

**The Effect of Organic Loading on Process Performance and Membrane Fouling in a  
Submerged Membrane Bioreactor Treating Municipal Wastewater**

by

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## **ABSTRACT**

The results of experiments on municipal wastewater primary effluent are presented for a pilot-scale submerged membrane bioreactor (SMBR). The SMBR pilot plant employed an ultrafiltration membrane with a nominal pore size of 0.035  $\mu\text{m}$  and was operated at a constant membrane flux of 30  $\text{L}/\text{m}^2\text{h}$ . The mixed liquor suspended solids (MLSS) concentration was maintained at  $8\pm 2$   $\text{g}/\text{L}$  and steady-state fouling rates were determined for 10, 5, 4, 3, and 2-d MCRTs, corresponding to food to microorganism (F/M) ratios of 0.34, 0.55, 0.73, 0.84, and 1.41  $\text{gCOD}/\text{gVSSd}$ , respectively. Membrane fouling rates increased as the F/M was increased. Steady-state membrane fouling rates were correlated with total soluble microbial products (SMP) concentrations. The membrane fouling rates did not correlate well with soluble COD measured on a 0.45  $\mu\text{m}$  membrane filtrate of mixed liquor or with soluble COD rejection (Effluent COD/Soluble COD).

## **KEYWORDS**

Food to microorganism ratio (F/M), membrane fouling, soluble microbial products (SMP), extracellular polymeric substances (EPS), mean cell residence time (MCRT), membrane bioreactor (MBR)

## **BACKGROUND**

A submerged membrane bioreactor (SMBR) is a biological wastewater treatment process in which ultrafiltration or microfiltration membranes immersed in the mixed liquor are used for solid-liquid separation. The effect of biological conditions on reactor design, process performance and membrane performance is poorly defined. The purpose of this work was to determine the effect of biological conditions on mixed liquor properties and their effects on the process and membrane performance.

*Process Limitation:* The principal SMBR process limitation is membrane fouling. When a conventional activated sludge system with a gravity secondary clarifier (CAS) fails, the effluent quality deteriorates. When an SMBR process fails, the effluent quality is typically unaffected, but the effluent flow rate decreases due to severe membrane fouling. Regardless of mean cell or hydraulic residence times, SMBR effluents contain undetectable total suspended solids (TSS) concentrations ( $< 2$  mg/L) and have low chemical oxygen demand (COD) because of the filtration provided by the membrane (Adham et al., 2001, Bouhabila et al., 2001, Bouhabila et al., 1998, Cicek et al., 2001, Cicek et al., 1998, Cote et al., 1997, Cote et al., 1998, Muller et al., 1995, Rosenberger et al., 2002, Trussell et al., 2000, Xing et al., 2000). At frequent intervals (min) in a vacuum-driven SMBR operating cycle, the vacuum is released and most of the materials accumulated on the membrane surface are removed by coarse bubble aeration, termed relaxation. In an alternate mode of operation, product water is pumped in reverse back through the membrane to encourage increased cake removal. Regardless of the mode of operation, some TSS, colloidal particles and macromolecules remain on the membrane

and can lead to declining membrane permeability over time (Bouhabila et al., 2001, Chang et al., 2002, Cicek et al., 2002, Tardieu et al., 1998). At much longer intervals (months), the membrane is chemically cleaned to remove accumulated materials and restore permeability. Rapid loss of membrane permeability increases cleaning frequency, increases operating and maintenance (O&M) costs, decreases membrane lifetime and may reduce plant capacity (Sablani et al., 2001). The fouling rate (loss of membrane permeability over time) is a critical design parameter for determining whether an SMBR is an economically feasible treatment alternative (Kaiya et al., 2000).

*Rationale:* To maintain membrane permeability, the SMBR process is limited to maximum mixed liquor suspended solids (MLSS) concentrations of 10 - 20 g/L (Cote et al., 1998, Mourato et al., 1999, Shimizu et al., 1996, Trussell et al., 2005, Ueda et al., 1997). Current practice is to operate the SMBR process at constant MLSS levels of approximately 10 g/L (typically 8 gVSS/L). A cost effective SMBR design requires optimization of the process tank volume where a small process tank will provide immediate capital cost savings, but will increase the organic loading (F/M). High F/M may increase the O&M costs for maintaining membrane permeability (Cicek et al., 2001, Trussell et al., 2005). The goal of this research was to provide a fundamental understanding of SMBR performance at a range of F/M values so that economical design and operation, especially at high F/M, would be possible.

## MATERIALS AND METHODS

*Pilot-Scale Submerged Membrane Bioreactor (SMBR):* A pilot-scale SMBR (Figure 1), designed to operate at a range of hydraulic residence times ( $\theta_H$ ) while maintaining a constant membrane flux of 30.6 L/m<sup>2</sup>·h (LMH), was custom-built by ZENON Environmental Services Inc. (Oakville, Ontario, Canada). The reactor working volume was 1,514 L. One full-scale ultrafiltration module (ZENON 500c) was immersed in the membrane tank and intermittent (10 s on/10s off) coarse-bubble aeration of 14 L/s ( $G=630 \text{ s}^{-1}$ ) was used to control membrane fouling. The membrane had a nominal pore size of 0.035  $\mu\text{m}$  and an absolute cutoff of 0.1  $\mu\text{m}$ .

The membrane was operated at a constant, but higher, flow rate (flux) than required for maintaining the desired  $\theta_H$ .  $\theta_H$  was controlled by recycling permeate through the system.  $\theta_H$  was adjusted to maintain an MLSS concentration of 8 g/L at each operating condition in the range from 1 to 4 h. To minimize the effect of the permeate recycle on membrane performance, the mixed liquor recycle rate,  $Q_R$ , between the aeration tank and the membrane tank was constant and high, ranging between 5 and 26  $Q$  with the highest  $Q_R/Q$  being used at the highest  $\theta_H$ -(highest permeate recycle flow). Because the recirculation rate was high at all times, the MLSS concentrations were very similar in both tanks with the MLSS concentration in the membrane tank 1.2 to 1.04 times the MLSS concentration in the aeration tank.

The aeration tank was equipped with fine-bubble air diffusers. Aeration air was supplemented with pure oxygen to maintain adequate dissolved oxygen (DO)

concentrations at the higher organic loading rates. All liquid stream flow data was collected electronically and periodically checked using a graduated cylinder and stopwatch. Vacuum pressure was measured with a pressure transducer and verified by an analog vacuum gauge. Temperature was monitored using an in-tank analog thermometer and was periodically verified using a mercury thermometer.

*Feedwater Characteristics:* The pilot-scale SMBR was fed with primary effluent from the Southeast Water Pollution Control Plant (SEP), San Francisco, CA (Table 1). A centrifugal pump was immersed in the SEP primary effluent channel and operated continuously. A level sensor in the aeration tank opened and closed a feed valve to maintain the desired liquid operating volume. The SMBR was shutdown for 5 h approximately once each week while the feed line was soaked with a sodium hypochlorite solution (2 g/L) to prevent biofilm growth that could seed the SMBR with filaments (Gabb et al., 1989) and alter the feed composition.

*Reactor Operation and Membrane Performance:* The reactor was operated for 3 MCRTs prior to steady-state data collection at each MCRT tested. ML DO concentration was  $\geq 2$  mg/L and  $\text{Na}_2\text{CO}_3$  was added to the feed wastewater to control the aeration basin pH to  $\geq 6.5$  during nitrification. Sludge wasting was performed on a continuous basis by pumping from the upper portion of the membrane tank.

The effects of F/M on membrane fouling were determined by holding the membrane flux and MLSS concentration constant for all conditions tested. The steady-state membrane-

fouling rate, defined as the specific flux decline at 20°C over a 2-week period, was determined at each MCRT once steady-state conditions were achieved. The membranes were chemically cleaned with a 1,000 mg/L NaOCl solution for 12 h between each operating condition.

*Foam Control:* The SBR process traps microorganisms larger than the membrane pores including those that float and may cause foam. Biological foaming caused by nocardioform bacteria was controlled in the SBR by mechanical means (Figure 2) rather than with an NaOCl surface spray (Trussell et al., 2000). When the membrane tank intermittent aeration was on, its liquid level was higher than that in the ML recycle line; when the membrane tank intermittent aeration was off, its liquid level was lower than that in the ML recycle line. This cycling of liquid levels pulled the foam into the ML recycle line, mixing it into solution as it fell into the aeration tank where foaming was controlled by continuous surface mixing.

*MLSS and COD:* MLSS and effluent COD were measured by Standard Methods, Methods 2540D and 5220D, respectively (APHA, 1998). Mixed liquor was filtered through a 0.45 µm membrane and the filtrate COD was defined as soluble COD.

*Soluble Microbial Products (SMP):* ML samples were collected, immediately cooled to 4°C, and analyzed within 2 h from the time of collection. Although SMP contains highly complex organic molecules, polysaccharides and proteins were used to quantify and better characterize the SMP because they comprise a major component of SMP found in

the literature (Barker and Stuckey, 1999). The untreated ML was centrifuged for 15 min at 12,000 g, and the protein and carbohydrate concentrations were determined on the supernatant to represent the soluble fraction (Soluble Microbial Products, SMP). Supernatant carbohydrate and protein concentrations were measured colorimetrically by the methods of Dubois et al. (1956) and Lowry et al. (1951), respectively.

## **RESULTS AND DISCUSSION**

The pilot-scale SMBR was operated at 10, 5, 4, 3, and 2 d MCRTs to evaluate the effects of F/M on process and membrane performance. Table 2 summarizes the pilot-scale SMBR operating conditions. The highest F/M tested was 1.41 gCOD/gVSS·d (2-d MCRT) and the lowest F/M tested was 0.34 gCOD/gVSS·d (10-d MCRT). Reported SMBR F/Ms are typically much lower than those tested here. However, Ng and Hermanowicz (2005) operated an SMBR on synthetic wastewater at F/Ms up to 11 gCOD/gVSS·d and Ognier et al. (2004) operated an external MBR (EMBR) on synthetic wastewater at an F/M of 1.7 gCOD/gVSS·d.

*Process Performance:* The median MLSS concentrations for the pilot-scale SMBR (Table 3) ranged from 6.9 to 8.6 g/L and the median percent volatile fraction was 85.5% at all conditions tested. Effluent TSS concentrations were always below the detection limit of 2 mg/L, indicating good membrane integrity.

The pilot-scale SMBR reduced the influent COD (median= 345 mg/L) to a median effluent COD concentration (eff COD) of 24 mg/L (93% removal) and a 90<sup>th</sup> percentile

value of 32 mg/L at all conditions tested. In previous SBR experiments on municipal wastewater, Trussell et al. (2005) obtained an eff COD concentration of 22 mg/L and a 90<sup>th</sup> percentile value of 28 mg/L over the range of MCRT values from 1.5 d to 35 d. At MCRTs below 2 d, Trussell et al. (2005) found that the eff COD increased to a median value of 30 mg/L and a 90<sup>th</sup> percentile value of 32 mg/L. At an MCRT of 2 d, Cicek et al. (2001) found that the median eff COD increased to 23 mg/L from 3.5 mg/L at a 5-d MCRT treating synthetic wastewater. However, Holler and Trosch (2001) operated an EMBR process on synthetic wastewater and found that overall COD removal was 95 to 99% and independent of F/M. For the data presented here, eff COD concentrations were relatively constant (~24 mg/L) for MCRTs greater than 2 d (Table 4) while at 2-d MCRT they increased to 34 mg/L. The soluble COD was relatively constant, ranging from 56 to 78 mg/L, and this is consistent with published literature for MCRTs less than 10 d (Cicek et al., 2001). The ultrafiltration membrane rejected between 56 and 67 % of the soluble COD in the mixed liquor, confirming that membrane filtration retains a portion of the soluble organics in the bioreactor (Xing et al., 2003). Complete nitrification ( $\text{NH}_4^+ < 1$  mg-N/L) occurred at all conditions except at 2-d MCRT (Table 4) where the effluent  $\text{NH}_4^+$  concentration was 7.0 mg/L.

*Membrane Performance:* Figure 3 shows the SBR membrane performance at a 10-d MCRT (F/M = 0.34 gCOD/gVSS·d). The 90-d start-up period allowed adequate time for conditioning the new membrane and establishing stable SBR pilot operation. The membrane was chemically cleaned on Day 93 prior to the start of the 10-d MCRT operating period. Membrane flux was constant (30 LMH) except for the periods

immediately after foaming incidents and air valve failures (Days 124 and 135). The specific flux decreased to 74% of its initial value after 87 d of operation and the steady-state fouling rate at 20°C was 0.18 LMH/bar d.

Membrane performance results for the 5-d MCRT ( $F/M = 0.55$  gCOD/gVSS·d) are also shown in Figure 3. Initially, residual NaOCl from chemical cleaning caused foaming and the membrane flux was reduced to 13 LMH until 4 d after last foaming event (Day 185). After stabilizing, the reactor was operated for 82 d at 30 LMH by which time the specific flux had decreased to 67% of its initial value. The steady-state fouling rate at 20°C was 0.39 LMH/bar d.

Figure 4 presents membrane performance results at the 4-d MCRT ( $F/M = 0.73$  gCOD/gVSS·d). The membrane flux and coarse airflow rate were reduced following the chemical cleaning (Day 180) and when malfunctions of the intermittent air valve caused foaming (Days 295 to 297). The reactor was operated for 54 d by which time the specific flux had decreased to 84% of its initial value. The steady-state fouling rate at 20°C was 0.59 LMH/bar d.

The routine weekly feed line cleaning with NaOCl solution strongly influenced membrane performance at the 3-d MCRT ( $F/M = 0.84$  gCOD/gVSS·d) (Figure 4). The specific flux increased significantly following each feed line cleaning. The weekly cessations of permeate production from the SMBR provided an extended membrane relaxation that helped to restore the membrane permeability. It is also possible, that at this

high F/M condition, the cessation of wastewater feed allowed the biology adequate time to consume influent organics and contributed to the restoration of membrane permeability. The reactor was operated for 35 d and the steady-state fouling rate was estimated for the longest operational period (12 d) to be 1.57 LMH/bar d at 20°C.

Figure 5 shows the membrane performance data at the 2-d MCRT ( $F/M = 1.41$  gCOD/gVSS·d). After initially operating for 3 d at reduced flux because of a foaming event at start-up (Day 394), the membrane performance was evaluated over a 2-week period. Routine chlorination of the feed line was suspended during this period of operation. The steady-state fouling rate at 20°C was 3.65 LMH/bar d.

Figure 6 shows the effect of F/M on steady-state membrane fouling rates and for the approximately 4-fold range of F/Ms tested (from 0.34 to 1.41 gCOD/gVSS·d), the membrane fouling rate increased 20-fold (from 0.18 to 3.65 LMH/bar d). No membrane fouling rates have previously been reported for high F/M conditions in MBRs. Cicek et al. (2001) found that membrane permeability decreased 1.5 times for an approximate 3-fold increase in F/M from 0.30 to 1.05 gCOD/gVSS·d (MCRT from 5 to 2 d) for an EMBR treating synthetic wastewater. Steady-state membrane fouling rates were not measured and the EMBR was operated at constant pressure rather than at constant flux as in our experiments. Kimura et al. (2005) concluded that the F/M ratio has an important effect on membrane fouling rates, but differences in MLSS concentrations and membrane fluxes between different F/M ratios make interpretation of these results indefinite.

Figure 7 shows that neither soluble COD concentrations nor % COD rejection =  $(100 * 1 - \text{eff COD} / \text{soluble COD})$  by the membrane in the pilot-scale SMBR have an effect on membrane fouling rate. Cicek et al. (2002) obtained similar results from a pilot-scale EMBR treating synthetic wastewater at 30-d MCRT. The soluble COD ( $<0.1 \mu\text{m}$  filtration) concentration correlated well ( $R^2=0.80$ ) with membrane permeability, but there was little correlation ( $R^2= 0.63$ ) between membrane permeability and soluble COD using  $0.45\text{-}0.22 \mu\text{m}$  filtration (similar to the  $0.45 \mu\text{m}$  filtration that used in our experiments).

Carbohydrate SMP ( $\text{SMP}_c$ ) and total SMP but not protein SMP ( $\text{SMP}_p$ ) correlated well with steady-state membrane fouling rates in the pilot-scale SMBR (Figure 7).

Rosenberger and Kraume (2003) found good correlation between  $\text{SMP}_c$  concentration and membrane fouling rates for 8 different SMBR sludges in a batch cross-flow membrane cell. Cicek et al. (2002) also found good correlation between soluble carbohydrate concentrations and membrane permeability. Although membrane fouling rates correlate well with  $\text{SMP}_c$ , the authors believe that the total SMP concentration controls the membrane fouling rates, due to an exposure to increased soluble organic content, regardless of whether it is  $\text{SMP}_c$  or  $\text{SMP}_p$ . Recently, Zhou et al. (2005) attributed mixed liquor colloidal organic content as the primary mixed liquor property to impact the membrane critical flux in SMBRs. Colloidal organic content was determined by centrifugation of the mixed liquor and measuring total organic content. This measurement and conclusion supports our findings that the total SMP concentration controlled the membrane fouling rates for SMBRs at high F/Ms.

## CONCLUSIONS

A pilot-scale SBR was operated at high F/M to evaluate process and membrane performance. Over the range of F/M tested (0.34 to 1.41 gCOD/gVSS·d), effluent quality was consistently excellent with CODs ranging from 22 to 34 mg/L and effluent TSS levels below the detection limit (< 2 mg/L). Steady-state membrane fouling rates increased 20-fold over a 4-fold increase in F/M. Soluble COD (0.45 µm membrane filtrate of ML) and soluble COD rejection did not correlate with steady-state membrane fouling rates while total SMP and SMP<sub>c</sub> concentrations correlated well with steady-state membrane fouling rates. Although biological foaming is ubiquitous in systems with a “trapping environment” like that of the SBR (Jenkins et al., 2004, Narayanan, 2003), mechanical mixing of the foam layer into solution controlled biological foaming in this pilot-scale SBR.

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Table 1. Feed Wastewater (Primary Effluent) Characteristics

Analyte	Units	Median	Range		
Total alkalinity	mg CaCO <sub>3</sub> /L	188	89	-	248
Ammonia	mg N/L	27	14	-	33
COD	mg/L	345	78	-	695
TKN	mg N/L	30	20	-	34
TSS	mg/L	99	57	-	242

Table 2. Summary of pilot-scale SMBR operating conditions

Parameter	Units	Value
Reactor volume	L	1514
Hydraulic retention time, $\theta_H$	h	1.1-3.6
MCRT	d	10, 5, 4, 3, 2
F/M Ratio	gCOD/gVSS·d	1.41, 0.84, 0.73, 0.55, 0.34
Membrane flux	LMH (gfd)	30 (18)
Coarse Bubble Aeration	L/s	14.2
Active membrane area	m <sup>2</sup>	61.3
MLSS concentration	g/L	8±2
Recirculation rate	Q <sub>r</sub> /Q	5-26

Table 3. MLSS, VSS%, effluent COD, soluble COD, and ammonia concentrations

MCRT, d	MLSS, g/L	VSS, %	COD, mg/L		NH <sub>4</sub> <sup>+</sup> , mg-N/L
			<i>Effluent</i>	<i>Soluble</i>	
2	6.9	84	34	78	7.0
3	7.6	85	22	59	0.4
4	8.2	85	24	62	0.5
5	8.6	87	25	76	0.5
10	7.8	85	24	56	0.1

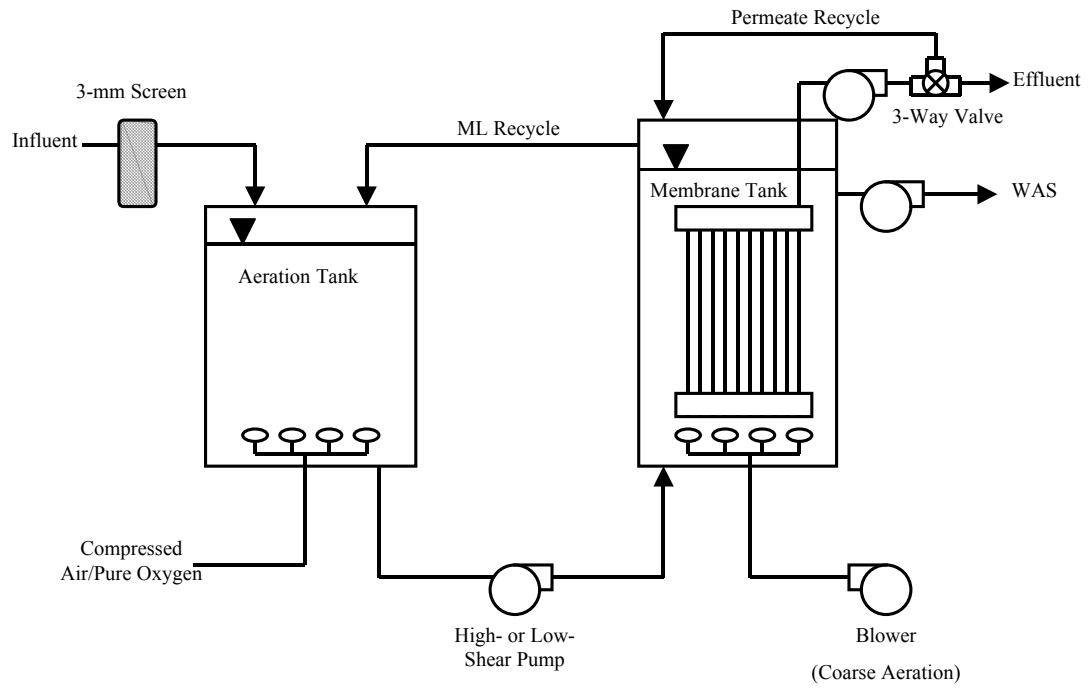
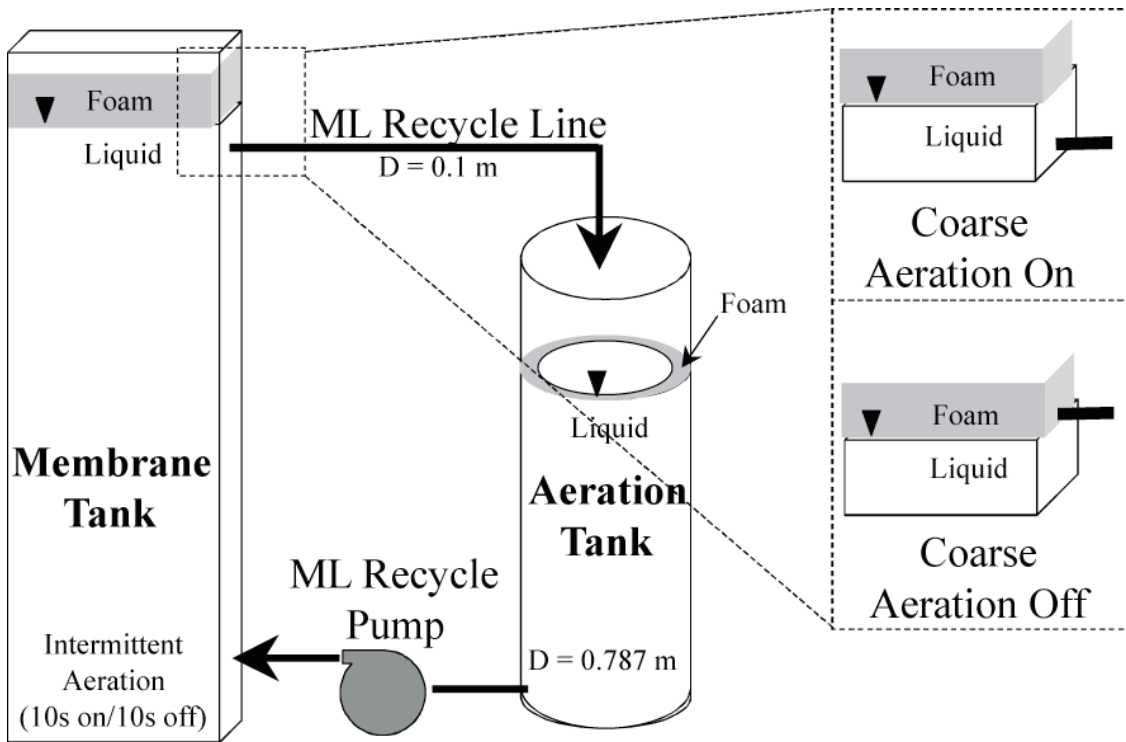
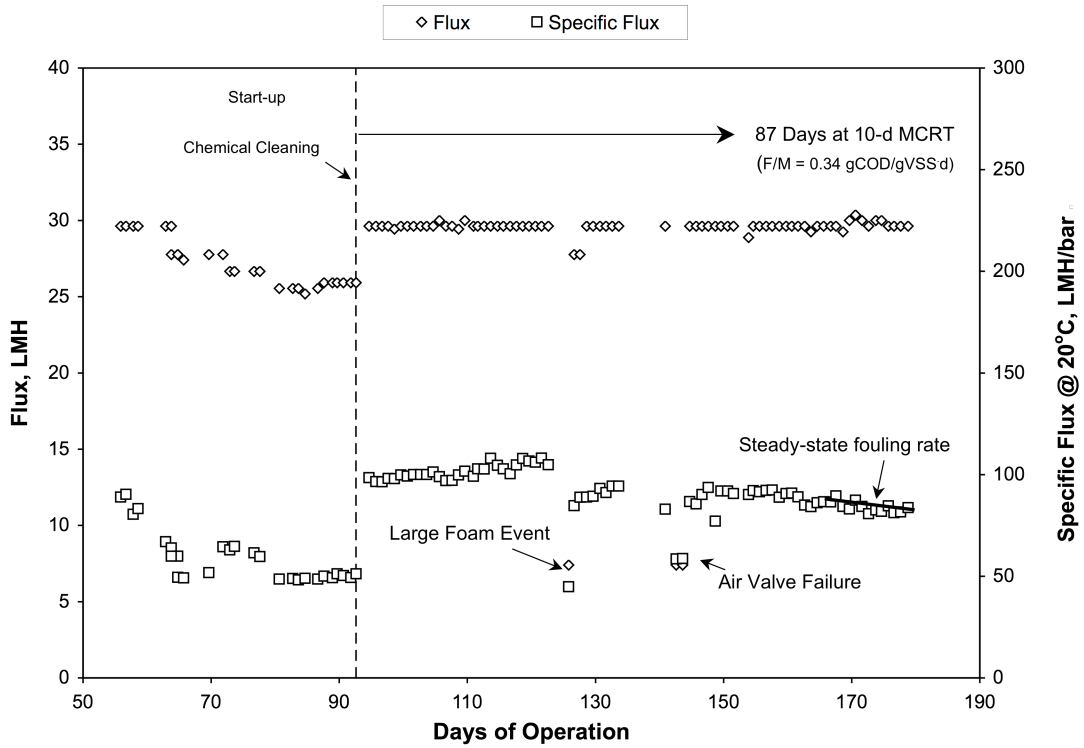


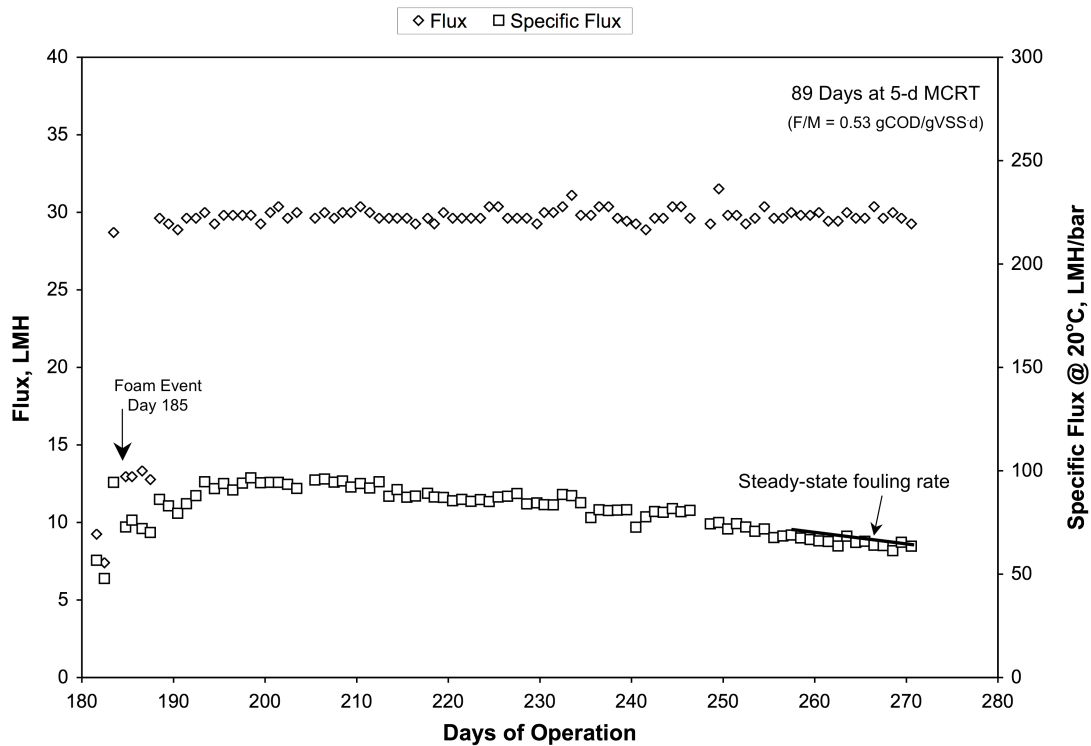
Figure 1. Schematic of the pilot-scale SMBR



**Figure 2. Illustration of mechanical foam control**

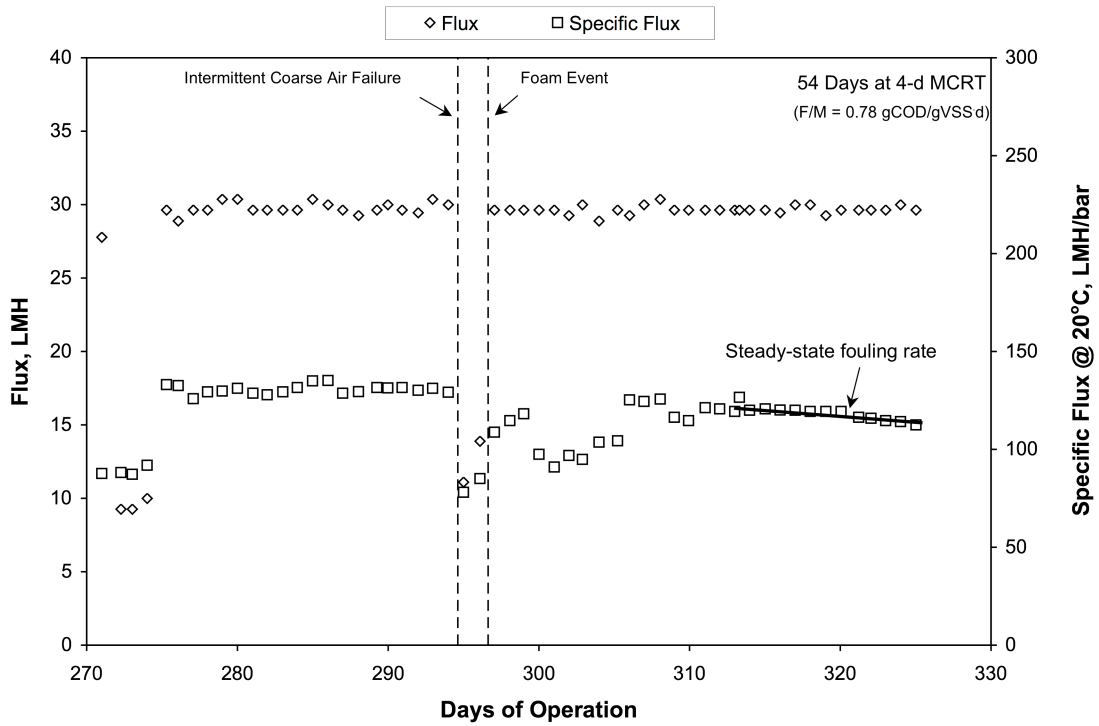


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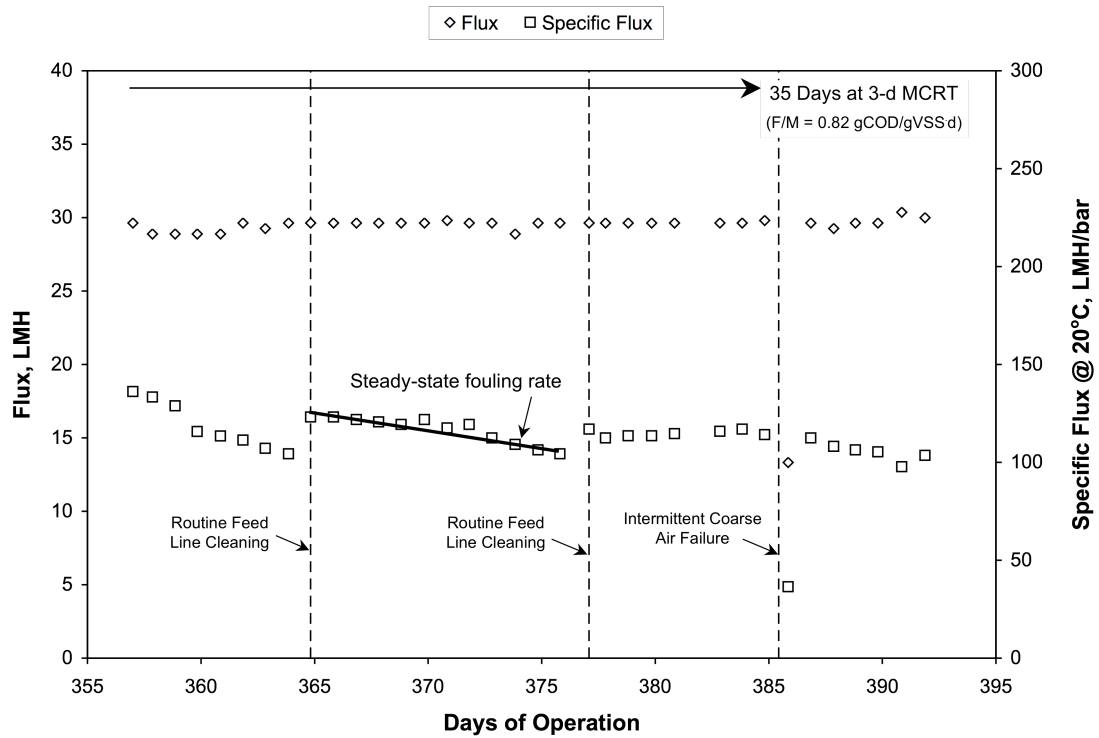


b)

**Figure 3. Membrane performance at a) 10-d and b) 5-d MCRTs**



a)



b)

**Figure 4. Membrane performance at a) 4-d and b) 3-d MCRTs**

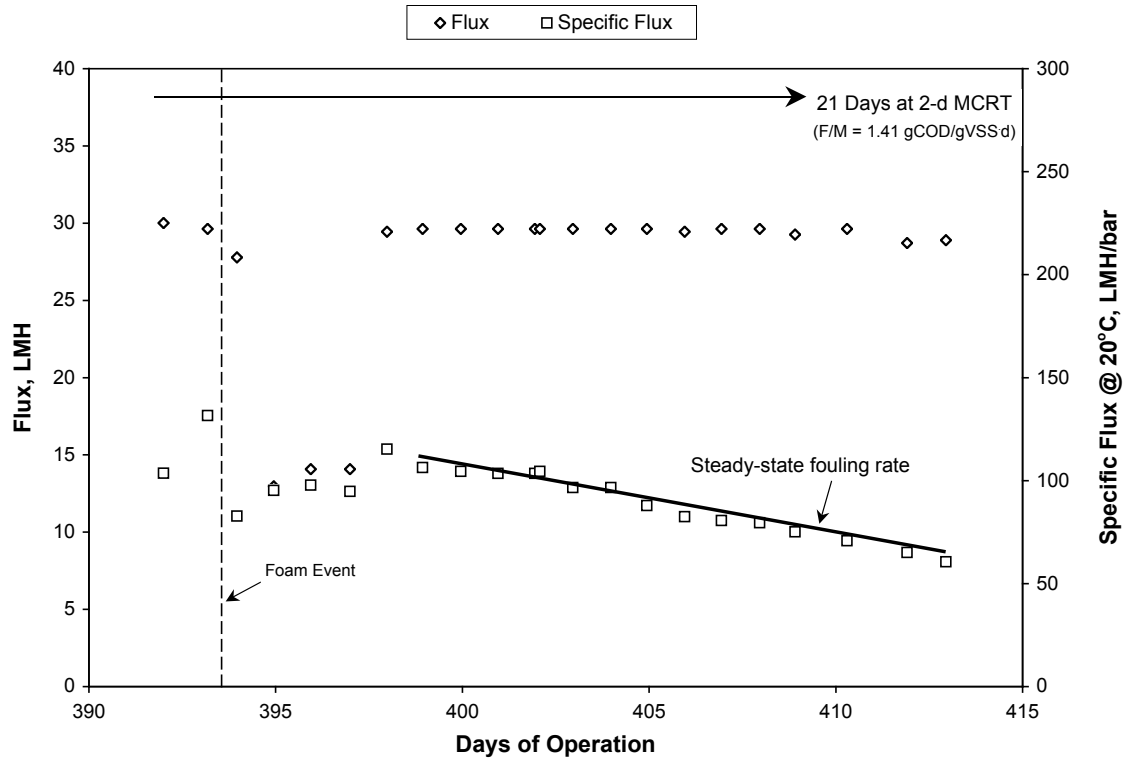


Figure 5. Membrane performance at 2-d MCRT

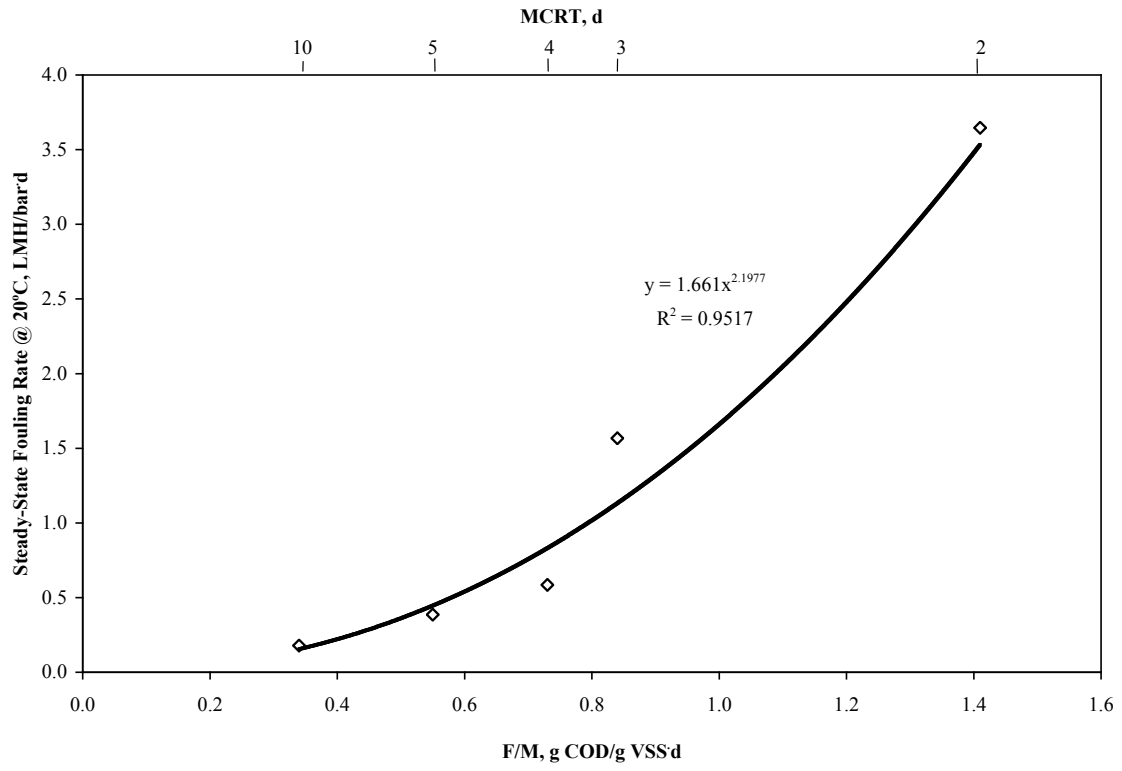
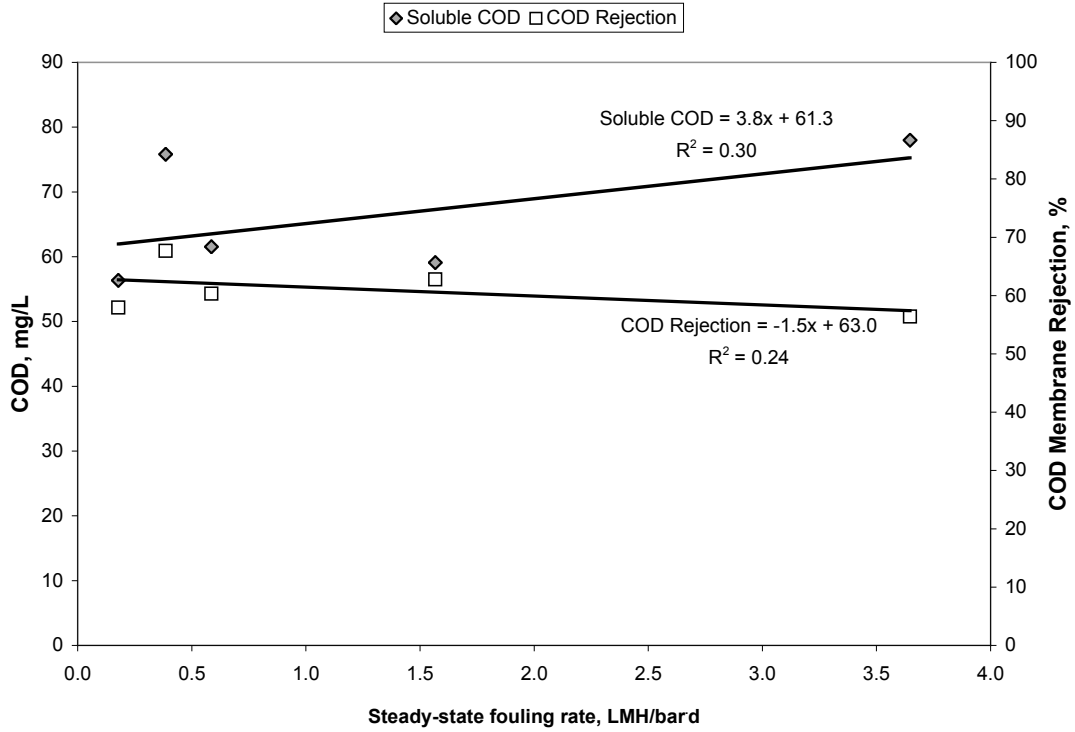
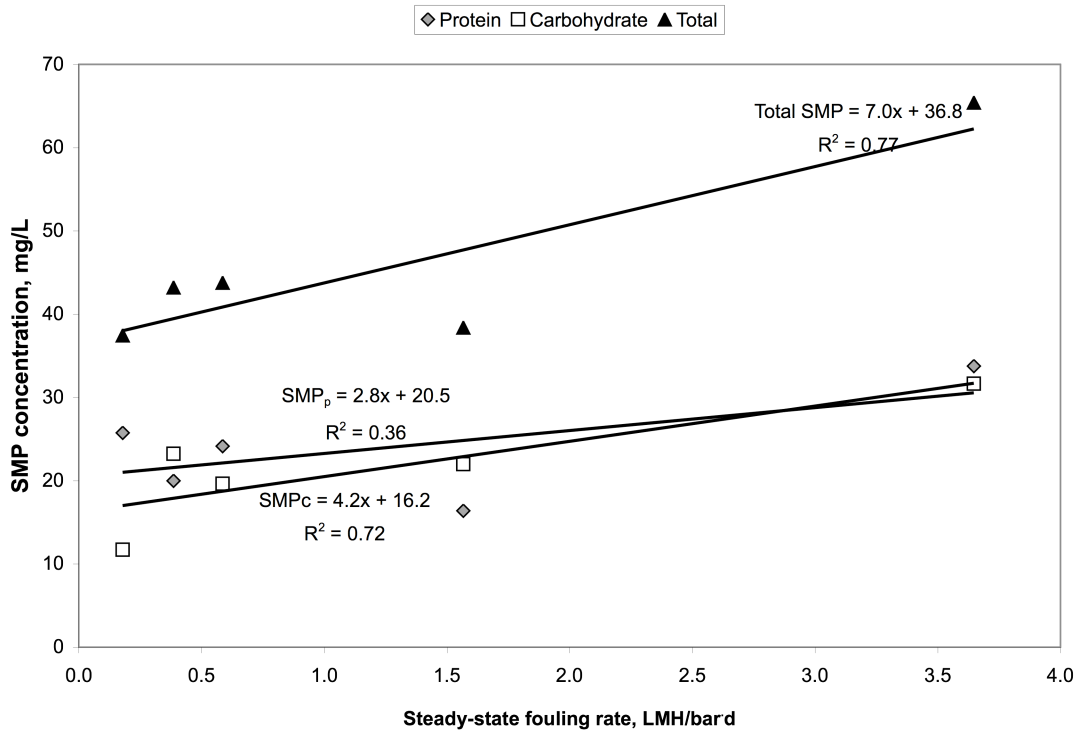


Figure 6. Effect of F/M on steady-state membrane fouling rate



a)



b)

**Figure 7. Correlations of steady-state membrane fouling rates to a) soluble COD and COD rejection and b) SMP<sub>c</sub>, SMP<sub>p</sub>, and total SMP**