

A MATHEMATICAL MODEL FOR A SEQUENTIAL BATCH MEMBRANE BIOREACTOR PILOT PLANT

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Abstract. A mathematical model to quantify the nitrogen removal for a membrane bioreactor (MBR) has been presented in this study. The model has been applied to a pilot plant having a pre-denitrification MBR scheme. The pilot plant was cyclically filled with real saline wastewater according to the fill-draw-batch operation. The model was calibrated by adopting a specific protocol based on extensive field dataset. The Standardized Regression Coefficient (SRC) method was adopted to select the most influential model factors to be calibrated. Results related to the SRC method have shown that model factors of the efficiency of backwashing and the biological factors affecting the soluble microbial products (utilization-associated products) (namely, fUAP and KH,UAP) strongly affects the membrane resistance. In terms of model calibration excellent results in terms of model efficiency were found for the total membrane resistance model output (efficiency equal to 0.79). Regarding the biological model outputs acceptable were found in the case an high number of measured data was available. In terms of uncertainty, it was found that for the great part of the analyzed model outputs the measured data lay inside the uncertainty bands.

1. Introduction

Membrane Bioreactor technology (MBR) represents one of the best alternative technologies compared to the traditional ones (e.g. conventional activated sludge (CAS)) in order to achieve the very stringent requirements in terms of effluent quality of the treated wastewater, (Gabarraon et al., 2015). MBRs offer several advantages over the CAS (e.g. high effluent quality, reduced footprint, lower excess sludge, higher organic loading rates applicable) (Judd and Judd, 2010). Thus, the use of MBR has considerably increased during the last years (Judd and Judd, 2010). However, despite the numerous advantages of MBR over CAS the MBR technology is affected by crucial issues that may hamper a widespread application. Membrane fouling is certainly one of the major obstacles in MBR operation (Drews, 2010). Indeed, membrane fouling, causing the permeability reduction and/or an increasing of transmembrane pressure (TMP), leads to the increase of the operating costs. Due to its crucial aspect in MBR operation, membrane fouling has been widely investigated in order to better identify factors strongly affecting its worsening (Pretel et al., 2016). During the last years many researchers recognized that the Soluble Microbial Products have an important role in membrane fouling (Drews, 2010). SMPs have been divided

into two main fractions: utilization-associated products (UAP) and biomass-associated products (BAP) (Namkung and Rittmann, 1986). UAPs are produced from the substrate degradation. Conversely, BAPs can be produced by the during the decay of the active biomass or due to the hydrolysis of bound extracellular polymeric substances (EPS) or during both processes (Aquino and Stuckey, 2008). Despite the useful insights gained by previous experimental studies, there are still some gaps in the knowledge for understanding the role played by the overall operating conditions in the definition of the optimal conditions for reducing fouling (i.e. economic costs). Indeed, experimental studies may present some limits in terms of both economic costs and investigation time requirements.

In this context, MBR mathematical models represent an useful tool to predict membrane fouling and to select the best operating conditions to reduce fouling (Mannina and Cosenza, 2013). MBR models have the advantages of providing the possibility to explore a wide range of operating conditions and compare different solutions prior to their effective realization/application.

From the literature three MBR modeling approaches can be pin down (Fenu et al., 2010): biomass kinetic models, membrane fouling models and integrated models. The kinetic models are based on the activated sludge models (ASMs) taking also into account the formation and degradation of the soluble microbial products (SMPs) in the MBR (Mannina and Di Bella, 2012). The hybrid models enable to describe the influences of SMPs in the biological processes and effluent quality (Zuthi et al., 2012). Membrane fouling models takes into account the physical processes modelling. Finally, the integrated models, basically couple the kinetic models with the fouling one (such the resistance-in-series model) and they often consider the formation and degradation of SMP. Recently Zuthi et al. (2013) addressed the importance of using integrated modeling approach with the use of resistance-in-series models in order to better simulate the membrane fouling. During the last years several modelling efforts have been also performed with the aim of introducing the role of SMP in the physical fouling mechanism process. Therefore, several mathematical models have been developed introducing SMP kinetics into the bioprocess of MBR (Oliveira-Esquerre et al., 2006; Jiang et al., 2008; Mannina and Di Bella, 2012) or with the extension of ASMs. However, the integration of the SMP kinetics modelling into ASMs has complicated their structure by including new processes, state variables and model parameters (Zuthi et al., 2013). Thus making their use un-adequate in real plants if not accurately calibrated and validated with real data. In this context the assessment of the uncertainty may improve the calibration process. With this aim the sensitivity and uncertainty analysis could help modeller to identify the key source affecting model outputs (Sweetapple et al., 2013).

In order to detail the fouling and pollutants removal modelling in MBR plant, in this work a mathematical model has been presented. The mathematical model has been applied to a sequential batch (SB) MBR pilot plant fed with real saline wastewater. The model has been calibrated by adopting a specific protocol (Mannina et al., 2011). A long-term data base, acquired during an extensive gathering campaign, was adopted for the model calibration. Uncertainty analysis has also been performed.

2. Materials and methods

2.1. The mathematical model

The proposed model couples the ASM1 model (Henze et al., 2000) with the SMPs modelling

(formation/degradation of both utilisation associated products and biomass associated products) in order to take into account their influence on membrane fouling.

The mathematical model is divided into two sub-models: a biological sub-model and a physical sub-model. The biological sub-model involves: 16 biological processes (aerobic and anoxic); 19 state variables, which include dissolved N_2O and CO_2 (S_{N2O} and S_{CO2} , respectively) and 68 model factors. In the Appendices A and B, the Gujer Matrix and the process rate equations of the biological model are reported, respectively. According to the Hiatt and Grady (2008) approach the nitrogen removal process is described as a two steps nitrification and four steps denitrification processes. With this regard the autotrophic biomass is modelled as ammonia-oxidising biomass (X_{AOB}) and nitrite oxidising biomass (X_{NOB}). Regarding the denitrification process four corrections factors for the heterotrophic anoxic growth rate have been introduced. Specifically, factors related to the reduction from

 S_{NO3} to S_{NO2} (μ_{g2}), S_{NO2} to S_{NO} (μ_{g3}), S_{NO} to S_{N2O} (μ_{g4}) and S_{N2O} to S_{N2} (μ_{g5}) have been considered. For example, the process rate related to the anoxic growth of heterotrophic biomass on soluble biodegradable organics (S_{S}) reducing S_{NO} to S_{N2O} is reported in Equation 1. As reported in Equation 1 both the switch functions related to the alkalinity (S_{ALK}) and the ammonia (S_{NUL}) have been here inserted

witch functions related to the alkalinity
$$(\underline{S_{ALK}}, S_{ALK})$$
 and the ammonia $(\underline{S_{NH4}}, S_{NH4})$ have been here inserter $K_{ALK} + S_{ALK}$

according to the corrections of the AMS1 as proposed by Hauduc et al. (2011).

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$$\mu_{H} \cdot \eta_{g4} \cdot \left[\frac{K_{O2,H}}{K_{O2,H} + S_{O2}}\right] \cdot \left[\frac{S_{NO}}{K_{NO} + S_{NO} + \frac{S^{2}_{NO}}{K_{I4NO}}}\right] \cdot \left[\frac{S_{S}}{K_{S} + S_{S}}\right] \cdot \left[\frac{S_{NH4}}{K_{NH4,H} + S_{NH4}}\right] \cdot \left[\frac{S_{ALK}}{K_{ALK} + S_{ALK}}\right] \cdot \left[\frac{K_{I3NO}}{K_{I3NO} + S_{NO}}\right] \cdot X_{H}$$
(1)

In order to model the SMP formation/degradation the aerobic and anoxic hydrolysis processes related both to the UAP and BAP have been added in the ASM1 (see Appendix B). With this aim two state variables have been added (S_{UAP} and S_{BAP}). The S_{BAP} production proportional to the biomass decay coefficient f_{BAP} (fraction of S_{BAP} generated per biomass decayed). The S_{BAP} reduction comprises first-order kinetics that are based on the hydrolysis rate coefficient $k_{H,BAP}$. The rate of the anoxic hydrolysis of S_{BAP} is provided in Equation 2. The S_{UAP} formation/degradation processes occur similarly to that of S_{BAP} .

$$k_{h,BAP} \cdot \eta_{NO3,HYD} \cdot \left[\frac{K_{O2,HYD}}{K_{O2,HYD} + S_{O2}} \right] \cdot \left[\frac{S_{NO3}}{K_{NO3,HYD} + S_{NO3}} \right] \cdot S_{BAP} \cdot X_H$$
(2)

The biological model takes into account the influence of the salinity both for the autotrophic and heterotrophic biomass according to Park and Marchland (2006). More precisely, the maximum growth rate of both autotrophic and heterotrophic biomass has been reduced of the Is coefficient. This latter coefficient has been evaluated according to the Equation 2.

$$I_{s} = \frac{I_{s}^{*}(\% NaCl)}{0.01 + \% NaCl}$$
(3)

Where I*s represent the inhibition factor evaluated and %NaCl is the percentage of salinity expressed as NaCl content.

The physical sub-model simulates the main physical processes that occur in the MBR which are influenced by or may influence the biological sub-model. The physical sub-model involves 6 model factors. Specifically, several processes are taken into account: cake layer formation during suction and back- washing phases; COD removal throughout cake layer which acts as a filter; COD removal due to physical membrane; pore fouling; pore blocking; and influence of SMP on pore fouling. The membrane is modeled by dividing its surface into N equal fractions (areal sections) according to the sectional approach method (Li and Wang, 2006). A different shear intensity of the fluid turbulence (G) is considered as a function of the distance from the aeration systems. Both reversible and irreversible fouling is modeled. More specifically, irreversible fouling is modeled as the sum of two contributes: pore fouling, which is caused by the deposition of solutes inside the membrane pores, and stable cake fouling which is caused by deposition of particles on the membrane surface not removed by backwashing. The deposition of solutes inside the pore is carefully taken into account, as it can be crucial for assessing SMP concentration inside the MBR tank and eventually the membrane fouling. Reversible fouling is modeled as dynamic cake fouling caused by deposition of particles removed during backwashing phase. For a detailed description of the physical sub-model reader is referred to the literature (Mannina and Cosenza, 2013).

2.2. The case study

An SB-MBR pilot plant consisted of two reactors in-series, one anoxic (volume 45 L) and one aerobic (volume 224 L), according to a pre-denitrification scheme (Figure 1) was monitored for almost three months. The pilot plant was equipped with an hollow fiber membrane module (Zenon Zeewed, ZW10) installed into a separate aerated compartment (volume 50 L) for the solid liquid separation. An oxygen depletion reactor (ODR) was placed in the recycling line in order to ensure anoxic conditions inside the anoxic reactor despite the intensive aeration in the aerobic tank. The aerobic, anoxic and MBR reactors were equipped with specific covers that guaranteed the gas accumulation in the headspace.

The SB-MBR pilot plant was discontinuously fed with real domestic wastewater (stored in a feeding tank of 320 L volume) according to fill-draw-batch operation approach. More in detail, 40 L of wastewater (V_{IN}) (previously mixed inside the mixing tank with salt, in order to meet the design salinity concentration) were cyclically fed in, whereas the permeate was extracted at 20 L h⁻¹ (Q_{OUT}).



Figure 1. Layout of the SB-MBR pilot plant (where VIN = 40 L = influent wastewater volume; ODR = Oxygen Depletion Reactor; MBR = membrane Bioreactor; QRAS = 80 L h-1 = recycled sludge from MBR to ODR; QR1 = 80 L h-1 = sludge feeding from aerobic tank to MBR; QOUT = 20 L h-1 (only during the MBR filtration phase = effluent flow rate)

Each cycle had the duration of 3 hours that were split into 1 hour of biological reaction and 2 hours of MBR filtration. During the biological reaction time the permeate extraction pump was turned out, thus Q_{OUT} was equal to zero. During the cycle, 80 L h⁻¹ (Q_{R1}) were continuously pumped from the aerobic to the MBR tank. Furthermore, a recycling activate sludge stream (Q_{RAS}), equal to 80 L h⁻¹ during the reaction period and to 60 L h⁻¹ (Q_{R1} - Q_{OUT}) during the filtration phase, was recycled from the MBR to the anoxic tank via the ODR tank. The experimental campaign was divided into six phases each characterized by a specific salt concentration from 0 up to 10 g NaCl L⁻¹. The NaCl concentration in the influent was increased at step of 2 g NaCl L⁻¹ on a weekly basis. The Phase VI had a duration of 26 days. During plant operations, the influent wastewater, the mixed liquor inside the anoxic and aerobic tank and the effluent permeate have been sampled and analyzed for total and volatile suspended solids (TSS and VSS), total chemical oxygen demand (COD_{TOT}), supernatant COD (COD_{SUP}), ammonium nitrogen (NH₄-N), nitrite nitrogen (NO₂-N), nitrate nitrogen (NO₃-N), total nitrogen (TN), total carbon (TC) and inert carbon (IC). Further, transmembrane pressure (TMP) [bar] data were achieved by means of an analogic data logger every 1 minute. Moreover, instantaneous permeate flow rate ($Q_{OUT,i}$) were measured also every day in order to evaluate the total membrane resistance R_T [m⁻¹] according to Equation 4.

$$R_T = \frac{TMP}{\left(Q_{OUT,i} / A\right) \cdot \mu} \tag{4}$$

Where: A $[m^2]$ represents the membrane surface, μ [Pa s] is the permeate viscosity; the unit of the TMP is Pascal [Pa], Q_{OUT,i} is expressed as cubic meter per second $[m^3 s^{-1}]$.

2.3. Calibration protocol

Model calibration has been performed by adopting the calibration protocol as proposed by Mannina et al. (2011). After a first trial and error calibration, the aforementioned protocol takes into account the selection of model factors of being calibrated for the model outputs of interest by using a sensitivity analysis and later the model factors calibration on the basis of the measured data.

2.4. Sensitivity analysis

In this study sensitivity analysis has been performed by adopting a global sensitivity method (GSA). More precisely, the standardized regression coefficient (SRC) method has been adopted to select important model factors (Saltelli et al., 2004). The SRC method consists of a Monte Carlo simulation (with random sampling of the model factors) and a multivariate linear regression between the model output and the considered model factors.

The absolute value of the standardized regression slopes of the regression (SRC or β i) represents the measure of sensitivity. The sign of β i indicates if the model factor "i" has positive (+) or negative (-) influence on the considered model output. The β i represents a valid measure of sensitivity when the coefficient of determination (R²) is greater than 0.7, as suggested by Saltelli et al. (2004). However, when compared results of the SRC method with other more sophisticated GSA methods (e.g. Extended-FAST) literature studies have demonstrated that SRC method can be adopted to select important model factors even at lower R² value (Cosenza et al., 2013). In the case of a linear model, R² is equal to 1, and the SRC method can be applied to select important and non-influential model factors. Conversely, when the R² is less than 1, the model factors

interact, and the SRC method does not provide any information about the interacting factors. the SRC method can be applied to non linear models only in terms of the selection of important model factors. To apply the SRC method, at least 500 and 1000 simulations are required as suggested in the literature (Cosenza et al., 2013).

2.5. Model parameter calibration

The protocol takes into account the adoption of the generalized likelihood uncertainty estimation (GLUE) methodology (Beven and Binley, 1992); based on Monte Carlo simulations: a large number of model parameter sets are generated from the multidimensional parameter space, each with random parameter values selected from uniform probability distributions for each parameter in order to explore the whole confidence region. The acceptability of each set is assessed by comparing predicted to observed data throughout a chosen likelihood measure/efficiency. In this study the same likelihood measure as adopted by Mannina et al. (2011) was used.

Regarding the uncertainty analysis, non important parameter are fixed to their default or trial and error calibration value. Further, only the model factors classified as important are considered to be uncertain and varied in the uncertainty range according to a random sampling. The results of the Monte Carlo simulations were interpreted by evaluating the trend related to the 5th and 95th percentiles of the model outputs.

3. Results and discussion

3.1. Model application and numerical settings

Simulations were run using input time series employed on the basis of the measured data according to the pilot plant feeding operation. Simulation period has the duration of 84 days. Four different sections of the SB-MBR plant were considered, in particular, the anoxic tank (section 1), aerobic tank (section 2), MBR tank (section 3) and permeate tank (section 4). In order to apply the SRC method model outputs are defined as the average values of the 84 days of simulated time series. Fifteen model outputs of the biological sub-model were taken into account for the GSA: COD_{TOT} for all the four sections; COD_{SUP} for sections 1, 2, and 3; S_{NO3} for sections 1, 3, and 4; ammonia (S_{NH4}) for sections 3 and 4; total nitrogen (TN) for the section 4; total suspended solids (X_{TSS}) for sections 1 and 2. Further, one model output of the physical sub-model was also considered: membrane total resistance (R_T). To apply SRC method 1200 model simulations have been performed. According to the literature suggestion, a threshold value of 0.1 has been chosen for the absolute value of β_i to discriminate between important and non influential model factors (Cosenza et al., 2013).

Uncertainty bands have been performed by employing 1000 Monte Carlo runs by varying only the most important model factors for all the model outputs taken into account. Likelihood distributions for each simulation time step and for each model output were then used for calculating uncertainty bands (5% percentile and 95% percentile of the 1000 runs for each model outputs).

3.2. Sensitivity analysis

The application of the SRC method has provided for each model output taken into account an R^2 value around 0.7. Despite this value is outside the range of applicability of SRC, previous studies have demonstrated that for complex environmental models (such as that under study) reliable results can also be obtained even if R^2 is lower than 0.7 (Cosenza et al., 2013). By applying the SRC method 20 model factors have been selected to

be important at least for one of the sixteen model output taken into account.

Figure 2 summarizes the results of the SRC method application for five (COD_{TOT,1}, S_{NO3,1}, COD_{SUP,2}, S_{NO3,2} and R_T) of the sixteen model output taken into account. By analyzing data reported in Figure 2 one can observe that the model factors mostly affecting the model output COD_{TOT,1} and COD_{SUP,2} is μ_{H} . Indeed, the β_i value of μ_{H} for both COD_{TOT.1} and COD_{SUP.2} is equal to 1; having a positive influence. Indeed, with the increasing of the maximum growth rate of heterotrophic bacteria the increase of the particle fraction of COD takes place. Further, COD_{TOT,1} is also affected by η_{g3} and η_{g4} which respectively control the rate of the heterotrophic anoxic growth when S_{NO2} (nitrite) is reduced to S_{NO} (η_{g3}) and S_{NO} (η_{g4}) is reduced into S_{N2O} . The model output $S_{NO3,1}$ is mostly influenced by the half saturation coefficients for free ammonia (K_{FA}) for nitrous oxide-nitrogen (K_{N2O}). Such a results is mainly due to the fact that this coefficients control the amount of nitrate that can be produced inside the aerobic tank and consequently recycled inside the anoxic one. Similarly, µAUT.NOB and IN.Ss influence the amount of nitrate that can be produced inside the aerobic tank and consequently $S_{NO3,1}$ (Figure 2b). Indeed, $\mu_{AUT,NOB}$ is the most important model factor for S_{NO3,2} (Figure 2d). In terms of resistance, the set of important model factors i: f_{UAP} (fraction of S_{UAP} generated in biomass decay), K_{H,UAP} (hydrolysis rate coefficient for S_{UAP}), λ (screening parameter) and C_E (efficiency of backwashing) (Figure 2e). Among these factors f_{UAP} and K_{H,UAP} are related to the biological sub-model; f_{UAP} and K_{H,UAP} positively influence R_T due to the fact that with their increase, the increase of the SUAP production takes place, thus influencing the membrane fouling. Such a result has paramount interest because suggests that by optimizing biological processes in order to reduce the SMP



Figure 2. Results of the important model factors for $COD_{TOT,1}$ (a), $S_{NO3,1}$ (b), $COD_{SUP,2}$ (c), $S_{NO3,2}$ (d) and R_T (e). production a substantial reduction of the membrane resistance (which means a reduction of operational costs)

can occur. Model factors λ and C_E are directly connected with the physical sub-model. The negative influence of C_E is due to the fact that with the increase of the backwashing efficiency the amount of the cake layer deposited on the membrane surface decreases thus reducing the TMP value at fixed permeate flux.

3.3. Model calibration

Model calibration have been performed by varying all the important model factors selected during the sensitivity analysis. All the other model factors have been fixed at their default value or at the value obtained during the initial trial and error calibration as suggested in the protocol of Mannina et al (2011).

The model calibration has been performed by comparing simulated data with measured data acquired during the sampling campaign. Simulations which provided model efficiency greater than 0.2 were selected as behavioral. The selection of the calibrated parameter values have been performed on the basis of the maximum model efficiency value.

Table 1 summarizes the results of the model calibration on the basis of the efficiency obtained for each model output of the biological sub-model. By analyzing data of Table 1 one can observe that acceptable efficiency were obtained for the model outputs of sections 1, 2 and 4. Indeed, as reported in Table 1 the average value 0.42 was obtained for the efficiencies of the model outputs related to the section 1; 0.41 for the model outputs of the section 2; 0.33 for the model outputs of the section 4. Conversely, the low efficiency values were obtained for the model outputs of the section 3 (0.28 on average). Such a result is mainly debited to the lower number of measured data for the section 3 with respect to the other sections. For the R_T a quite high efficiency value (0.79) was obtained. Thus underlying the excel ability of the model to reproduce the membrane fouling mechanisms.

Section 1	Anoxic tank			
Model output	COD _{TOT,1}	COD _{SUP,1}	X _{TSS,1}	S _{NO3,1}
Efficiency	0.42	0.52	0.31	0.54
n° data	14	14	16	17
Section 2	Aerobic tank			
Model output	COD _{TOT,2}	COD _{SUP,2}	$X_{TSS,2}$	
Efficiency	0.36	0.52	0.36	
n° data	14	14	14	
Section 3	MBR tank			
Model output	COD _{TOT,3}	COD _{SUP,3}	S _{NH4,3}	S _{NO3,3}
Efficiency	0.25	0.29	0.31	0.28
n° data	8	8	8	8
Section 4	Permeate			
Model output	$COD_{TOT,4}$	S _{NH4,4}	$S_{NO3,4}$	TN _{,4}
Efficiency	0.35	0.34	0.36	0.3
n° data	15	17	17	12

Table 1. Results of the model calibration in terms of efficiency related to each model output

3.4. Calibrated results and uncertainty bands

Figure 3 shows the result of the calibrated model in terms of measured data, calibrated modelled trends and uncertainty bands (5% and 95% band).



Figure 3. Results of the uncertainty analysis for COD_{TOT} in the anoxic (a), aerobic tank (b) and permeate (e), COD_{SUP} in the MBR (c), S_{NO3} in the MBR (d) and RT (f).

By analysing data reported in Figure 3 one can observe that the uncertainty band width (as average

difference between 95% and 5% uncertainty band value) changes with the model outputs in the different plant sections (e.g., greater for $COD_{TOT,1}$, $COD_{SUP,3}$ and $COD_{TOT,4}$) (Figure 3 a-e).Such a result is mainly due to the fact that some model outputs entail different level of complexity in terms of involved phenomena in the different plant sections. Indeed, for example the variation of the total COD involves the combination of the variation of different state variables of the model: X_S, X_i, X_H, X_{AOB}, X_{NOB}. Similarly, the variation of supernatant COD involves the variation of S_{BAP}, S_{UAP}, S_I, S_S. Moreover, the band width of model outputs $COD_{TOT,1}$, $COD_{SUP,3}$ and $COD_{TOT,4}$ is greater than others because an higher number of the model factors for which the uncertainty has been studied (important model factors) was important for these model outputs. Indeed, as reported in Figure 3f the band width of the model output R_T is very narrow due to the high accuracy of the model in reproducing the membrane fouling and due to the low number of model factors that resulted to be important for R_T. Note that during the period between the day 62nd and 78th technical failure of the TMP acquisition system occurred, thus the measured value were deeply erroneous (Figure 3f).

Globally, the measured data lays inside the uncertainty band. However, for $COD_{TOT,4}$ a significant proportion of the measured data fall near or on the extremes of the uncertainty bands. Such a fact confirms even more the importance in the quantification of the model uncertainty. Indeed, the quantification of the uncertainty pointed out that the model structure has to be improved in order to provide a better reproduction of the simulated phenomena.

4. Conclusions

The main conclusions deduced from this study are:

- ✓ Model factors affecting the UAP production strongly influence the model resistances coupled with the backwashing efficiency.
- ✓ The calibrated model is able to reproduce in an excellent way the physical processes occurring inside the modelled systems.
- ✓ For the biological model outputs the calibrated model shows a good adaptation between modelled and measured data for the case an high number of measured data is available.
- ✓ Model uncertainty has shown the possibility to improve the model structure to improve the reproduction of some phenomena involved in the modelling of the permeate total COD.

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