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# **Techno-economic feasibility of recovering phosphorous, nitrogen and water from diluted human urine via forward osmosis**

Federico Volpin<sup>1</sup>, Huijin Heo<sup>2</sup>, Md Abu Hasan Johir<sup>1</sup>, Jaeweon Cho<sup>2</sup>, Sherub Phuntsho<sup>1,\*\*</sup> and Ho Kyong Shon<sup>1\*</sup>

<sup>1</sup> School of Civil and Environmental Engineering, University of Technology, Sydney (UTS), City Campus, Broadway, NSW 2007, Australia.

<sup>2</sup> School of Urban and Environmental Engineering, Ulsan Institute of Science and Technology (UNIST), UNIST-gil 50, Ulsan 689-798, Korea.

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\* Corresponding author: Tel.: (+61) 02 9514 2629; email: Hokyong.Shon-1@uts.edu.au

\*\* Corresponding author: Tel.: (+61) 04 2264 5628; email: Sherub.Phuntsho@uts.edu.au

22

## 23 **Abstract**

24

25 Due to high phosphorus (P) and nitrogen (N) content, human urine has often proven to suitable raw  
26 material for fertiliser production. However, most of the urine diverting toilets or male urinals dilute  
27 the urine 2 to 10 times. This decreases the efficiency in the precipitation of P and stripping of N. In this  
28 work, a commercial fertiliser blend was used as forward osmosis (FO) draw solution (DS) to  
29 concentrate real diluted urine. During the concentration, the urea in the urine is recovered as it  
30 diffuses to the fertiliser. Additionally, the combination of concentrate  $\text{PO}_4^{3-}$ , reverse  $\text{Mg}^{2+}$  flux from  
31 the DS and the  $\text{Mg}^{2+}$  presents in the flushing water, was able to recover the  $\text{PO}_4^{3-}$  as struvite. With 50%  
32 concentrated urine, 93% P recovery was achieved without the addition of an external  $\text{Mg}^{2+}$ .  
33 Concurrently, 50% of the N was recovered in the diluted fertiliser DS. An economic analysis was  
34 performed to understand the feasibility of this process. It was found that the revenue from the  
35 produced fertilisers could potentially offset the operational and capital costs of the system.  
36 Additionally, if the reduction in the downstream nutrients load is accounted for, the total revenue of  
37 the process would be over 5.3 times of the associated costs.

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40 **Keywords:** Urine treatment; forward osmosis; phosphorous recovery; Nitrogen recovery; struvite.

## 1. Introduction

The latest report from the United Nations Food and Agriculture Organisation (FAO) estimates that between 2015 - 2018, the world demand for fertilisers (N,  $P_2O_5$ ,  $K_2O$ ) has increased, on an average, 1.8 percent per annum (FAO 2017). As in 2015, the demand was estimated to be 184'017'000 tonnes, every year the fertiliser consumption increases of about 3'351'300 tonnes. With a world population expected to reach 11.2 billion people by 2100, this trend is expected to keep rising (United Nations 2017). The rise in the need for fertilisers would affect both the raw material availability as well as the greenhouse gas (GHG) emissions and eutrophication. In fact, at present, 1 - 2% of the world primary energy consumption is due to the production of ammonia fertiliser (130 million tonnes in 2008) (Bicer et al. 2017, Canfield et al. 2010). As a by-product, about 2 to 3 tons of  $CO_2$  are released for each ton of ammonia produced. Thereby the ammonia production industries are subsidising for about 0.93% of the world greenhouse gasses emission (Bicer et al. 2017). At the same time, the high quality and high-grade phosphate rock reserves, which constitute the primary source of phosphorous (P) fertilisers, are depleting. However, the projections of P depletion rates vary, with a recent estimate that suggests a 20 - 60% resource depletion by 2100 (Elser and Bennett 2011, Mogollón et al. 2018, Van Vuuren et al. 2010).

The current growth in the size and density of cities is leading to increasing nutrients discharge to the downstream treatment plants. The conventional biological oxidation and reduction of ammonium ( $NH_4^+$ ) to unreactive nitrogen gas ( $N_2$ ) is an energy-intensive process due to the high amount of oxygen required for the ammonia and nitrite oxidising bacteria (AOB and NOB) (Kavvada et al. 2017, Ma et al. 2016). Recent investigations determined a theoretical energy requirement of 1.2 – 12.5 kWh.Kg $_N^{-1}$  (Batstone et al. 2015, Maurer et al. 2003a, Wett et al. 2013) (lowest Sharon-Anammox, highest nitrification-denitrification) for the removal of N, and a unit cost of 93 – 134 \$.Kg $_P^{-1}$  for the removal of P (via Integrated Fixed-Film Activated Sludge Systems with Enhanced Biological Phosphorus removal

IFAS-EBPR) (Bashar et al. 2018). Consequently, two challenges are likely to become increasingly relevant to achieve sustainable waste management. First, finding new nutrient sources and, second, reducing the costs associated with WW treatment.

One of the proposed approaches to pursue these goals it is to “shortcut” the N and P cycles by recovering  $\text{NH}_3$  and  $\text{PO}_4^{3-}$  directly from the WW (Larsen et al. 2016, Maurer et al. 2006),(Larsen and Gujer 1997). In particular, if the different WW streams (i.e., faeces, urine, light and heavy greywater) are separated at source, the economics of nutrients and water recovering might be more favourable (Larsen et al. 2016). Among them, source separated human urine could be a suitable raw material for fertiliser production due to its high nutrients concentration (i.e. more than  $5 \text{ Kg}_\text{N}/\text{m}^3$  and up to  $0.5 \text{ Kg}_\text{P}/\text{m}^3$ ) (Fumasoli et al. 2016, Maurer et al. 2006, Maurer et al. 2003b). The phosphorous in the urine can be recovered individually via struvite crystallisation (de Boer et al. 2018, Etter et al. 2011). In this process, external magnesium ( $\text{Mg}^{2+}$ ) source is added to hydrolysed urine ( $\text{pH} > 9$ ) to trigger the precipitation of a magnesium ammonium phosphate salt (i.e., struvite). Vegetables were grown using struvite produced from human urine, showing an insignificant human health risk (de Boer et al. 2018). Currently, the primary issue with this process is that, in most male urinals, urine is heavily diluted with flushing water, which decreased the P-recovery efficiency (Liu et al. 2014, Udert et al. 2003). Additionally, only about 3% of the N in the urine can be recovered via struvite precipitation (Maurer et al. 2006). Therefore, there is a need to find suitable technologies to concentrate urine for N and P recovery while the energy consumption is low.

Among different processes that can be used to concentrate urine to recover N and P forward osmosis (FO) is one of them. FO is a process that relies on the osmotic pressure difference between two solutions, separated by a semi-permeable membrane, to drive the water transport. As the solutes are not able to diffuse through the membrane, the water in the low concentration solution (FS) will dilute the high concentration one (DS) until the thermodynamic equilibrium is reached (Phuntsho et al. 2012). As water transport is occurring by diffusion rather than advection, FO membranes were found

to be more fouling resilient compared to RO membranes (Siddiqui et al. 2018). Additionally, when fertiliser is used as a DS, which is called fertiliser-drawn FO (FDFO), the final diluted solution can be directly used for irrigation. This means that this process would possibly be low energy intensive as no high-pressure pumping is required (Van Der Bruggen and Luis 2015). The major drawback of this process is the occurrence of reverse diffusion of the ions in the DS to the FS, also called Reverse Salt Flux (RSF). This happens due to the non-ideality of the available FO membranes (i.e., the ionic rejection is not 100%).

The idea of taking advantage of the RSF phenomena was firstly investigated in our previous study (Volpin et al. 2018), for simultaneous urine concentration, urea/ $\text{NH}_3$  and  $\text{PO}_4^{3-}$  recovery. In this study, concentrated  $\text{Mg}(\text{NO}_3)_2$  fertiliser is used to dewater non-hydrolysed human urine. The urea and  $\text{NH}_3$  in the urine can diffuse across the membrane, to the fertiliser DS, due to the combination of poor rejection and high concentration gradient. At the same time, the combination of urine concentration and reverse  $\text{Mg}^{2+}$  flux from the fertiliser to the urine can enhance the  $\text{PO}_4^{3-}$  recovery as struvite.

In this work, a commercial fertiliser blend was used to extract water and nitrogen from real urine. Also, the combination of urine concentration and the  $\text{Mg}^{2+}$ -RSF from the fertiliser allowed almost full phosphorous recovery. Water and nitrogen flux, urine concentration factor and membrane fouling/scaling were also investigated. Finally, the data obtained were used to calculate the payback period of the plant. Operational and capital costs (OPEX and CAPEX) were benchmarked with the revenue from the produced fertiliser. The savings due to the reduction in the downstream nutrients load was also considered.

## **2. Materials and methods**

### **2.1 Process description**

Figure 1 shows the schematic representation of the process. Further details on the set-up used for the experiments can be found in our previous studies (Chekli et al. 2017, Phuntsho et al. 2012). Real fresh (i.e., non-hydrolysed) urine was used as feed solution (FS) while pure and blended fertilisers were used as DS. The solutions were separated by a commercial thin-film composite (TFC) polyamide (PA) membrane (Toray Chemical Korea Inc., South Korea), with an active membrane area of 20.02 cm<sup>2</sup>. Membranes were stored in distilled water at 4 °C before use and were oriented to active layer-feed solution (i.e., AL-FS, FO mode) during the tests with the feed solution being the real urine. The osmotic gradient between the two solutions will allow the permeation of water to the draw channel, thereby diluting the fertiliser. Urea would diffuse to the fertiliser side due to the combination of the concentration gradient, low urea molecular weight and absence of charge. Phosphorous, on the other hand, is crystallised after the FO. The increase in the PO<sub>4</sub><sup>3-</sup> concentration plus the Mg<sup>2+</sup> reverse flux from the DS allowed for the precipitation of phosphorous as struvite. This would happen once the pH of the concentrated urine would rise to 8.5 – 9 as a consequence of the hydrolysis of the remaining urea (M. Fidaleo and R. Lavecchia 2003, Randall et al. 2016, Ray et al. 2018).

### **2.2 Urine collection, fertiliser preparation, storage and characterisation**

Real urine was collected and immediately frozen at -40 °C to avoid bacterial growth (Bernini et al. 2011). After the collection, the urine was thawed at 4 °C, mixed to homogenise the solution, separated in 500 mL batches, and re-frozen. No pre-filtration of the urine samples was performed. The urine composition is given in Table 1.

Single fertiliser and a commercial fertiliser blend were used as DS. Firstly, magnesium nitrate hexahydrate ( $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ) employed. A magnesium nitrate solution was chosen as a control solution due to the (I) high solubility of  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , which generates high osmotic pressure, (II) high  $\text{Mg}^{2+}$  content and (III) high water flux (Volpin et al. 2018). Afterwards, a commercial fertiliser blend was used to simulate real applications such as fertigation of the green wall or other hydroponic applications (Xie et al. 2015),(Zou and He 2016),(Chekli et al. 2017). The commercial hydroponic nutrient solution (Optimum Grow - twin pack hydroponic nutrient) used in this study as DS was obtained from Fernland Agencies Pty Ltd (Queensland, Australia). This is a hydroponic nutrient solution usually employed in plant nurseries and commercial greenhouses. The composition of the fertiliser solution is displayed in Table 1. This nutrient solution consists of part A and B, to be diluted separately and then mixed. Part B was chosen for these experiments as it contains  $\text{Mg}^{2+}$ .

The following analytical equipment was used for the measurement of the fertiliser and urine composition. The anions concentration were measured using Ion Chromatography (IC, Thermo Fisher Scientific, USA) and cell test kits using a spectrophotometer (Spectroquant NOVA 60; Merck, Germany). The cations were measured using Microwave Plasma Atomic Emission Spectrometry (MP-AES) (MP-AES 4100, Agilent, USA). Urea and ammonia were measured by UV-Spectrophotometer (UV-1700, Shimadzu, Japan), at 340 nm wavelength, using urea/ammonium Megazyme kit (Megazyme, Australia).



## Experimental plan and performance evaluation

Experiments were first conducted using 0.5 M  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  as DS. Afterwards, the commercial fertiliser blend was used to conduct long-term experiments (i.e. > 10 days). The concentration of the DS was chosen to achieve a water flux similar to the one generated by the commercial fertiliser.

### 2.3.1. Effect of pH on membrane scaling

The first set of experiments aimed at assessing the optimal pH of urine to avoid struvite precipitation on the membrane surface. In this regards, the pH of fresh urine was adjusted to 6, 6.5, 6.75, 7 and 7.5 using 1 M NaOH or 1 M citric acid. FO flux decline was measured over time, and the used membrane was analysed by scanning electron microscopy (SEM, Zeiss Supra 55VP, Carl Zeiss AG, Germany). Samples were firstly dried by purging of air and then lightly coated with Au/Pd. The SEM imaging was performed at an accelerating voltage of 10 kV at different magnifications and various points.

### 2.3.2. Experimental protocol and performance evaluation

Once the optimal feed pH was selected, experiments using 2 L of real urine as FS and 0.5 L of 0.5 M  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  as DS were performed. Water flux, reverse magnesium flux ( $\text{Mg}^{2+}$  - RSF), urea/ $\text{NH}_3$  forward flux and N, P recovery rates were measured. Mass balance calculations were done to estimate the nitrogen recovery. The specific nitrogen flux was calculated as the ratio between  $J_N$  [ $\text{g}_N/\text{m}^2 \cdot \text{h}^{-1}$ ] and  $J_w$  [ $\text{L}/\text{m}^2 \cdot \text{h}^{-1}$ ]. Magnesium reverse salt flux was also measured to estimate the amount of  $\text{Mg}^{2+}$  that has migrated to the urine. This is necessary to estimate the maximum phosphorous recovery. The experiments were conducted in triplicates and at three different urine dilution factors, i.e., 1:1, 1:2 and 1:5. Dilutions were performed to mimic the different designs of urinals and urine-diverting toilets (e.g., Dubbletten, Wostman-DS) (Zhang et al. 2014).

The experiments were run upon reaching a urine concentration equal to the one of undiluted urine (i.e., 80% and 50% concentration for 1:5 and 1:2 urine dilution respectively). Undiluted urine was still concentrated 50% as a means of comparison. At the end of the filtration test, urea and  $\text{NH}_3$

concentrations were measured both in the concentrated urine and in diluted DS ( $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ). Afterwards, 10  $\mu\text{L}$  of urease enzyme solution was spiked in the concentrated urine to speed up the urea hydrolysis to raise the pH up to 9 – 9.5 and trigger struvite precipitation. The hydrolysed urine was then stirred for 2 hours and filtered using a 0.45  $\mu\text{m}$  pore-size filter (Merck, Millipore).  $\text{PO}_4^{3-}$  was measured in the filtered solution to calculate the P-recovery.

### 2.3.3. Long-term experiments with commercial fertiliser blend

After preliminary tests, a long-term experiment (i.e. two weeks) was carried out with 5 L of diluted urine (1:5) as FS and 0.35 L of commercial fertiliser as DS. Tap water was used for the dilutions, and its composition is presented in Table 1. The experiment was run until  $\sim 50\%$  urine concentration and about eight times fertiliser dilution was reached. To avoid microorganisms growth in the FS and DS, 0.5 mL of hydrogen peroxide (30% v/v) was applied once a week. The outcomes from this test were used for the economic analysis below discussed.

To better identify the impact of fouling of the transmembrane flux, the theoretical FO water flux was modelled via the classical solution-diffusion model coupled with the diffusion-convection transport in the membrane support layer (Ali et al. 2018, Tang et al. 2010). Equation 1 was used for the flux calculation.

$$J_w = \sigma K_m \ln \left( \frac{A\pi_D + B_s}{A\pi_F + J_w + B_s} \right) \quad (\text{AL-FS}) \quad (1)$$

Where  $\sigma$  is the reflection coefficient, assumed as unity (complete rejection of the solute),  $K_m$  is the mass transfer coefficient, of the selected DS, given by the ratio between the diffusivity of the salt and the structural parameter (S) of the membrane (i.e.  $K_m = D/S$ ). The transport coefficients for water and solutes are expressed as A and B<sub>s</sub>. Finally,  $\pi_D$  and  $\pi_F$  are the DS and FS bulk osmotic pressure, respectively. The input and derivation of eq. 1 is described in Section 1.3 of the SI.

### 2.3.4. Membrane fouling and scaling

After each experiment, the membrane was cleaned with a solution of 0.1 M NaOH (pH = 12) for 10 minutes and then flushed with de-ionised water (Kim et al. 2014, Mi and Elimelech 2008). Alkaline cleaning procedure was chosen as most of the fouling layer was resulting from organic deposition (e.g., creatinine). After cleaning, membrane flux recovery test was conducted using 0.5 M NaCl solution and de-ionised water, as DS and FS respectively.

### 2.3 Economic analysis

Understanding the economic feasibility of this process is crucial for practical application. The cost of the system as operational and capital costs (OPEX and CAPEX) was analysed and benchmarked with the revenues (i.e., urea/NH<sub>3</sub> and struvite). Additionally, the theoretical savings of reducing the nutrients load to the downstream treatment plant were included in the analysis. Finally, an economic analysis was performed to investigate the investment return period with different process performances. Starting with the costs, CAPEX was calculated assuming a plant treating 10 m<sup>3</sup>/day of urine with (I) plant availability of 0.95, (II) 6% interest rate, (III) pump efficiency of 0.85. Construction and equipment costs were also included (Kim et al. 2017b). The construction cost includes pressure vessels, pumps, piping and others (i.e., civil engineering, intakes, working capital and contingencies) while the equipment cost includes membranes and materials (Chekli et al. 2017, Valladares Linares et al. 2016). The required membrane area was calculated based on the plant capacity and average measured flux. CAPEX amortization costs were then calculated as follows:

$$\text{Amortized CAPEX } (\$. \text{m}^{-3}) = \frac{\text{CAPEX} \times i}{1 - (1 - i)^{-n}} \times \frac{1}{Q \times 365} \quad (2)$$

Where *i* is the interest rate, *n* is the plant lifetime, and *Q* is the plant capacity.

Secondly, the OPEX was calculated based on the membrane replacement rate (i.e., 10%), FO membrane cost (i.e., 1000 \$/element), electrical cost, chemical cleaning and others such as repairs,

laboratory fees, labour and insurance (Valladares Linares et al. 2016). For the pumping energy calculations, pressure losses of 2 bar, for the feed channel, and of 0.5 bar for the draw channel were assumed (Kim et al. 2017b). All the detailed information on the input of the cost analysis are summarised in the supporting information (SI) (Table S1). The revenue from the recovery of urea/ $\text{NH}_3$  and struvite was calculated using the recovery rates measured in this work and the commercial prices of urea and struvite (World Bank Group 2018),(Forrest et al. 2008). All the detailed information on the input of the analysis of the revenues are summarised in the SI (Table S2). The theoretical savings in the removal of N and P were also calculated according to Maurer et al. (2003b). Current Australian energy prices, 2.4 AUD/ $\text{Kg}_\text{N}$  and 2.26 AUD/ $\text{Kg}_\text{P}$  were assumed for the calculations (AEMC 2013, Maurer et al. 2003a). Finally, for the economic analysis, the plant construction time was hypothesised to be one year with the construction costs distributed during the construction year.

### **3. Results and discussion**

#### **3.1 Effect of pH of urine on struvite production and FO performance**

The solubility of struvite crystals is strongly dependant on the pH of the solution. Struvite solubility decreases at  $\text{pH} > 6$ , to reach the minimum solubility at pH above 9 (Doyle and Parsons 2002). As most commercial fertilisers contain  $\text{Mg}^{2+}$ , some of which might back diffuse during the FO filtration process, it is important to understand what the optimal urine pH is to avoid membrane scaling caused by struvite formation on the membrane active layer. In this regards, 0.5 M  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  was used to dewater real urine with a pH of 6, 6.5, 6.75, 7 and 7.5. Real urine was used instead of synthetic urine as the presence of organic acids affects the thermodynamics of struvite crystals formation (Song et al. 2014). Each test was run for 20 hours to have a reliable indication of the flux decline.

The results presented in Figure 2, show that pH above 6.75 can cause severe membrane scaling resulting in higher flux decline. This is due to the formation of struvite crystals on the membrane surface (SEM picture inserted in Figure 2). Kim et al. (2017a) showed that, the precipitation of solid struvite decreases from pH lower than 7.5 to reach a minimum at pH of 6.5 (Kim et al. 2017a).

To achieve 20% concentrated urine at pH 7.5, the flux decline was almost 100% whereas it was ~30% when the urine pH was  $\leq 6.75$ . Based on these results, subsequent experiments were conducted with urine having a pH of 6 - 6.5.

#### **3.2 Effect of urine dilution on the water flux and membrane cleaning**

The concentration of  $\text{PO}_4^{3-}$  and  $\text{Mg}^{2+}$  in the urine greatly affects the P recovery efficiency via struvite crystallisation. Previous studies found that, diluted urine often shows less P-recovery as struvite as well as lower struvite crystals size under the same experimental conditions (i.e. at same pH and  $\text{Mg}^{2+}$  load) (Liu et al. 2014, Udert et al. 2003). In this work, it was also found that P removal was less than 73% when the urine was diluted five times, compared to a 99.5% removal of P with the undiluted urine (Figure S1, see section 1.3 in the SI for further details).

In this section, the effectiveness of FO in concentrating different urine solutions was investigated. Water flux and membrane cleaning effectiveness were evaluated for urine diluted to 1, 2 and 5 times. The following average fluxes were achieved: 6.6 L.m<sup>-2</sup>.h<sup>-1</sup> (undiluted, 50% concentration, 67.5 h), 8.6 L.m<sup>-2</sup>.h<sup>-1</sup> (2 times dilution, 50% concentration, 64.5 h), 8.1 L.m<sup>-2</sup>.h<sup>-1</sup> (5 times dilution, 80% concentration, 93 h). Figure 3(A) shows the temporal variation of water flux as a function of the urine concentration factor. Flux decline was attributed to the combination of DS dilution, urine concentration and membrane fouling. Particularly at high urine concentration factor (i.e., > 15 mS/cm), the formation of a brown layer was observed on top of the membrane surface (Figure S5). Membrane cleaning with de-ionised water only partially removed the brown layer, however, the brown layer was removed entirely when alkaline cleaning with 0.1 M NaOH was conducted for 10 minutes. Figure 3(B) confirms the effectiveness of the cleaning strategy by showing that the flux of the clean membrane was nearly the same to that of the virgin membrane with a minor variation of  $\pm 5\%$ . This was despite the different experimental time and urine concentration factor. It can be then concluded that real urine can be concentrated over 2 times without any pre-treatment. Alkaline cleaning was found to be effective in fully restoring the initial water flux.

### 3.3 Nutrients flux and recovery for different urine dilution factors

Nitrogen and phosphorous recovery is key parameters for understanding the economic feasibility of the process. Hence, the nitrogen flux from the urine to the fertiliser solution together with the P recovery after struvite precipitation was investigated.

The urine dilution factor strongly influences the urea/NH<sub>3</sub> flux (Figure 3A). This can be explained by Fick's law, as it regulates the membrane diffusion rate (i.e.,  $J_N = D_N \cdot A \cdot \Delta C_N / x$ ) where the concentration difference  $\Delta C_N$  is a dominant factor in the diffusion rate  $J_N$ . In this case, the nitrogen diffusivity  $D_N$ , membrane area  $A$  [m<sup>2</sup>] and thickness  $x$  [m] is the same for each experiment. Therefore, the high initial concentration of urea/NH<sub>3</sub> in undiluted urine is responsible for the higher diffusion rate. Additionally, the higher water flux ( $J_w$ ) (in the case of diluted urine) also lowers the specific nitrogen flux. Figure S2

shows the linear relationship between the urine concentration factor and the total nitrogen recovery (as the sum of urea and  $\text{NH}_3$ ).

Because of the same principle, the  $\text{Mg}^{2+}$  reverse flux was almost the same in all cases. In fact  $\Delta C_{\text{Mg}}$  depends only on the  $\text{Mg}^{2+}$  concentration in the DS, which was the same in each test.

The experiments showed that the recovery of P and N was ~99% and 40-65%, respectively (Figure 4 B). The total N recovery was found to be 1-2% lower than the sum of urea-N and  $\text{NH}_3$ -N. This is because of the  $\text{NO}_3^-$  reverse salt flux from the  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  DS to FS (Volpin et al. 2018). Also, due to the  $\text{Mg}^{2+}$ -RSF and P concentration factor, the removal of phosphorous was completely high (>99%) in all the concentrated urine solutions without any addition of additional magnesium.

### **3.4 Diluted real urine with commercial fertiliser as the draw solution**

After having tested the effectiveness of a pure fertiliser, a commercial fertiliser blend was used to dewater real diluted urine. A long-term experiment (i.e., over 11 days) was performed to investigate:

- i. Achievable urine concentration and fertiliser dilution,
- ii. Clean in place effectiveness, and
- iii. Nitrogen and phosphorus recovery.

The results obtained in long-term test were used for the economic analysis described in the following section 3.6.

Figure 5 (A) shows the temporal variation of flux over the urine concentration factor. The average flux to achieve 50% urine concentration was about  $4.2 \text{ L.m}^{-2}.\text{h}^{-1}$ . The gradual flux declination is mainly attributed to the decrease in the osmotic driving force ( $\Delta\pi$ ), as over time the fertiliser gets diluted and the urine gets concentrated. However, membrane fouling could also play an essential role. To assess the effect of membrane fouling on the flux decline, flux modelling and clean in place (CIP) with 0.1 M NaOH were conducted. The impact of flux loss due to fouling can be noticed after 20% urine concentration, and it becomes more severe in the final part of the filtration. By looking at the flux

improvement after the CIP, at about 33% urine concentration, a 29% flux improvement was measured (i.e., from 3.3 to 4.6 L.m<sup>-2</sup>.h<sup>-1</sup>). This corresponds to an 88% flux recovery compared to the theoretical flux.

On the other hand, Figure 5 (B) shows the calculated N and P recovery based on nutrients mass balance. The results show that over 93% of the P was removed after struvite precipitation and over 50% of the N was recovered in the fertiliser. This is in line with the previous results using Mg(NO<sub>3</sub>)<sub>2</sub>.6H<sub>2</sub>O as DS (Figure 4).

To conclude, urine was concentrated 50% using a commercial fertiliser as DS, while fouling was observed. About 93% of the P and 50% of the N were removed after the filtration-precipitation without the addition of extra Mg<sup>2+</sup> source. Assisting the process with hydraulic pressure (pressure assisted osmosis or PAO), especially at the end of the filtration, could improve the overall average flux (Chekli et al. 2017). However, the impact that the hydraulic pressure would have on the N/P recovery and fouling should be further investigated.

### **3.5 Assessment of the system's economic feasibility**

In this section, the costs associated with the construction and operation (i.e., CAPEX and OPEX respectively) were benchmarked with the revenue from the produced fertiliser and the downstream WW treatment savings. The assumptions considered during cost estimation are shown in Tables S1 and S2 in the supporting information. The impact of the average water flux in the return in the capital investment was also analysed. Scenario A uses  $J_w = 4.2 \text{ L.m}^{-2}.\text{h}^{-1}$  as input (measured value), scenario C uses  $J_w = 8 \text{ L.m}^{-2}.\text{h}^{-1}$  (ideal no fouling process) and scenario B used  $J_w = 6.5 \text{ L.m}^{-2}.\text{h}^{-1}$  (moderate fouling, in between A and C).

Figure 6B and Figure S6 show that, without accounting for the reduction in the N and P discharge, the payback period for the Scenario A is of over 20 years. That is because, the revenue from the struvite and urea/NH<sub>3</sub> recovery is only 0.8% higher than the OPEX of the system (Figure 6A). This is because membrane replacement costs account for the most significant portion of the OPEX, i.e., 64% (Figure



S3). However, Figure 6B shows that just by increasing the average flux to 6.5 or 8 L.m<sup>-2</sup>.h<sup>-1</sup> (scenario B and C) the payback period can be moved to 5.5 or 2.7 years respectively. This could be achieved by having a more frequent CIP or by assisting the process with hydraulic pressure. Chekli et al. (2017) showed that applying a small pressure (e.g., 2 - 4 bar) at the end of the filtration, the flux can be increased by over 57% (Chekli et al. 2017). This should be investigated further as a mean to reduce the OPEX and CAPEX of the system.

Regarding the returns from the process, Figure S4 shows that struvite and urea/NH<sub>3</sub> would yield similar revenues. It can be argued that the urea recovered is already mixed with the fertiliser draw solution. Therefore, instead of revenue, its recovery would be more of saving in the addition of nitrogen to the fertiliser solution to meet the required concentration. To have a more conservative estimation, the savings from the water extracted from the urine were not included in the calculation.

Finally, the theoretical savings on the reduced nutrient load to the downstream treatment plant were calculated. The results are shown in Figure A and B. Due to the high nitrogen content of urine, and the relatively high cost to oxidise NH<sub>4</sub><sup>+</sup> to NO<sub>3</sub><sup>-</sup> and reduce NO<sub>3</sub><sup>-</sup> back to unreactive N<sub>2</sub>, a 50% reduction in the N load from urine was found to have a significant impact on the economics of the process. In particular, the savings in the downstream treatment costs can be as high as 6.2 \$/m<sup>3</sup> (which is 3.5 times higher than the sum of CAPEX and OPEX). Nonetheless, at present, this reduction in nutrients could not have a direct economic benefit to the plant operators.

To conclude, a preliminary economic analysis was performed to estimate the economic viability of this process. Even though with the current fertiliser prices, the direct revenues from the extracted fertiliser would only be adequate to offset the FDFO process costs, if the savings to the downstream treatment plant are included, the total revenue was calculated to be over 4.3 times of the process costs.

#### 4. Conclusions

Forward osmosis (FO) was successfully utilised in dewatering real human urine, diluted five times, using a commercial fertiliser blend as DS. Phosphorous recovery from the urine was achieved without the addition of an external  $Mg^{2+}$  source. Instead, the combination of over 50% urine concentration, reverse  $Mg^{2+}$  flux from the DS and the  $Mg^{2+}$  present in the tap water were found to be adequate to remove 93% of the P that presents in the urine. Simultaneously, about 50% of the N was recovered in the diluted fertiliser DS. The N recovery was found to be due to the poor rejection of the urea and ammonia in the urine. Urea and ammonia can, therefore, diffuse to the DS due to the concentration gradient across the membrane. Organic foulants in the urine were found to affect the FO water flux, especially at high urine concentration factors. However, alkaline cleaning with 0.1 M NaOH was able to recover the initial flux values. Inorganic scaling, on the other hand, was effectively avoided by keeping the urine pH below 6.5.

The experimental outcomes were used to perform an economic analysis of the process. Specifically, the OPEX and CAPEX of achieving 50% urine concentration, and over eight times fertiliser dilution, were compared with the revenue from the P and N recovery. Additionally, the theoretical savings from reducing the nutrients load to the downstream treatment plant were included. The low average filtration flux was found to yield high OPEX, mainly due to the membrane replacement cost. With the average flux measured in this study (i.e.,  $4.2 \text{ L.m}^{-2}.\text{h}^{-1}$ ), the payback period for this plant would be over 20 years. However, if the average flux is increased to  $8 \text{ L.m}^{-2}.\text{h}^{-1}$  (via fouling mitigation or pressure assisted osmosis), the return on the investment can be reached after 2.7 years. Finally, if the savings to the downstream treatment plant are included, even with average flux of  $4.2 \text{ L.m}^{-2}.\text{h}^{-1}$ , the total revenue of the process would be over 4.3 times of the associated costs.

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