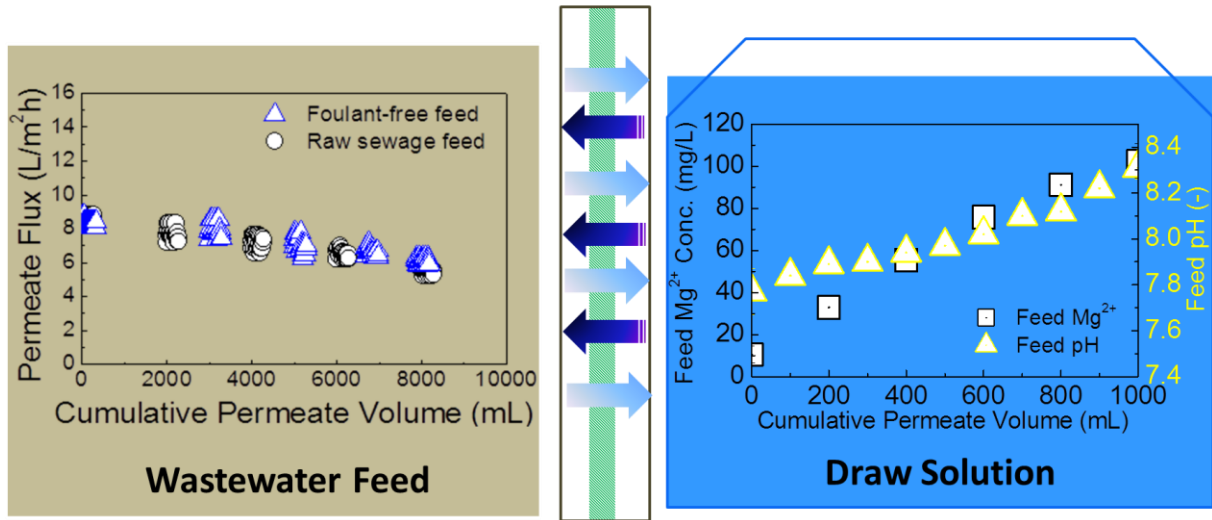


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880 **Figure 1:** Illustrations of the critical challenges in wastewater nutrient recovery. (A) Struvite
 881 precipitation efficiency as a function of initial phosphate concentration; there is a critical
 882 need for membrane processes enabling higher concentration factor. Data points were
 883 summarised from literatures (Çelen et al. 2007, Guadie et al. 2014, Jaffer et al. 2002, Liu et al.
 884 2011, Münch and Barr 2001, Pastor et al. 2008, Pastor et al. 2010, Ronteltap et al. 2010, Song
 885 et al. 2011) (B) Presence of toxic heavy metal ions in struvite precipitates from waste streams;
 886 there is a critical need for membrane processes with high selectivity. Data points were
 887 collected from literatures (Lin et al. 2013, Ma and Rouff 2012, Pizzol et al. 2014, Rouff 2012,
 888 Rouff and Juarez 2014).

Forward Osmosis



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891 **Figure 2:** Unique mass transfer properties of forward osmosis (FO) enhance nutrient
892 recovery efficiency from wastewater. Data reproduced from (Xie et al. 2014a, Xie et al.
893 2014b).

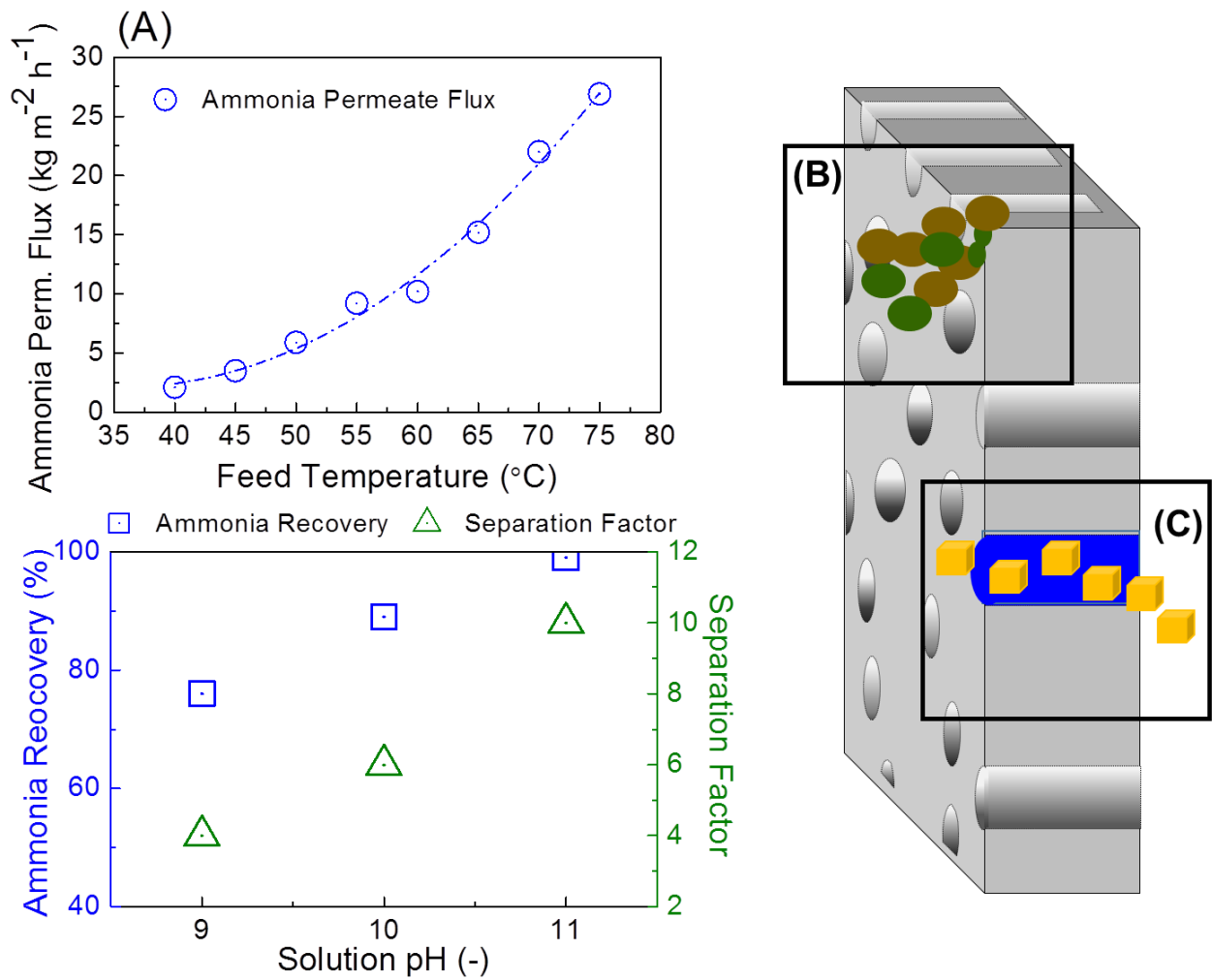


Figure 3: Conceptual illustration of membrane distillation (MD) for wastewater nutrient recovery. (A) Ammonia vapour selectively permeates through the membrane pores as a function of feed temperature (upper panel) and solution pH (lower panel where separation factor was calculated as the ratio of ammonia concentration in the feed and permeate); experimental data were reproduced from references (Ding et al. 2006, El-Bourawi et al. 2007, Qu et al. 2013, Xie et al. 2009). (B) Fouling of MD membrane leading to detrimental effect on process productivity (such as flux decline). (C) Wetting of MD membrane pores and permeate quality, such as feed solute (yellow cubes) flowing directly across membrane.

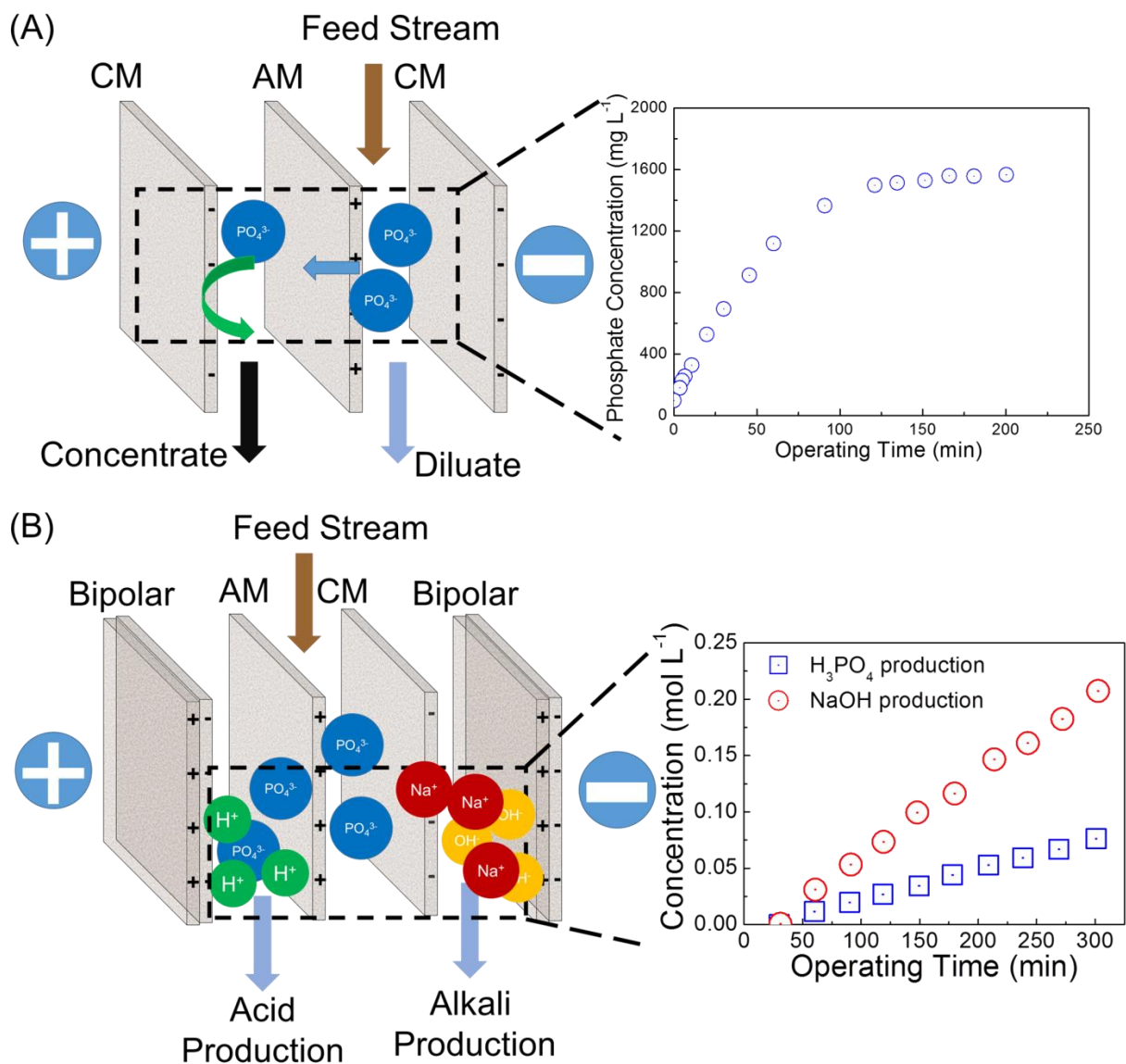


Figure 4: Conceptual illustration of electro dialysis (ED) for wastewater nutrient recovery. (A) Conventional ED process selectively concentrates phosphate in waste stream, where phosphate ion concentration in the concentrate stream increased as a function of time. (B) ED process with bipolar membrane selectively produces phosphoric acid from waste stream, where phosphoric acid concentration increases as a function of operating time. Data reproduced from Wang et al. 2013.

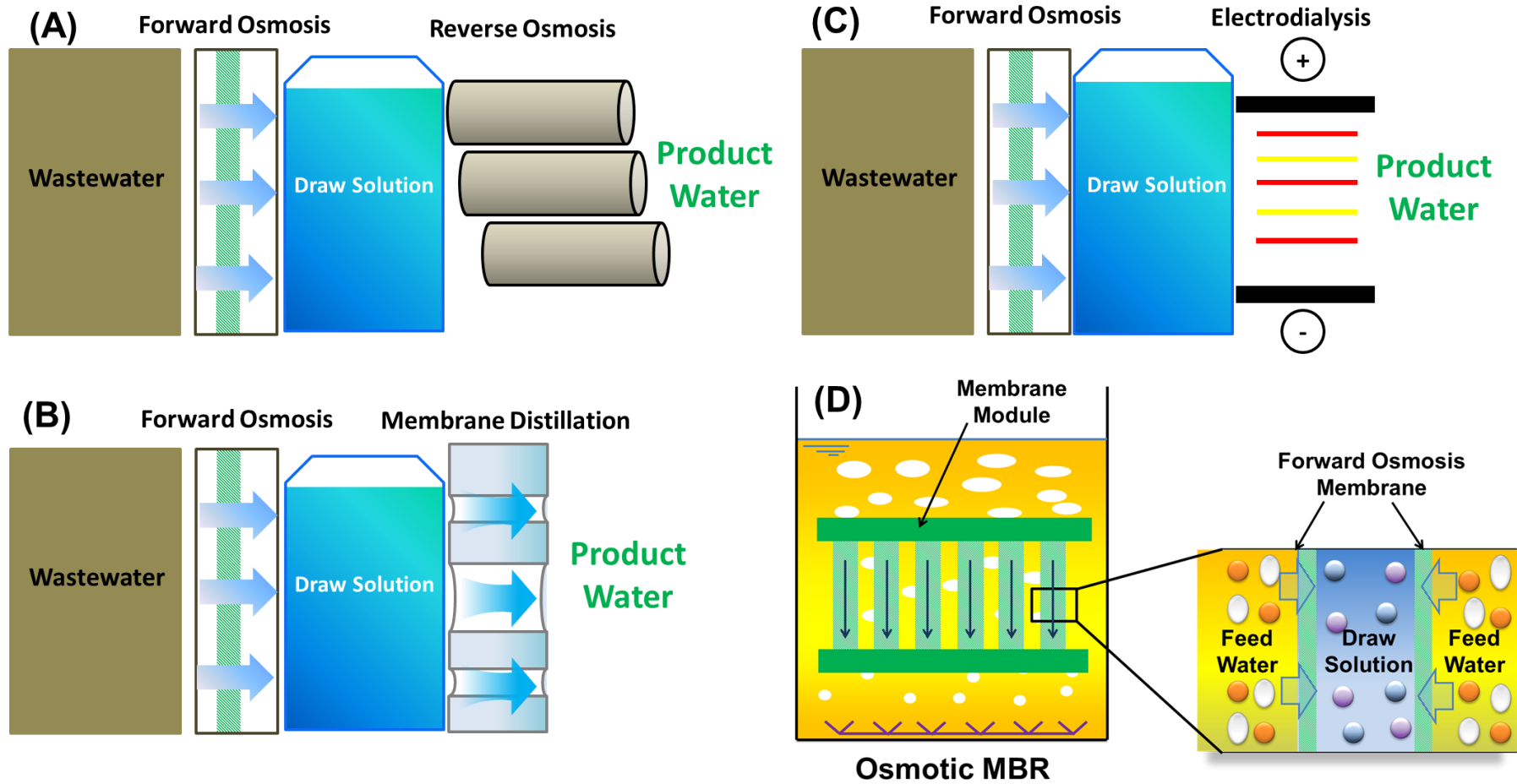


Figure 5: Conceptual illustrations of forward osmosis (FO)-based membrane processes for wastewater nutrient recovery: (A) FO-RO hybrid process; (B) FO-MD hybrid process; (C) FO-ED hybrid process; and (D) Osmotic MBR with FO membrane.

Table 1: Summary of nutrient and water recovery by pressure- (nanofiltration and reverse osmosis), osmotically- (forward osmosis), thermally- (membrane distillation) and electrically- (electrodialysis) driven membrane processes in terms of process performance, membrane fouling, and operating cost and energy consumption.

Driving force	Source water	Nutrient recovered	Key membrane process	Performance	Membrane fouling	Operating cost and energy consumption
Pressure-driven	Urine (Maurer et al. 2006)	Ammonium, phosphate, potassium	RO	Concentration factor up to 5. Ammonium: 70%; phosphate: 73%; potassium: 71%.	Membrane scaling	Operating pressure of 50 bar
	Synthetic urine (Pronk et al. 2006b)	Urea, ammonium, phosphate	NF	Urea: 10% ammonium: 55% phosphate: 94%	N.A.*	Operating pressure of 20 bar
	Synthetic wastewater (Niewersch et al. 2014)	Phosphate, potassium	NF	Phosphoric acid: 50% potassium: 30%	N.A.	Operating pressure of 12 bar
	Digested sludge (Blöcher et al. 2012)	Phosphate	NF	phosphate: 50%	N.A.	Operating pressure of 25 bar Operating cost can be covered by phosphate recovery revenue
Osmotically-driven	Urine (Zhang et al. 2014a)	Ammonium, phosphate, potassium	FO	Ammonium: 50-80% phosphate: >90% potassium: >90%	N.A.	N.A.
	Urine (Gormly and Flynn 2007, Michael et al. 2012)	Water	FO (X-Pack™, Water Well®)	Total nitrogen >95% urea > 93% total organic carbon > 95%	N.A.	N.A.
	Synthetic	Water	FO with	Diluted fertilizer for	N.A.	N.A.

	wastewater (Phuntsho et al. 2012)		fertilizer draw solution	agricultural irrigation		
	Secondary treated effluent (Hancock et al. 2013)	Water	FO-RO	Nitrate >72% phosphate >99% dissolved organic carbon > 98%	Cake layer formation	N.A.
	Raw sewage (Xie et al. 2013)	Water	FO-MD	Total organic carbon > 99% total nitrogen >99%	Cake layer formation	Draw solution temperature 40 °C
	Secondary treated effluent (Zhang et al. 2013b)	Water	FO-ED	Total organic carbon >90% Near 100% rejection of heavy metal ions (Cd, As, Pb)	N.A.	€3.32-4.92 per m ³ product water
	Activated sludge (Nguyen et al. 2013)	Water, ammonium, phosphate	FO	Ammonium >96%, phosphate >98% dissolved organic carbon > 99%	Cake formation	N.A.
	Activated sludge (Hau et al. 2014)	Water, ammonium, phosphate	FO-NF	Ammonium >97% phosphate >99%	Cake formation	NF operating pressure: 80 psi
	Activated sludge (Holloway et al. 2007)	Water, ammonium, phosphate	FO-RO	Ammonium >92.1% phosphate >99.8%	Pore blocking and surface fouling	4 kWh/m ³ at 75% water recovery
	Anaerobic sludge (Xie et al. 2014a)	Water, phosphate	FO-MD	Ammonium >90% phosphate >97% struvite product	Cake formation	Draw solution temperature 40 °C
Thermally-driven	Urine(Zhao et al. 2013)	Water, ammonia, organic matters	Vacuum MD	Organic matter: >99% Ammonia: 41-75% Water: 32-49%	Organic fouling with salt crystallization	Feed temperature: 50-70 °C Vacuum pressure: 9.5kPa.

	Synthetic wastewater (Xie et al. 2009)	Ammonia	Sweep gas MD	Ammonia: >96%	N.A.	Feed temperature: 65°C sweep gas flowrate: 3 L/min
	Synthetic wastewater (El-Bourawi et al. 2007)	Ammonia	Vacuum MD	Ammonia: >90%	N.A.	Feed temperature: 50 °C Vacuum pressure: 6.3kPa.
	Synthetic wastewater (Ahn et al. 2011)	Ammonia	Direct contact MD	Ammonia: >92%	N.A.	Feed temperature: 35°C Ammonia stripping solution: 1 M H ₂ SO ₄
	Synthetic wastewater (Qu et al. 2013)	Ammonia	Direct contact MD	Ammonia: >99%	N.A.	Feed temperature: 55°C Ammonia stripping solution: 0.1 M H ₂ SO ₄
	Swine manure (Zarebska et al. 2014)	Ammonia	Direct contact MD	Ammonia: >99%	Organic fouling followed by pore wetting	Feed temperature: 40°C Ammonia stripping solution: 0.5 M H ₂ SO ₄
	Swine manure (Thygesen et al. 2014)	Ammonia	Direct contact MD	Ammonia: >98%	Organic fouling	Feed temperature: 35°C Ammonia stripping solution: 0.5 M H ₂ SO ₄
Electrically-driven	Urine (Pronk et al. 2006a)	Ammonium, phosphate, potassium	ED with ion exchange membrane	Concentration factors: ammonia (2.9), potassium (3.1), phosphate (2.7); Eliminating micropollutants	N.A.	Applied current density: 22.5 mA/cm ² Current efficiency: 50%
	Municipal Wastewater (Zhang et al. 2013a)	Phosphate	ED with ion exchange membrane	Concentration factors: phosphate (6.5)	N.A.	Applied current density: 31.25 A/cm ² Current efficiency: 72% Energy consumption: 16.7 kWh/(kg PO ₄ ³⁻)

	Synthetic wastewater (Wang et al. 2013)	Phosphate	ED with ion exchange membrane	Concentration factors: phosphate (4.2)	N.A.	Applied current density: 71.5 mA/cm ²
	Synthetic wastewater (Wang et al. 2013)	Phosphate	ED with bipolar membrane	Concentration factors: phosphate (16); product phosphorus acid of 0.075 mol/L	N.A.	Applied current density: 50 mA/cm ² Current efficiency: 75% Energy consumption: 29.3 kWh/(kg H ₃ PO ₄)
	Swine manure (Mondor et al. 2008, Mondor et al. 2009)	Ammonium	ED with ion exchange membrane	Concentration factors: ammonium (5.3)	Calcium and colloidal particle deposition	Applied current density: 2.7 A/cm ² Current efficiency: 77.9%

* not applicable