Electronic Supplementary Material (ESI) for Environmental Science: Water Research & Technology. This journal is © The Royal Society of Chemistry 2019

## **Appendix A. Supplementary Material for**

# Reduction of Reverse Solute Flux Induced Solute Buildup in the

## **Feed Solution of Forward Osmosis**

Matthew Ferby <sup>a</sup>, Shiqiang Zou <sup>b</sup>, and Zhen He<sup>a,c\*</sup>

<sup>a</sup> Department of Civil and Environmental Engineering, Virginia Polytechnic Institute and State

University, Blacksburg, Virginia 24061, USA

<sup>b</sup> Department of Civil and Environmental Engineering, Stanford University, Stanford, CA 94305

USA

<sup>c</sup> Department of Energy, Environmental and Chemical Engineering, Washington University in St.

Louis, St. Louis, MO 63130, USA

Total pages: 4 excluding cover page

Total tables: 2

\*Corresponding author. Phone: (540) 231-1346; fax: (540) 231-7916; e-mail: zhenhe@wustl.edu

#### Solute Buildup and Concentrating Effect

Solute buildup (*SBU*) (g m<sup>-2</sup> h<sup>-1</sup>) is used to quantify the solute concentration due to accumulation cause by RSF and can be calculated using Eq. (S1)

$$SBU = \frac{\left(n_{f,F} - n_{i,F}\right) \times MW}{A \times \Delta t} \tag{S1}$$

where  $n_{i,F}$  and  $n_{f,F}$  represent the DS in the feed solution's initial and final mole, respectively, MW is the molecular weight of the solute, A is the effective surface area of the FO membrane, and  $\Delta t$  is the operation time.<sup>1</sup>

The concentrating effect (*CE*) quantitatively describes the effect of water recovery on the solute concentration, if already present, in the feed solution (Eq. (S2)):

$$CE = \frac{V_{recovered} \times C_{i,F}}{A \times \Delta t}$$
(S2)

Note that CE (g m<sup>-2</sup> h<sup>-1</sup>) utilizes the amount of recovered water -  $V_{recovered}$  (L) and the initial concentration of the feed solution -  $C_{i,F}$  (g L<sup>-1</sup>) to find the rate of concentration of the solute in the feed solution relative to the water recovered in the draw solution. This is different from the change of the absolute amount of solute in the feed solution after the membrane treatment duration. These two variables can be used to determine the movement of solute ( $J_s$ ) across the membrane by subtracting the *SBU* from the *CE*, unless DI water is used as the feed solution resulting in  $J_s = SBU.^2$ 

#### **Physical Separation Case Study**

In order to showcase the use of the two proposed parameters- solute removal/recovery rate (SRR) and removal/recovery ratio (ReR), a mass balance was completed on the osmotic membrane bioreactor (OMBR) of a previous study.<sup>3</sup> The data used to generate Figure 3A was both calculated and provided (Table S1).

Table S1. Mass balance variable used for physical separation case study

Mass Balance Variable*	Value
Average Water Flux	3.5 LMH
Average Reverse Salt Flux	0.03 g m <sup>-2</sup> h <sup>-1</sup>
Salinity Accumulation	0.58 g day <sup>-1</sup>
Mg <sup>2+</sup> Removal	4.06 mg L <sup>-1</sup>

\* Variables are either directly reported or calculated based on the data in the study

The salinity accumulation in the OMBR was determined as described by Eq. S3.

Accumulation  
= 
$$\frac{(TDS_f - TDS_i) * V}{\Delta t} = \frac{(EC_f * \alpha - EC_i * \alpha) * V}{\Delta t}$$

Where  $EC_f$  and  $EC_i$  represent the final and initial electrical conductivity (mS cm<sup>-1</sup>), respectively;  $\Delta t$  (days) represents the operation time;  $\alpha$  is the conversion factor between conductivity and TDS;  $TDS_f$  and  $TDS_i$  represent the final and initial TDS (g L<sup>-1</sup>) concentration; V is the bioreactor's feed solution volume. MF flux used in the SRR calculation was estimated using a study that investigated MF flux rates compared to FO flux.<sup>4</sup> The draw solution concentration was considered in this assumption as well.

#### **Chemical Precipitation Case Study**

For the chemical precipitation case study, the information was both calculated and provided to generate Figure 4A (Table S2). The mass balance was based off a previous FO study.<sup>5</sup>

Mass Balance Variable*	Value
Draw Solution Molarity (MgCl <sub>2</sub> )	0.5 M
Average Water Flux	2.32 L m <sup>-2</sup> h <sup>-1</sup>
Average Reverse Salt Flux	1.55 g m <sup>-2</sup> h <sup>-1</sup>
Struvite Precipitation	0.39 g m <sup>-2</sup> h <sup>-1</sup>

Table 2S. Mass balance variable used for chemical separation case study

\* Variables are either directly reported or calculated based on the data in the study

This case study presented many of the details necessary to compare to other DS precipitation studies. The RSF was converted to more widely used units  $(g m^{-2} h^{-1})$  to determine the other parameters. In the paper, the authors reported that ~25% of the DS precipitated as struvite. The struvite composition was determined using scanning electron microscopy-energy dispersive spectroscopy. In addition to struvite, it was reported that a portion of the Mg<sup>2+</sup> precipitated as Mg(OH)<sub>2</sub>. With struvite being the precipitate of interest, the SRR would be calculated using Eq. S4.

$$SRR = \frac{C_{Mg,struvite} * V_{feed}}{A * \Delta t}$$
(Eq. S4)

Where  $C_{Mg,struvite}$  (m) is the concentration of magnesium precipitated as struvite,  $V_{feed}$  is the volume of the concentrated feed solution. However, not enough information was provided to make this distinction and resulted in a slightly higher ReR. Based on information provided by the authors, the amount of reversed fluxed Mg<sup>2+</sup> that precipitated as struvite was assumed to be the ReR (0.25 or 25%). The assumed ReR did not include Mg<sup>2+</sup> that was naturally present in the digested swine wastewater (feed). Moreover, removal techniques rely heavily on a precipitate being formed over the chemical purity. This caused us to consider calculating the SRR for this case study by manipulating the ReR parameter (Eq. S5).

$$SRR = ReR * RSF$$
 (Eq. S5)

In the event that more information is provided, the ReR can be denoted if a particular precipitate is desired. This would suggest that future research could present ReR that determine how much of the reverse fluxed DS precipitated as one compound compared to another. For example, distinction between Mg(OH)<sub>2</sub> and struvite could help determine what operation conditions exist when comparing removal efficacies between different studies that use MgCl<sub>2</sub> as a DS. This same concept would apply to other DS and precipitates, as well.

### References

1. Zou S, He Z. Enhancing wastewater reuse by forward osmosis with self-diluted commercial fertilizers as draw solutes. Water Res. 2016;99:235-43.

2. Zou S, Qin M, He Z. Tackle reverse solute flux in forward osmosis towards sustainable water recovery: reduction and perspectives. Water Res. 2019;149:362-74.

3. Chang H-M, Chen S-S, Nguyen NC, Chang W-S, Sinha Ray S. Osmosis membrane bioreactor–microfiltration with magnesium-based draw solute for salinity reduction and phosphorus recovery. Int Biodeter Biodegr. 2017;124:169-75.

4. Wang J, Pathak N, Chekli L, Phuntsho S, Kim Y, Li D, et al. Performance of a novel fertilizer-drawn forward osmosis aerobic membrane bioreactor (FDFO-MBR): Mitigating salinity build-up by integrating microfiltration. Water. 2017;9(1).

5. Wu Z, Zou S, Zhang B, Wang L, He Z. Forward osmosis promoted in-situ formation of struvite with simultaneous water recovery from digested swine wastewater. Chemical Engineering Journal. 2018;342:274-80.