Dynamic characterization of a FeCl₃-dosed anaerobic membrane bioreactor (AnMBR) treating municipal wastewater
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ABSTRACT
A transient study was conducted at pilot scale to assess the impact of Fe dosage on the dynamics of biological and membrane performance of an anaerobic membrane bioreactor (AnMBR) treating authentic municipal wastewater. A transient model of the AnMBR system was employed to assist with interpretation of the observed responses in the mixed liquor under different FeCl₃ dosages. A high dosage (43 mg FeCl₃/L sewage) resulted in a significant accumulation of fixed suspended solids and volatile suspended solids (VSS) and reduction of colloidal COD in the mixed liquor. The elevated dosages appeared to reduce the biodegradability of VSS that was present in the raw wastewater. Intermediate dosages of FeCl₃ (21–12 mg/L) had less effect on these responses and did not appear to affect VSS biodegradation. Membrane performance was significantly affected by FeCl₃ dosage as indicated by reversible resistance (RR) and physically irreversible resistance (IR). RR was closely related to the colloidal COD in the mixed liquor, thus responded quickly to Fe dosage. Physically, IR had a delayed response to changes in the colloidal COD concentrations in the mixed liquor and this was attributed to the effect of slow mass transfer of colloidal matter between the mixed liquor and the membrane.

Key words | bioavailability, colloidal matter, ferric iron, membrane fouling

INTRODUCTION
There is growing interest in developing sustainable technologies for wastewater treatment to recover, other than remove, organic matter, nitrogen and phosphorus as sources of energy and fertilizer (McCarty et al. 2014). Anaerobic membrane bioreactors (AnMBRs) are of particular interest because the anaerobic process has low energy and nutrients requirements, low sludge production, and can generate biogas that can be directly employed as an energy source (Rittmann & McCarty 2000). In AnMBRs, the membrane can completely retain biomass in the bioreactor to maintain long solids retention time (SRT) without prolonging the hydraulic retention time (HRT) (Huang et al. 2011; Smith et al. 2012). However, the application of AnMBRs has been limited due to concerns about membrane fouling, which can offset the advantages of AnMBRs.

Membrane fouling may result from the formation of a loosely-attached cake on the surface of the membrane and the development of a strongly-attached cake with solutes, e.g. a gel layer and pore narrowing or pore blocking. The former is often assumed to be reversible while the latter is deemed to be irreversible (Wang et al. 2014). The reversible resistance (RR) is assumed to represent the presence of foulants as a loosely-attached cake layer and can be removed by physical cleaning such as air or biogas sparging and mixed liquor recirculation during relaxation. The irreversible resistance (IR) is generally assumed to be indicative of the accumulation of foulants as either a strongly-attached cake layer or pore blocking. The term irreversible fouling is also known as physically irreversible fouling, since it typically cannot be removed by physical cleaning (Wang et al. 2014).

The addition of FeCl₃ to the mixed liquor may improve membrane performance. It has been shown that the addition of FeCl₃ can effectively mitigate membrane fouling in aerobic MBR systems (Fan et al. 2007; Koseoglu et al. 2008; Song et al. 2008; Zhang et al. 2008; Ji et al. 2010). In these prior studies, the reduction in fouling was attributed...
to increased particle sizes and lower concentrations of soluble and colloidal materials. However, the interaction of the FeCl₃ with components in mixed liquor was not assessed. There is limited information available that describes the transient responses of MBRs, and specifically AnMBRs, to FeCl₃ addition.

Therefore, this study employed a transient approach to characterize the impacts of FeCl₃ addition on pilot-scale AnMBR biological and membrane processes. It was anticipated that this approach would provide insight into how these systems will dynamically respond to changes in Fe addition. Further, it was anticipated that the study would provide insight into time-dependent processes such as mass transfer of foulants from the mixed liquor to the membrane surface. In order to assist with interpretation of the system dynamics, a general transient model of the AnMBR system was assembled and subsequently employed as a framework to provide dynamic baselines of the responses based on specific assumptions regarding contaminant behaviour. By comparing the baselines predicted by the model with the observed data, the dynamics of the measured responses were interpreted. The results of this study provide operators of AnMBRs with insight into how these systems will dynamically respond to changes in Fe addition. The study also sought to provide insight into time-dependent processes such as mass transfer of foulants between the mixed liquor and the membrane surface.

MATERIALS AND METHODS

A pilot scale AnMBR that was fed with 3 mm screened sewage from the Burlington Skyway Wastewater Treatment Plant (Ontario, Canada) was employed in this study (Figure 1). The pilot AnMBR consisted of a 550 L completely mixed anaerobic digester (AD) and an 80 L membrane tank (MT) that held a polyvinylidene fluoride hollow fibre membrane module with a surface area of 5.4 m² and a pore size of 0.04 μm (GE: ZeeWeed 500). The AD contents were mixed by recirculation of the contents at a flow of 3,600 L/h with a positive displacement pump. The AD contents were circulated through the MT using a centrifugal pump that withdrew mixed liquor from the bottom of the AD and pumped it to the bottom of the MT, after which it flowed to the top of the AD by overflow. This circulation mixed the MT contents and generated a cross flow velocity that would enhance surface shear for membrane fouling control. Biogas produced in the AD was released from the head space of the AD and its production was measured by an electrical gas flow meter. Biogas was recirculated through the MT at a flow of 0.786 m³/h (20 °C and 1 atm) with a blower (KNF NEUBERGER, PM23820-150.1.2) to reduce membrane fouling.

Throughout the study, the temperature of the digester was maintained at 23 ± 1 °C by a heat tape that was controlled by a temperature controller which was informed by
a temperature sensor in the digester. The system tended towards lower pH due to elevated CO₂ partial pressures. The pH of the digester was controlled through NaHCO₃ addition to maintain a value in the range of 6.7–6.8, as it is relatively close to 7.0 and should have no significant impact on the anaerobic biological process, but saved cost in dosing NaHCO₃. Pilot operation and data acquisition were controlled using a programmable logic controller. The feeding of raw sewage and the wasting of AD contents for SRT control were controlled on the basis of the weight of the digester, which was monitored by load cells installed at the base of the digester.

The pressures of the headspace in the MT and permeate line were recorded using digital pressure gauges. The transmembrane pressure (TMP) was calculated as the difference between the pressure in the MT and that of the permeate line. To reduce membrane fouling, a discontinuous filtration mode was incorporated into the pilot operation. The discontinuous filtration mode consisted of a repeating cycle that had filtration for 8 minutes and relaxation for 2 minutes.

Membrane performance was characterized by monitoring the TMPs before (TMP(b)) and after (TMP(a)) relaxation. Hence, based on the classification by Wang et al. (2014), the TMP(a) values were defined to be indicative of physically irreversible fouling that included all the fouling that could not be removed by mixed liquor recirculation and biogas sparging. The TMP recovery observed during relaxation (TMP(b-a)), which was equal to the difference between TMP(b) and TMP(a) values, was defined to characterize the presence of reversible fouling. This consisted of loosely bound materials that accumulated on the membrane surface during each filtration cycle but were removed during relaxation. The subsequent discussion of the membrane performance employs this terminology.

The impact of FeCl₃ dosing on membrane performance was assessed by examining membrane resistances on the basis of the implemented flux values and the observed TMP responses. TMP(b-a) values were employed to calculate the RR (Equation (1)). As the TMP(a) included the impact of the resistance of the virgin membrane, the TMP(a) at Day 1 was subtracted from the measured TMP(a) values to exclude the resistance of the virgin membrane. Therefore, the modified TMP(a) values were employed to calculate physically IR based on Equation (1) (Lin et al. 2009).

\[ R = \frac{\Delta P}{J \times \mu} \]  

(1)

where

\[ R = \text{Resistance, m}^{-1} \]  
\[ \Delta P = \text{Trans-membrane pressure (TMP), kPa} \]  
\[ \mu = \text{Viscosity of water, Pa·s} \]  
\[ J = \text{Membrane flux, L/m}^2\text{/h} \]

Wastewater and mixed liquor samples were collected twice per week from the outlets of the feed pump (upstream of the FeCl₃ dosing) and anaerobic reactor, respectively. The samples were analyzed for total suspended solids (TSS), volatile suspended solids (VSS), and fixed suspended solids (FSS), which was determined as the difference between TSS and VSS and chemical oxygen demand (COD). All analyses were conducted according to standard methods (APHA 2005). The methane concentration in the biogas was measured using a gas chromatograph (Agilent Technologies G2802A).

The sewage and mixed liquor were analyzed for colloidal COD (cCOD) to provide insight into the presence of potential foulants. The 1.5 µm-filtered COD was analyzed after centrifuging the sewage or mixed liquor sample at 4,000 rpm for 12 minutes then filtering the supernatant through a 1.5 µm glass fiber filter. The 0.45 µm-filtered COD was analyzed after filtering the 1.5 µm-filtered COD sample through a 0.45 µm glass fiber filter. The cCOD was calculated by subtracting 0.45 µm-filtered COD from the 1.5 µm-filtered COD (Fan et al. 2006).

Prior to the onset of this study, the pilot AnMBR was operated at an HRT of 14.5 hours (flux of 10 LMH) and an SRT of 70 days for 85 days without FeCl₃ addition. During the entire experiment, the SRT was maintained at 70 days and the HRT was varied by adjusting the flux over a range from 14.5 hours (flux = 10 LMH) to 8.5 hours (flux = 17 LMH). At the beginning of the study a virgin membrane was installed, and hence the first stage of the work was assumed to be at biological steady state as indicated by a stable VSS concentration of 6.5 ± 0.4 g/L in the bioreactor. Therefore, the data collected in the first stage of the study was deemed to represent bioprocess steady state and was employed in the subsequent comparison when FeCl₃ was added to the influent.

The research was conducted in four stages (Figure 2) where the Fe dose and the membrane flux were varied. Initially the system was operated without Fe addition (Stage 1) and this was followed by sequential periods of high (Stage 2), intermediate (Stage 3) and high (Stage 4) dosage. The membrane flux was maintained at a low value (10 LMH) until the midpoint of Stage 3, after which it was sequentially ramped up to 17 LMH.
RESULTS AND DISCUSSION

Equation (2) was assembled based on specific assumptions regarding contaminant behaviour and subsequently employed as a framework to evaluate the dynamic responses of the concentrations of FSS, cCOD and VSS. The values predicted by the equation provided a baseline to compare with the observed data from where the dynamics of the measured responses were interpreted.

\[
\frac{dX}{dt} V = QX_0 - Q_w X_{AD}
\]  

(2)

where

\[
\frac{dX}{dt} = \text{rate of change of concentration in reactor, g/L/d} \\
V = \text{reactor volume, L} \\
Q = \text{influent flow, L/d} \\
X_0 = \text{concentration of particulate in influent, g/d} \\
Q_w = \text{waste sludge flow, L/d} \\
X_{AD} = \text{concentration of particulate in reactor, g/L.}
\]

As will be subsequently discussed, Equation (2) was employed in several contexts to evaluate hypotheses regarding the role of iron addition in the system. In most cases the model simulations, which employed raw wastewater characteristics (Table 1) as inputs, were compared with observed responses in the digester. A review of Table 1 reveals that while there was variability in the raw wastewater characteristics, there were no substantial changes in this stream with time. Hence, it was assumed that the subsequently described responses that were observed in the digester were due to changes in the operating conditions and not due to changes in the wastewater properties.

The concentrations of total Kjeldahl nitrogen (TKN) and total phosphorus (TP) in the influent are presented in Table 1 and it can be observed that they were within the typical range of values present in municipal wastewater. FeCl₃ dosing had no significant influence on the removal of TKN and TP during the experiment. This was consistent with our previous work in a steady-state study (Dong et al. 2015). The absence of an effect on TP and TKN removals is hypothesized be attributed to the reduction of ferric iron to ferrous iron in the anaerobic environment, which would modify the nature of the complexes and precipitates that were formed.

It was hypothesized that the Fe that was dosed into the wastewater would either coagulate organic matter or precipitate in the digester. In either case, it would contribute to the FSS present in the digester and hence changes in the FSS concentration in the digester could be employed to evaluate the fate of Fe and to assess the validity of the dynamic modeling approach employed in this study. Figure 3 presents the FSS concentrations that were observed in the AnMBR over the duration of the study. From Figure 3 it can be observed that during Stage 1 the FSS concentrations
were essentially constant with time and this material was attributed to the FSS that were present in the raw wastewater. Upon addition of Fe in Stages 2–4, the FSS concentration increased with time and the rate of increase was a function of the Fe dose.

Equation (2) was employed to predict the FSS concentrations during the period of Fe dosing by using the measured FSS concentration in the digester contents as the initial conditions. The integrated form of Equation (2) was then applied to each time interval that had a different combination of Fe dose and membrane flux to predict the FSS in the mixed liquor on the assumption that only the added Fe contributed to the FSS. It was anticipated that this approach would yield a conservative estimate of the FSS, as it did not account for precipitated inorganic anions. The fraction of the Fe that formed precipitates was not known, and hence the model simulations provided a reference point for assessing the fate of Fe.

Table 1 | Sewage characteristics

<table>
<thead>
<tr>
<th>Stage</th>
<th>Total BOD₅ (mg/L)</th>
<th>Total COD (mg/L)</th>
<th>cCOD (mg/L)</th>
<th>VSS (mg/L)</th>
<th>FSS (mg/L)</th>
<th>Fe (mg/L)</th>
<th>TKN (mg/L)</th>
<th>TP (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>140 ± 21.4</td>
<td>295 ± 64.8</td>
<td>10.3 ± 2.7</td>
<td>102.9 ± 35.7</td>
<td>21.6 ± 10.3</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>152 ± 20.1</td>
<td>262 ± 36.7</td>
<td>15.0 ± 3.8</td>
<td>79.1 ± 15.8</td>
<td>14.5 ± 4.7</td>
<td>1.19 ± 0.40</td>
<td>45.2 ± 9.1</td>
<td>4.1 ± 0.8</td>
</tr>
<tr>
<td>3</td>
<td>186 ± 26.9</td>
<td>291 ± 59.3</td>
<td>13.7 ± 4.0</td>
<td>99.4 ± 20.5</td>
<td>28.3 ± 6.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>162 ± 26.2</td>
<td>274 ± 29.7</td>
<td>14.7 ± 4.3</td>
<td>92.6 ± 19.7</td>
<td>22.8 ± 5.5</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 3 compares the model simulations with the observed FSS concentrations over the duration of the study. From Figure 3 it can be observed that the model was able to effectively predict the FSS concentrations over most of the period of Fe dosing. In Stages 2 and 4, the predicted values were somewhat less than the observed values, suggesting that there was some formation of inorganic precipitates at the high dose rate. This was less apparent at the intermediate dosages in Stage 3; however, the simulation periods in this stage were shorter and hence there was less operational time for the formation of inorganic precipitates to be observed. Overall, it was concluded that the modeling approach employed was valid and hence it was employed to further evaluate the behavior of other particulate species in the AnMBR.

The addition of Fe to aerobic MBRs (Fan et al. 2007; Koseoglu et al. 2008) and AnMBRs (Dong et al. 2015) has been previously demonstrated to coagulate colloidal matter, in that is likely responsible for membrane fouling.
Hence, the colloidal COD (cCOD) of the mixed liquor was monitored to evaluate the response of the system to FeCl₃ addition (Figure 4). From Figure 4 it can be observed that without Fe addition (Stage 1) the cCOD concentrations in the mixed liquor were in the range of 900–1,200 mg/L. Immediately after the start of a high level of Fe dosing in Stage 2, the cCOD concentrations declined significantly to reach approximately 600 mg/L by the end of the stage. When reduced iron dosing was employed in Stage 3, the cCOD concentrations started to gradually increase while the membrane flux was maintained at 10 LMH. When the flux was increased, the cCOD concentrations increased more rapidly despite increases in Fe dosing in Stage 3. Only when a high level of Fe dosing was employed in Stage 4 was the cCOD concentration reduced under high flux operation. The results indicate that cCOD concentrations in the AnMBR were a function of both the Fe dosage and the membrane flux, which affected the ratio of HRT to SRT.

It was hypothesized that the cCOD in the mixed liquor would either have entered with the raw wastewater or was generated in the digester by the biological processes. It was however not clear how Fe addition influenced cCOD from these two sources. Equation (2) was therefore employed to evaluate the role of Fe addition on cCOD in the system. In this application, simulations were conducted on the assumption that Fe addition would immediately coagulate all of the cCOD in the feed wastewater and that cCOD that was generated in the digester was not impacted. Figure 4 presents the results of these simulations in comparison to the observed results. From Figure 4 it can be seen that the simulated values in Stages 2 and 4, which had a FeCl₃ dosage of 43 mg/L, declined somewhat more slowly than the observed values. Hence it would appear that under high dosage conditions the dosed Fe not only coagulated all of the cCOD in the sewage, but also coagulated some of the cCOD that was present in the AD and/or mitigated the cCOD that was generated in the AD.

In Stage 3, which was conducted at lower FeCl₃ dosages (21 to 12 mg/L), the simulations that were conducted on the assumption of complete coagulation of feed cCOD consistently underpredicted the observed values. The increase in the observed cCOD values suggested that either some of the feed cCOD was not coagulated or sufficient cCOD was generated in the AD to exceed the coagulation capacity of the Fe addition. To further investigate the contribution of Fe dosing to cCOD removal, additional simulations were conducted in Stage 3 where it was assumed that none of the feed cCOD was coagulated. When these values were considered together with the previously discussed values they formed an envelope of potential responses. From Figure 4 it can be seen that the observed values fell within this envelope. Hence, it was concluded that the lower dosages introduced insufficient Fe to coagulate all cCOD.

![Figure 4](http://iwaponline.com/wst/article-pdf/2017/2/481/216954/wst2017020481.pdf)
in sewage and mitigate cCOD generation. The dosage of FeCl₃ can therefore be employed as an operational parameter to control the cCOD in the mixed liquor.

It was anticipated that FeCl₃ would impact on the VSS concentrations in the AnMBR as it was previously demonstrated that cCOD was removed under high dosage conditions. Figure 5 presents the observed VSS concentrations over the duration of the testing. From Figure 5, it can be seen that in the absence of Fe dosing (Stage 1) the VSS concentrations in the mixed liquor were effectively constant with time. By contrast, the VSS concentrations increased substantially with high Fe dosing in Stages 2 and 4. Under conditions of intermediate Fe dosing (Stage 3) the observed VSS concentrations increased only modestly. Preliminary calculations that were performed to evaluate the potential contribution of coagulated cCOD to the production of VSS in mixed liquor revealed that this could not explain the substantial increases in VSS that were observed in the AnMBR.

An alternate mechanism that was hypothesized for the increased VSS concentrations in the mixed liquor was that Fe addition was rendering normally biodegradable VSS to be non-biodegradable. To investigate this hypothesis, Equation (2) was employed to simulate the mixed liquor VSS with the assumption that all of the VSS in the feed sewage was non-biodegradable (Figure 5). From Figure 5 it can be seen that with the addition of FeCl₃ at a dosage of 43 mg/L in Stages 2 and 4 the model was able to reasonably simulate the accumulation of VSS in the AnMBR. Hence, the results suggest that the addition of FeCl₃ at 43 mg/L substantially reduced the biodegradability of VSS entering the AnMBR, and it was expected that this would impact upon methane production.

In Stage 3, with reduced FeCl₃ dosages (21 to 12 mg/L), the simulations that were conducted on the assumption of completely nonbiodegradable VSS in the feed consistently over predicted the observed values. Hence, these results implied that at reduced dosages, there was insufficient Fe interacting with the VSS in sewage to prevent biodegradation of this material. The results indicate that the use of Fe for fouling control should be carefully controlled such that cCOD is coagulated but the biodegradability of the VSS is not inhibited.

The production of methane is an important factor to consider when assessing the sustainability of AnMBRs, as it might be employed to compensate for the energy consumed in AnMBR operation. As previously discussed, the addition of Fe appeared to substantially reduce the biodegradability of VSS and it was hypothesized that methane production would be reduced and the sludge yield would be increased (Gossett et al. 1978; Dentel & Gossett 1982). The impacts of FeCl₃ addition on the methane and sludge yields were therefore investigated. In anaerobic treatment of dilute wastewaters, it is important to account for
both the gaseous methane in the biogas and the methane dissolved in the permeate as the latter can represent a substantial fraction of the produced methane. In this study the gaseous production was directly measured (20 °C and 1 atm) and the dissolved methane was estimated (Henry’s Law) to calculate the methane yield (as COD) of the system (Figure 6). The sludge yield was evaluated in terms of the COD equivalent of VSS in the wasted sludge. From Figure 6 it can be seen that the methane yield was 0.167 ± 0.017 gCH4 as COD/g CODfeed in Stage 1, and with the addition of FeCl3 at 43 mg/L the yield was reduced to approximately 0.079 gCH4 as COD/g CODfeed by the end of Stage 2. Similarly, the methane yield decreased from 0.134 to 0.044 gCH4 as COD/g CODfeed during Stage 4. In Stage 3, when FeCl3 dosages were reduced an increase in methane production from 0.044 to 0.145 gCH4 as COD/g CODfeed was observed. On the other hand, the sludge yield showed the opposite trend to that of the methane yield. These results confirmed the accumulation of VSS and the reduction in its biodegradability due to the interaction with Fe. Therefore, the dosage of FeCl3 should be carefully controlled as it can negatively affect methane production and the economical viability of an AnMBR.

As previously described, the addition of FeCl3 to the AnMBR feed substantially impacted upon the concentrations of cCOD in the AnMBR, which is considered to be a membrane foulant. It was hypothesized that the transient responses of the reversible fouling and physically irreversible fouling would differ, as the former could be attributed to direct filtration processes while the latter would be more dependent on slower mass transfer processes. Hence, the transient response of reversible fouling and physically irreversible fouling in the AnMBR to FeCl3 addition was explored.

It was anticipated that the dosed Fe would deposit on the membrane; however, it was not expected that this would generate significant resistance because most of the Fe was in a particulate form in the mixed liquor and the particulate mass formed a thicker but more porous fouling layer when deposited on the membrane (Dong et al. 2015). Therefore, the impact of the Fe deposits on membrane fouling was not included in the conceptual model.

From Figure 7 it can be observed that without Fe addition (Stage 1) the RR values increased from 2.8*10^8 to 8.6*10^8 m^-1 in Stage 1. Immediately after the start of a high level of Fe dosing in Stage 2, the RR values declined significantly to reach approximately 0.5*10^8 m^-1 by the end of the stage. When reduced iron dosing was employed in Stage 3, the RR stayed constant while the membrane flux was maintained at 10 LMH. When the flux was increased, the RR increased more rapidly despite increases in Fe dosing in Stage 3. Only when a high level of Fe dosing was...
employed in Stage 4 was the RR reduced under high flux operation. The results indicate that RR in the AnMBR was a function of both the Fe dosage and the membrane flux (ratio of HRT to SRT), which affected the potential foulants in the mixed liquor.

As previously discussed, cCOD concentrations in the AnMBR were also a function of both the Fe dosage and the membrane flux. Hence, it was hypothesized that the cCOD was the foulant that directly contributed to the reversible fouling. To evaluate this hypothesis, the relationship between RR and cCOD concentration was examined (Figure 8). From Figure 8 it can be seen that there appeared to be a positive trend between RR and cCOD. Thus RR was influenced by the dosage of FeCl₃ and the membrane flux that established the concentration of colloidal organic matter in the mixed liquor. Hence, the RR responded quickly to operational parameters that immediately affected the cCOD concentration in the mixed liquor.

From Figure 7 it can be seen that without FeCl₃ addition IR values increased from 0 to 5.7×10⁸ m⁻¹ in Stage 1 of the testing. In Stage 2, the addition of FeCl₃ at 43 mg/L resulted in an immediate but slower reduction in IR as compared to RR. Subsequently, in Stage 3, while the membrane flux was maintained at 10 LMH the IR values continued decreasing to a minimum value of 0.1×10⁸ m⁻¹ despite the reduced FeCl₃ dosage (21 to 12 mg/L). The trend in IR values during this period differed from the RR values, which remained constant. When the flux was increased, the IR values increased rapidly despite increases in Fe dosing in Stage 3. When a high level of Fe dosing was employed in Stage 4, the IR values stabilized at approximately 7.6×10⁸ m⁻¹ under high flux operation.

In the datasets, there were some instances when the IR value was higher than the RR value. This was typically observed in the latter parts of a phase where significant fouling had developed. This response was possible because the flux values were maintained by providing sufficient TMP to overcome the resistance caused by fouling. Under these conditions, the strongly attached foulants on the membrane surface and pore blocking accumulated significantly to yield a higher resistance than the loosely attached foulants that were removed by biogas sparging.

The results indicate that IR in the AnMBR was a function of both the Fe dosage and the membrane flux, but the mechanism by which these factors affected IR differed from that of RR. The results suggest that the effects of Fe dosage on IR were due to its impact on cCOD concentration in mixed liquor but that the response was not immediate. Previous studies have demonstrated that physically irreversible fouling was correlated to cCOD concentration as its presence can contribute to the formation of a strongly attached cake layer and pore blocking (Zhou et al. 2016; Meng et al. 2017). However, in the current study there was less correlation between IR and cCOD concentration than between RR and cCOD concentration in the stages when FeCl₃ was added. The lack of correlation of IR with cCOD
concentrations may be attributed to the slow mass transfer between the cCOD in mixed liquor and the physically irreversible foulant layer. Hence an extended time was required for colloidal matter to come in contact with the membrane and to enter the pore structure. This slow process would delay the IR response relative to the changes in the cCOD concentration in the mixed liquor. Therefore, IR responded slower than RR to the changes in cCOD concentration when Fe was added.

The membrane flux would have an impact on the mass transfer of physically irreversible foulants as it determines the loading of foulants to the membrane surface. The effect of flux on IR was evaluated by comparing the IR in Stages 2 and 4. From Figure 7 it can be observed that the high dosage of FeCl₃ at 43 mg/L had limited mitigation of IR in Stage 4 as compared with Stage 2, although the cCOD in the mixed liquor was significantly reduced in both stages. The limited mitigation of IR in Stage 4 was attributed to the increased flux. An increase in flux from 10 to 17 LMH would enhance foulant loading to the membrane surface, and hence in Stage 4 the IR stabilized rather than decreasing as it did in Stage 2 when high dosages of Fe caused a decline in cCOD concentrations. This behavior was consistent with the previous conclusion that the development of physically irreversible fouling is dependent on slower mass transfer processes.

CONCLUSIONS

Insights were obtained into physical, biological and membrane responses of a pilot scale AnMBR when sequentially operated over a range of dosages of FeCl₃ and membrane fluxes. The dynamic response of FSS in the mixed liquor revealed that Fe addition contributed to the production of FSS in the system. The cCOD and VSS responses revealed that the Fe dosage positively impacted on the extent of cCOD that was coagulated and negatively impacted the biodegradability of fed VSS. The impact of Fe dosage upon biodegradability of VSS was confirmed by the methane production data. Fe dosage should be optimized to balance coagulation of cCOD for fouling control and inhibition of biodegradability of the VSS for methane production.

The dynamic response of reversible fouling to FeCl₃ addition was rapid and was attributed to a direct filtration process that was dominated by the cCOD concentration in mixed liquor. The response of physically irreversible fouling was delayed relative to the IR response and appeared to be
controlled by mass transfer of cCOD between the mixed liquor and the physically irreversible foulant layer. The results suggest that the use of Fe addition can immediately control reversible fouling, but it can take long term dosing to mitigate physically irreversible fouling. Fe addition was less effective at higher flux values that modifying the balance between rates at which physically irreversible foulants accumulate on the membrane.

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