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## Effect of sludge characteristics on optimal required dosage of flux enhancer in anaerobic membrane bioreactors

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### ABSTRACT

Fouling is a major challenge for the economic feasibility and applicability of membrane bioreactors. Several strategies have been proposed for fouling control, and among them, addition of flux enhancers (FE) have shown promising results. Previous research revealed a high variability of the required optimal dosage of FE for different types of sludge; however, studies analysing the causes for such variability are lacking. Therefore, this research examines the effect of sludge characteristics on the optimal FE dosage required for sludge filterability improvement ( $D_{opt}$ ). We applied the cationic polymer Adifloc KD451 as FE, and determined  $D_{opt}$  with short-term cross-flow filtration tests. Sludge samples were obtained from five different pilot and full-scale anaerobic membrane bioreactors (AnMBRs) treating municipal or industrial wastewater. Results showed that  $D_{opt}$  was significantly correlated with colloidal organic matter concentration, sludge filterability, capillary suction time, and concentration of soluble polysaccharides. Furthermore, we derived empirical models to predict  $D_{opt}$  based on the significantly correlated sludge characteristics as input variables. This research identifies the factors affecting the required dosage of FE for different sludges and provides guidelines for safe continuous dosing of FE in AnMBR for fouling control.

### 1. Introduction

Anaerobic membrane bioreactor (AnMBR) technology is increasingly researched for wastewater treatment in a circular economy solution to recover nutrients and to produce reclaimed water and biogas. Nevertheless, membrane fouling is recognised as the major challenge limiting the economic feasibility and applicability of AnMBRs. Researchers extensively studied the causes and mitigation of membrane fouling in both aerobic and anaerobic membrane bioreactors [1–4]. Membrane fouling control strategies have focused on optimisation of membrane operational variables, such as: gas sparging, filtration-relaxation cycle and chemical cleaning. Although optimisation of operational variables is suitable when the sludge has good or moderate filtration quality, it may not be adequate or sufficient when fouling is caused by a sludge with poor filterability [5]. Previous works, mostly applied high shear stress near the membrane surface to mitigate fouling [6]. Nevertheless, high shear stress promotes the production of extracellular polymeric substances, floc breakage and detachment of soluble and colloidal material [1]. Reports have shown that soluble and colloidal organic matter plays

an important role in fouling due to the promotion of gel and cake layer formation, pore-blocking and adsorption into the membrane pores [1,7,8].

The application of flux enhancers (FE) for fouling control have been extensively investigated. FE are adsorbents, coagulants and flocculants that decrease fouling by changing the sludge characteristics, thereby improving sludge filterability. For example, some FE reduce the concentration of colloidal and soluble organic matter by charge neutralisation, sweep coagulation, bridging mechanisms or patch flocculation [9]. Previous studies used inorganic coagulants as FE such as poly-aluminium chloride,  $FeCl_3$ ,  $Al_2(SO_4)_3$  and polymeric ferric sulphate. However, without pH correction, these coagulants decrease pH when increasing their dosage, whereas organic polymers and adsorbents do not affect pH [9,10]. Powder activated carbon (PAC) is the most widely applied adsorbent for fouling control. Despite extensive research on PAC applied as FE, this might not be the most appropriate chemical to dose to an AnMBR because the PAC with adsorbed sludge has reduced adsorbent capacity and needs regular replacement [11]; consequently, the sludge adsorbed to the PAC is lost during replacement. Avoiding sludge loss is

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crucial in anaerobic processes because the biomass has a low growth rate. Moreover, Iversen [12] reported adverse effects of the residuals from chemical activation of PAC on biomass activity. Therefore, inorganic coagulants and adsorbents might not be the most promising FE, whereas organic polymers have shown to be advantageous.

The most commonly used organic polymers for fouling control are cationic polymers such as polyDADMAC and polyacrylamide based [13]. Previous studies have successfully applied commercially available cationic polymers from Nalco (MPE50 and MPE30) and Adipap (Adifloc KD452) for fouling control in municipal [14,15] and industrial [16,17] pilot and full-scale (aerobic) MBRs. However, to date, there are only four publications that apply cationic polymers to anaerobic sludge, and all at lab-scale [10,18–20]. So far, to our knowledge, there are no reported studies of cationic polymers addition at pilot or full-scale AnMBRs. Researchers have determined the optimal required dosage of different cationic polymers in a variety of sludge samples and obtained significantly different results. For aerobic sludge samples, reported dosages of Adiflocs range from 10 [21] to 70 mg L<sup>-1</sup> [15,22], and of MPE50 from 100 [23] to 600 mg L<sup>-1</sup> [16]. For anaerobic sludge, dosages of MPE50 from 300 to 1500 mg L<sup>-1</sup> have been applied. Wozniak (2010) has recommended to evaluate weekly the optimal dosage for a continuous application of FE to an MBR, to adapt the dosage to the variable conditions. Díaz et al. (2014) determined the optimal dosage of MPE50 based on specific resistance to filtration (SRF) measurements for disintegrated anaerobic granules (SRF = 2.2 × 10<sup>14</sup> m kg<sup>-1</sup>) and for sludge from a lab-scale AnMBR (SRF = 5.2 × 10<sup>13</sup> m kg<sup>-1</sup>), the optimal dosages were 0.3 g L<sup>-1</sup> and 1.5 g L<sup>-1</sup>, respectively. Braguglia et al. (2006) determined the optimal dosage of Praestol 644, which is a cationic polymer of high molecular weight and low charge density, during biodegradability tests of waste activated sludge. The latter authors obtained different values for the optimal dosage during the biodegradation test due to an increase in protein concentration in the soluble fraction. Despite the variability of the optimal dosage required for different sludge samples that has been reported in literature, there are no studies analysing the cause for such variability.

Moreover, several researchers have observed an adverse effect at FE dosages higher than the optimal dosage on different response variables [10,16,18,20,22,23], such as filtration performance. Furthermore, when overdosing FE, a fraction of FE remains unbound in the bulk liquid and can cause intense fouling by FE adsorption on the membrane surface and inside the pores [24] or potentially contaminate the permeate by passing through the membrane. Therefore, knowing the optimal dosage is crucial to avoid (accidental) deterioration of the filtration performance and permeate quality.

The required FE dosage would be ideally determined by adding FE directly to membrane bioreactors and quantifying fouling. However, this approach is not feasible because the FE at high dosages could have adverse effects on the reactor filtration performance, possibly causing irreversible fouling, and affecting biological activity [25]. Thus, assessment of the fouling potential of sludge samples is a more

appropriate method to determine optimal dosage. In previous research, optimal dosages were mostly determined based on soluble or colloidal organic matter removal and dead-end filtration measurements: time-to-filter, SRF, capillary suction time (CST) and volume of filtrate collected after a defined filtration time. However, cross-flow filtration mode methods, as mostly applied in full-scale membrane bioreactors, are expected to represent more accurately the fouling mechanisms occurring in membrane bioreactors than dead-end filtration methods. Particularly, the anaerobic Delft filtration characterisation method (AnDFCm) [26] is a cross-flow method to determine the filtration properties of sludge samples, defined as sludge filterability. The AnDFCm uses a dedicated set-up that comprises a tubular side-stream ultrafiltration membrane, combined with a well-defined measurement and cleaning protocol, allowing comparison of the filtration properties of different sludge samples.

The goal of this research is to study the effect of sludge characteristics on the optimal required dosage of FE for sludge filterability improvement ( $D_{opt}$ ) and to provide guidelines for dosing FE to AnMBRs.  $D_{opt}$  was determined based on sludge filterability measurements applying the AnDFCm, on seven sludge samples collected from five pilot and full-scale AnMBRs treating municipal or industrial wastewater.

## 2. Materials and methods

### 2.1. Sludge samples and flux enhancer

We selected Adifloc KD451 (Adipap SA, France) as FE based on its capacity to improve sludge filterability with small dosages [27]. Adifloc KD451 is a cationic polymer with low molecular weight and high charge density. The concentration of the FE stock solution was 30 g L<sup>-1</sup>, and it was prepared by mixing the polymer with demineralised water for at least 2 h, and prepared one day before use to provide enough time for chain opening.

Sludge samples were collected from five different AnMBRs, as summarised in Table 1. Sludges BWa, BWb and BWc were collected from one AnMBR at different operational periods, as follows. The AnMBR was spiked (pulse-addition) with the FE Adifloc KD451; BWa and BWb were collected before and three weeks after FE addition, respectively. BWc was collected one year after BWb, assuming that the FE had been removed with the excess sludge. When samples BWa and BWb were collected, the AnMBR was operated without sludge withdrawal, implying an infinite sludge retention time (SRT), whereas when BWc was collected, 30 L d<sup>-1</sup> of sludge was withdrawn, resulting in an SRT of 70–80 days.

### 2.2. Sludge characterisation

#### 2.2.1. Analytical methods

Chemical oxygen demand (COD), total phosphorus (TP), total nitrogen (TN) and ammonium-nitrogen (NH<sub>4</sub>-N) were measured with

**Table 1**  
Sludge samples details.

Sludge name	Reactor (configuration, membrane surface area)	Fed	Location	Time between sludge collection and testing (d)
LiqOFMSW	Pilot-scale AnMBR (sidestream, 20.5 m <sup>2</sup> )	Liquid fraction of digestate of OFMSW <sup>b</sup>	Spain	7
BWa, BWb, BWc <sup>a</sup>	Pilot-scale AnMBR (submerged, 6.25 m <sup>2</sup> )	Source separated blackwater	Spain	0 <sup>c</sup>
Food1	Full-scale AnMBR (sidestream, 1452 m <sup>2</sup> )	Wastewater from confectionery factory	the Netherlands	0.5
Food2	Full-scale AnMBR (sidestream, 1584 m <sup>2</sup> )	Wastewater and waste from confectionery and pet food factory	Poland	7, 8
Food3	Full-scale AnMBR (sidestream, 3432 m <sup>2</sup> )	Whey and wash water	South Africa	14, 15

<sup>a</sup> BWa, BWb and BWc were collected from one AnMBR at different operational conditions.

<sup>b</sup> OFMSW is the organic fraction of municipal solid waste.

<sup>c</sup> Tests performed at the AnMBR site immediately after sampling.

Hach Lange test kits. Because no oxidised N compounds were present, the total organic nitrogen (TON) was calculated as the difference between TN and  $\text{NH}_4\text{-N}$ . COD was measured in different fractions of the sample, obtained by consecutive filtration steps [10]. The COD measured in the unfiltered sample was designated as total COD (tCOD). The submicron COD (csCOD), which includes both colloidal and soluble COD, and soluble COD (sCOD) were measured in fractions below  $1\ \mu\text{m}$  and  $0.45\ \mu\text{m}$ , respectively. The supracolloidal COD (scCOD) is the fraction above  $1\ \mu\text{m}$  and it was calculated by subtracting csCOD from tCOD. The colloidal COD (cCOD) is the fraction between  $0.45$  and  $1\ \mu\text{m}$  and was calculated as the difference between csCOD and sCOD. To obtain the csCOD fraction the unfiltered sludge sample was centrifuged at  $6500g$  for 10 min, and the supernatant filtrated through a  $1\ \mu\text{m}$  paper filter Whatman GF/B (GE Healthcare, USA). A second filtration was made through a  $0.45\ \mu\text{m}$  syringe filter CHROMAFIL® Xtra PES-45/25 (MACHEREY-NAGEL, Germany) to obtain the sCOD.

Soluble microbial products (SMP) were measured in the soluble fraction, obtained by centrifuging the sludge sample at  $15,772g$  for 15 min and filtering the supernatant through a  $0.45\ \mu\text{m}$  pore syringe filter. SMP were characterised as polysaccharides (SMP-PS), proteins (SMP-PR) and humic substances (SMP-HS). Polysaccharides were determined with the phenol-sulfuric acid method [28], using D-Glucose monohydrate as standard. Proteins and humic substance were determined with the modified Lowry method [29], using bovine serum albumin and humic acid sodium salt as standards.

Particle size distribution (PSD) was measured in the range  $0.01\text{--}2000\ \mu\text{m}$  with a Microtrac Bluewave diffraction analyser (Malvern Instruments Ltd., UK). This analyser uses a light scattering technique providing results as volume-based PSD and, in samples with multiple particle sizes together, volume-based distributions neglecting the

smallest particles even when they are present in large numbers. For example, assuming spherically shaped particles, the volume distribution of a sample with equal number of particles with diameters of  $1$ ,  $10$  and  $100\ \mu\text{m}$  is  $0.0001\%$ ,  $0.0999\%$  and  $99.9\%$ , respectively. Thus, we assumed that the PSD represents the size of the larger particles, which are the flocs, and not of the submicron particles. We reported PSD as percentiles  $D_{10}$ ,  $D_{50}$  and  $D_{90}$ , where  $D_{50}$  is the median diameter.

Electrical conductivity and pH were measured with a benchtop multi-meter WTW Multi 9620 IDS (Xylem Analytics, Germany). Sludge dewaterability was measured as CST with a Type 304 M CST apparatus and filter paper (Triton Electronics Ltd, UK) at  $21\ ^\circ\text{C}$  following Standard Methods (APHA, 1999). The concentrations of total suspended solids (TSS) and volatile suspended solids (VSS) were measured following Standard Methods (APHA, 1999), applying a  $0.7\ \mu\text{m}$  pore size filter (AP4007000, Merck Millipore Ltd., Ireland).

### 2.2.2. Sludge filterability

Sludge filterability was measured with the AnDFCm in the side-stream ultrafiltration cross-flow filtration unit (AnDFCm installation) shown in Fig. 1. The AnDFCm consists of three steps: (1) filtering water to determine the resistance to water filtration ( $R_{T,\text{water}}$ ), (2) sludge filtration to estimate the additional resistance obtained when  $20\ \text{L}$  of permeate per  $\text{m}^2$  of membrane area are produced ( $\Delta R_{20}$ ), and (3) mechanical membrane cleaning.  $R_{T,\text{water}}$  includes the membrane intrinsic resistance and the irreversible and irrecoverable fouling resistances. Sludge filterability is inversely related to  $\Delta R_{20}$ . Moreover,  $\Delta R_{20}$  values can only be directly compared when the AnDFCm sludge filtration operational conditions (flux and cross-flow velocities) are the same. In this study, two different operational conditions were applied for different sludge samples, which are explicitly shown in the output

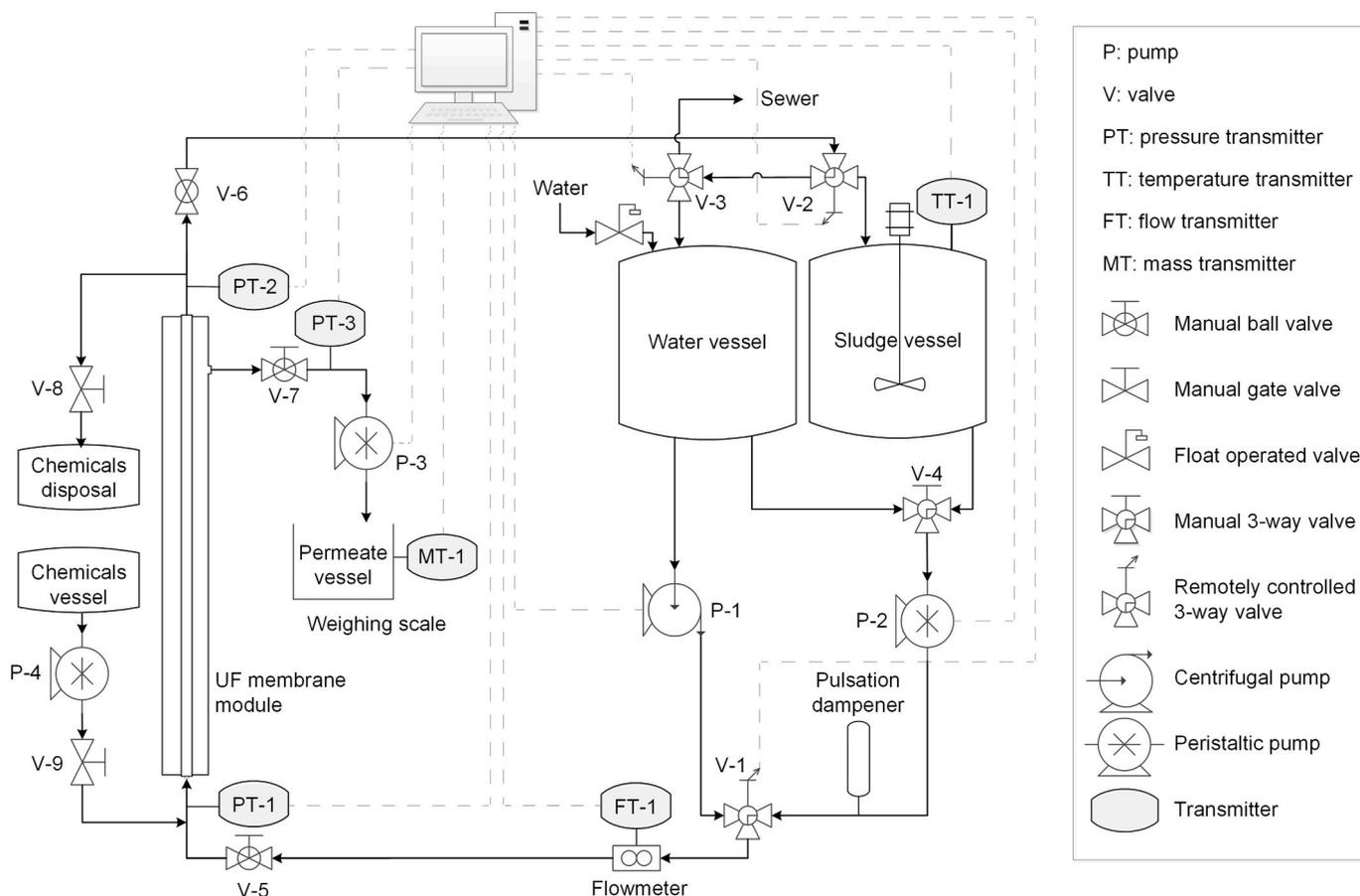


Fig. 1. Scheme of the anaerobic Delft filtration characterisation method (AnDFCm) installation used for sludge filterability assessment. The ultrafiltration (UF) membrane was an X-Flow (Pentair, the Netherlands): tubular,  $30\ \text{nm}$  pore size,  $8\ \text{mm}$  internal diameter, and  $95\ \text{cm}$  length.

measurement. The  $\Delta R_{20(1.5,60)}$  and  $\Delta R_{20(0.5,60)}$  are  $\Delta R_{20}$  measured at 60 L  $m^{-2} h^{-1}$  flux, with 1.5  $m s^{-1}$  and 0.5  $m s^{-1}$  cross-flow velocities, respectively. Moreover, we use  $\Delta R_{20}$  to refer to  $\Delta R_{20(0.5,60)}$  and  $\Delta R_{20(1.5,60)}$  simultaneously.

### 2.3. Flux enhancer dosage-step experiments

Dosage-step tests were performed for all sludge samples shown in Table 1, to determine  $D_{opt}$ . To assess the reproducibility of  $D_{opt}$  determination, the total volume of each sludge, i.e. BwC, Food2 and Food3, was divided in two samples of equal volume prior to performing the dosage-step tests; the resulting samples were further designated as: BwC, BwC-II, Food2, Food2-II, Food3 and Food3-II.  $D_{opt}$  was individually determined in each resulting sample.

The dosage-step test consisted of consecutive stepwise increasing additions of FE into a sludge sample. The protocol, detailed in Fig. 2, was an iterative process that comprised: (1) addition of FE, (2) mixing, (3) sampling for characterisation (PSD, csCOD, sCOD and CST), (4)  $\Delta R_{20(1.5,60)}$  measurement, (5) mechanical cleaning, and (6) determination of resistance to water filtration ( $R_{T,water}$ ); Step 1 to Step 6 were performed consecutively for each dosage. During Step 4,  $\Delta R_{20(1.5,60)}$  was measured for all sludge samples except Food2, where  $\Delta R_{20(0.5,60)}$  was measured instead, because Food2 presented such good filtration quality that no cake build-up was obtained when a cross-flow velocity of 1.5  $m s^{-1}$  was applied.

### 2.4. Data analysis

#### 2.4.1. Selection of optimal and critical dosages

We defined optimal dosage ( $D_{opt}$ ) as the dosage at which filterability improvement reaches a plateau, or saturation, above which no further significant improvement was observed with higher dosages. Critical dosage ( $D_{crit}$ ) is defined as the dosage above which an adverse effect on filterability was observed. Filterability improvement was calculated as  $\Delta R_{20}$  decrease. Additionally, we compared  $D_{opt}$  and  $D_{crit}$  with those obtained with the following alternative sludge characteristics, which are commonly reported in literature: csCOD removal, sCOD removal and CST decrease. From this comparison we verified if csCOD, sCOD or CST could be applied as quick alternative measurements for optimal dosage determination. Appendix A contains further details on dosages selection and calculation.

#### 2.4.2. Correlation and regression analyses

We used Kendall's correlation coefficient ( $\tau$ ) to identify which of the raw sludge characteristics, if any, had a statistically significant relationship with  $D_{opt}$ . Field [30] recommends using  $\tau$ , which is a

non-parametric rank correlation, for small data sets such as the one in our study (10 points). The statistical significance was assessed by comparing the probability value ( $p$ ) with two levels of significance: 0.01 and 0.05.  $\tau$  and  $p$  were computed with *corr* function in Matlab® R2019b.

Afterwards, the sludge characteristics that significantly correlated with  $D_{opt}$  were used to derive empirical models:  $D_{opt} = f(x)$ , where  $x$  is a significantly correlated variable and  $f$  is the model (for example: linear or exponential);  $f$  was selected based on the graphical representation of the results. The models were calibrated in Matlab® R2019b with *fit* function. The goodness-of-fit was assessed based on the coefficient of determination ( $R^2$ ). The prediction bounds (or prediction interval) for new observations were obtained with *fit*, considering all predictor values (simultaneous bounds), and a 95% confidence level [31].

## 3. Results

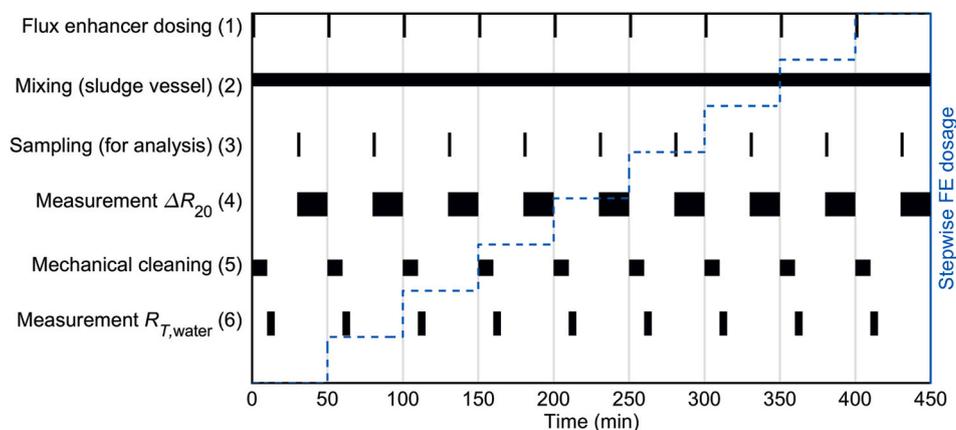
### 3.1. Raw sludge characteristics

The complete characterisation of raw sludges is presented in Table A1 in Appendix A; Table 2 summarises the most relevant characteristics.

The time lapses between collection and testing for LiqOFMSW, Food2, Food2-II, Food3 and Food3-II were sufficiently long to assume that sludge characteristics might have changed due to physicochemical and biochemical processes taking place during transportation and storage, such as degradation of substrates. Therefore, the physicochemical characteristics presented in Table A1 and Table 2 of the abovementioned samples might not be identical to the characteristics of the sludges at the moment of collection from the AnMBRs. Nevertheless, filterability was significantly worse (higher  $\Delta R_{20(1.5,60)}$ ) for LiqOFMSW and Food1 than for the remaining samples, which was in accordance with the deteriorated permeability ( $= \text{Flux}/\text{TMP}$ ) observed by the plant operators at the moment of collection (data not shown).

LiqOFMSW was noticeably different from other sludges, that is: it had considerably higher  $\Delta R_{20(1.5,60)}$ , CST, cCOD, sCOD,  $NH_4-N$ , conductivity, SMP-HS and SMP-PS. Moreover, in LiqOFMSW, SMP-PR was negligible compared to SMP-HS; thus, it was not possible to determine SMP-PR with the modified Lowry method because this method cannot measure proteins independently from humic substances.

the median floc size, represented by the median diameter  $D_{50}$ , was similar for all samples, except Food1. The volume fraction for particles below 1  $\mu m$  was zero in all samples tested (Appendix A). Govoreanu et al. [32] showed that when measuring particle size distribution of activated sludge with a light scattering technique, the distribution of small particles are likely to have errors. Therefore, the values measured below 1  $\mu m$  were likely not realistic. Therefore, we characterised the fraction of organic material below 1  $\mu m$  through cCOD.



**Fig. 2.** Scheme of the flux enhancer dosage-step protocol performed in the AnDFCm installation. The blue-dashed line represents the increasing stepwise FE dosage and the black lines the time frame for each step of the protocol: (1) FE was added to the sludge vessel containing 40 L of sludge, (2) sludge with FE was mixed (during the entire experiment), (3) after 30 min sludge was sampled for characterisation, (4)  $\Delta R_{20(1.5,60)}$  was measured, (5) the membrane was mechanically cleaned by forward-flushing with water at approximately 3  $m s^{-1}$  cross-flow velocity for 10 min, (6) resistance to water filtration ( $R_{T,water}$ ) was measured at 1.5  $m s^{-1}$  cross-flow velocity and 60 L  $m^{-2} h^{-1}$  flux, and (7) Step 1 to Step 6 were repeated for the next dosage. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

**Table 2**

Most relevant characteristics of raw sludges, Appendix A contains the complete characterisation.

Variable	Unit	LiqOFMSW	BWa	BWb	BWc	BWc-II	Food1	Food2	Food2-II	Food3	Food3-II
$\Delta R_{20(1.5,60)}$	$\times 10^{12} \text{ m}^{-1}$	30.0	13.2	6.5	3.1	3.4	24.8	3.0	0.6	14.2	15.3
CST	S	3712	ND	76	64	72	1458	204	204	393	497
SCOD	$\text{g L}^{-1}$	4.36	0.14	0.10	0.28	0.28	ND	0.11	0.10	0.35	0.44
CCOD	$\text{g L}^{-1}$	6.86	0.76	0.37	0.24	0.21	ND	0.14	0.21	1.12	0.95
Conductivity	$\text{S m}^{-1}$	3.0	0.21	0.24	0.11	0.11	ND	0.45	0.45	0.42	0.42
TSS	$\text{g L}^{-1}$	12.4	8.6	8.7	4.2	3.9	17.2	14.4	17.8	9.5	10.7
SMP-PR	$\text{mg L}^{-1}$	NA <sup>a</sup>	50	15	74	71	140	0	0	109	117
SMP-HS	$\text{mg L}^{-1}$	5164	80	71	89	83	47	32	33	0	0
SMP-PS	$\text{mg L}^{-1}$	339	14	7	19	19	62	16	16	20	22
D50	$\mu\text{m}$	21	19	27	22	23	61	21	21	27	28

Abbreviations: ND, not determined; NA, not applicable.

<sup>a</sup> SMP-PR could not be determined because SMP-HS was too high and the sample had to be diluted 20 times, limiting the meaningfulness of the result.

### 3.2. Flux enhancer dosage-step experiments

Fig. 3 shows the effect of increasing FE dosage on  $\Delta R_{20}$  obtained during the dosage-step experiments for each sludge sample. The FE improved filterability showing a saturation behaviour for all sludges. The required dosage of FE to improve filterability was noticeably different for each sludge: the ones with better filterability required less FE. Furthermore, high dosages caused an adverse effect, i.e. increased  $\Delta R_{20}$ , in LiqOFMSW, BWc, BWc-II, Food1, Food2 and Food3; for a more detailed visualisation see Appendix A, Figure A1. The dosages above which this adverse effect was observed, i.e.  $D_{\text{crit}}$ , varied from 0.10 to 2.5  $\text{g L}^{-1}$ . Moreover, the observed  $\Delta R_{20}$  decrease that was achieved for LiqOFMSW (52%) and Food1 (82%) was smaller than for the other samples (>90%). Therefore, sludges with worse filterability do not only require more FE, but also present a more limited improvement on filterability by FE addition.

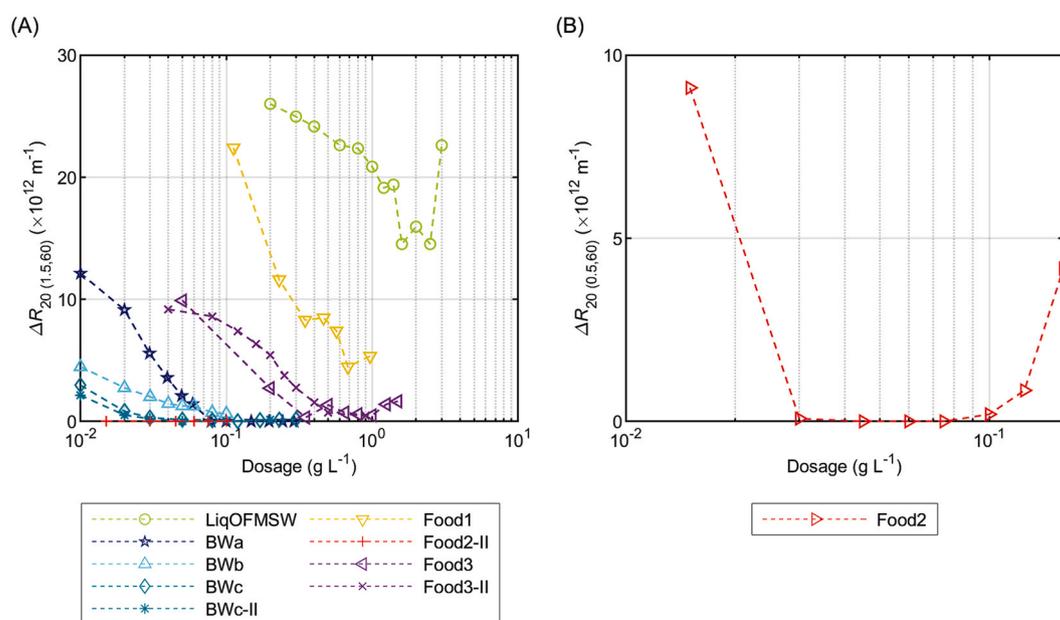
During dosage-step tests, we collected samples and analysed them after each FE addition, this is Step 3 in Fig. 2. Fig. 4 shows that FE decreased csCOD, sCOD and CST, and increased D50. Comparison between Figs. 4A and 3 shows that for most sludge samples, except for LiqOFMSW, the csCOD and  $\Delta R_{20}$  had a similar behaviour, that is: the addition of FE decreased csCOD with a saturation behaviour and had an adverse effect at high dosages, which was observed in BWc, BWc-II, and Food3. Moreover, Fig. 4B shows that sCOD presented a behaviour

similar to  $\Delta R_{20}$  and csCOD, which was characterised as decrease-saturation-increase. Nevertheless, in Food3, the sCOD increased at a lower dosage (0.5  $\text{g L}^{-1}$ ) than the csCOD and  $\Delta R_{20}$  (1.0  $\text{g L}^{-1}$ ).

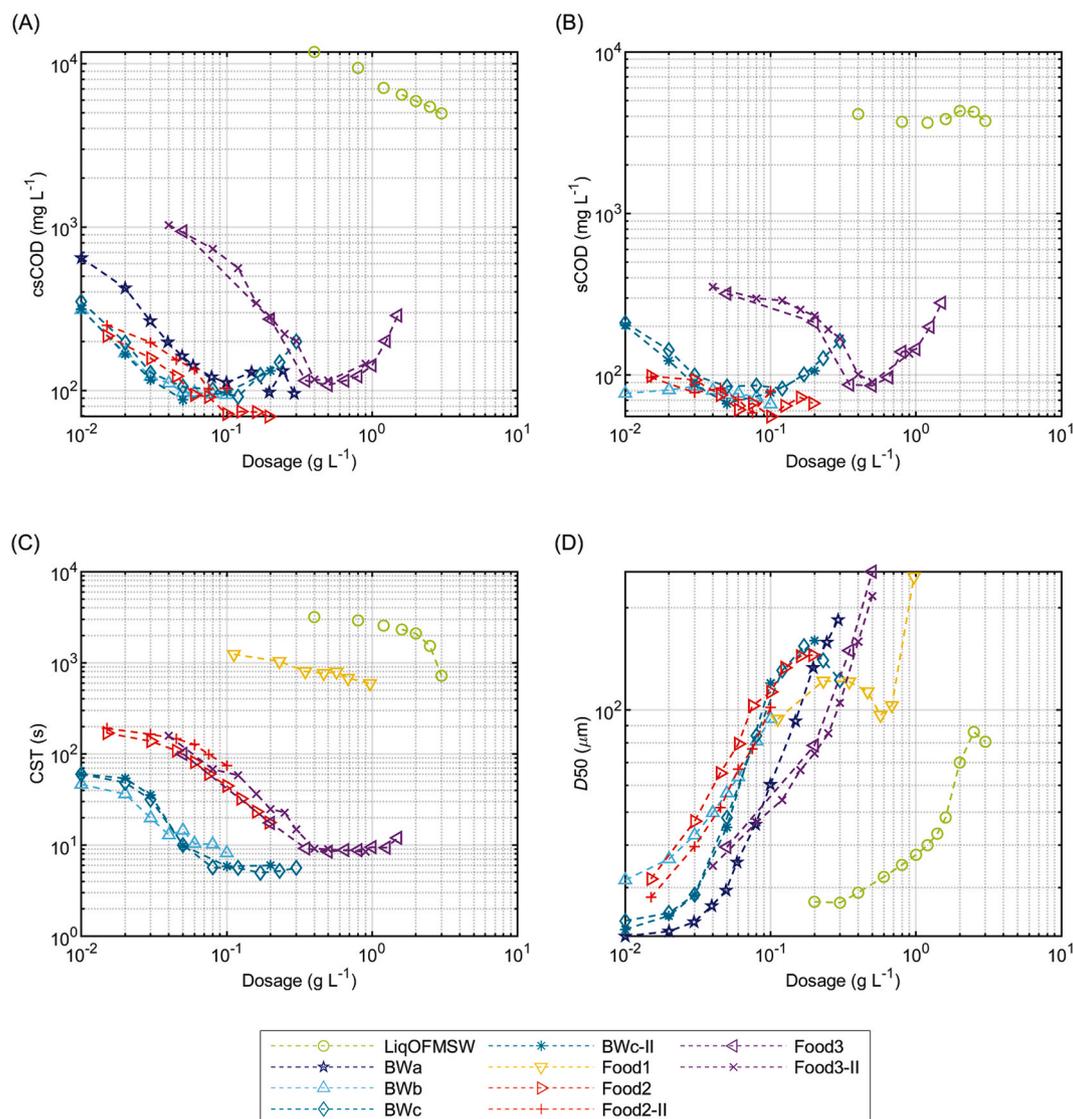
CST is a dead-end filtration method used to assess sludge dewaterability and has been previously used as an indicator of sludge filterability. In our study, CST (Fig. 4C) presented a similar saturation behaviour as  $\Delta R_{20}$  (Fig. 3) for most sludges tested. However, the adverse effect on  $\Delta R_{20}$  and csCOD at high dosages, was not reflected in the CST measurements.

In the FE dosage ranges tested for each sample, the D50 versus dosage curve displayed the saturation behaviour, observed in  $\Delta R_{20}$ , CST and csCOD, only in Food1. Moreover, the adverse effect at high dosages was only present in LiqOFMSW and BWc (Fig. 4D). Therefore, floc size, as quantified in this research, might not be a direct indication of filterability improvement and deterioration.

$R_{\text{T,water}}$  was measured in each cycle of the dosage-step test, after filtering sludge with different dosages of FE and cleaning the membrane, see protocol in Fig. 2. In Fig. 5, the  $R_{\text{T,water}}$  increased after filtering sludges with high FE dosages. For most sludges, the dosage above which  $R_{\text{T,water}}$  rapidly increased was similar to  $D_{\text{crit}}$  (Table 3) of each sludge sample. However, in Food3-II,  $R_{\text{T,water}}$  rapidly increased at dosages above 0.08  $\text{g L}^{-1}$  while no adverse effect was observed on  $\Delta R_{20}$  in the whole range tested, 0.04–0.90  $\text{g L}^{-1}$ . The membrane was chemically cleaned after measuring Food3-II with 0.20  $\text{g L}^{-1}$  of FE, causing a slight



**Fig. 3.**  $\Delta R_{20}$  during FE dosage-step test in the AnDFCm installation for sludge samples from different sources.  $\Delta R_{20}$  was measured by applying a flux of  $60 \text{ L m}^{-2} \text{ h}^{-1}$  and cross-flow velocities of (A)  $1.5 \text{ m s}^{-1}$  and (B)  $0.5 \text{ m s}^{-1}$ .



**Fig. 4.** Sludge characteristics during FE dosage-step test in the AnDFCm installation for sludge samples from different sources: (A) submicron organic matter concentration, (B) soluble organic matter concentration, (C) capillary suction time, and (D) floc size expressed as median diameter. The csCOD of Food1, CST of BWA, and sCOD of BWA and Food1 were not measured.

decrease in  $R_{T,water}$  at the next dosage step ( $0.25 \text{ g L}^{-1}$  of FE). Surprisingly, at dosages above  $0.25 \text{ g L}^{-1}$ ,  $R_{T,water}$  continuously decreased with increasing dosages. We could not explain this observation and the cause should be further investigated. Nevertheless, this behaviour was not observed in other sludges, including Food3 that originated from the same sample as Food3-II.

### 3.3. Optimal and critical dosages

The optimal and critical dosages are presented in Table 3. The dosages differed considerably between samples; particularly  $D_{opt}$  ranged from  $0.02$  to  $1.16 \text{ g L}^{-1}$  and  $D_{crit}$  from  $0.10$  to  $2.5 \text{ g L}^{-1}$ , which were determined based on  $\Delta R_{20}$  decrease.

We compared  $D_{opt}$  with  $D_{opt,csCOD}$ ,  $D_{opt,sCOD}$  and  $D_{opt,CST}$ , that were determined with csCOD, sCOD and CST, respectively. The average relative differences of  $D_{opt,csCOD}$ ,  $D_{opt,sCOD}$  and  $D_{opt,CST}$  with  $D_{opt}$  were 47%, 52% and 78%, respectively; where the relative difference is the absolute value of the difference between the correspondent optimal dosage and  $D_{opt}$ , divided by  $D_{opt}$ .

$D_{crit}$  was determined in 6 out of 10 performed dosage-step tests, because for BWA, BWb, Food2-II and Food3-II no adverse effect on

filterability was observed. Apparently, with the latter 4 sludges, the charge-saturation level of the applied FE was not reached and hence  $D_{crit}$  could not be identified. However, for Food2-II and Food3-II the maximum dosages tested were possibly too low to achieve an adverse effect, moreover these maximum dosages were lower than  $D_{crit}$  obtained with Food2 and Food3, respectively. Accordingly, for BWb the filterability improvement was below saturation at the maximum dosage tested ( $0.10 \text{ g L}^{-1}$ ); thus, the  $D_{crit}$  might be above  $0.10 \text{ g L}^{-1}$ . Surprisingly, for BWA the adverse effect was not observed despite applying a dosage 5-folds above  $D_{opt}$ . Therefore, BWA might be the only sludge that did not present an adverse effect at high dosages of FE.

We compared  $D_{crit}$  with their corresponding  $D_{crit,csCOD}$ ,  $D_{crit,sCOD}$  and  $D_{crit,CST}$ , presented in Table 3.  $D_{crit,csCOD}$  was equal to  $D_{crit}$  for 3 sludges (BWc, BWC-II and Food3) and differed for 2 sludges (LiqOFMSW and Food2); whereas  $D_{crit,sCOD}$  was equal to  $D_{crit}$  for 2 sludges (BWc and Food2) and differed for 5 sludges (LiqOFMSW, BWC-II, Food2-II, Food3 and Food3-II). Furthermore,  $D_{crit,CST}$  was not detected in any sample because there was no adverse effect on CST.

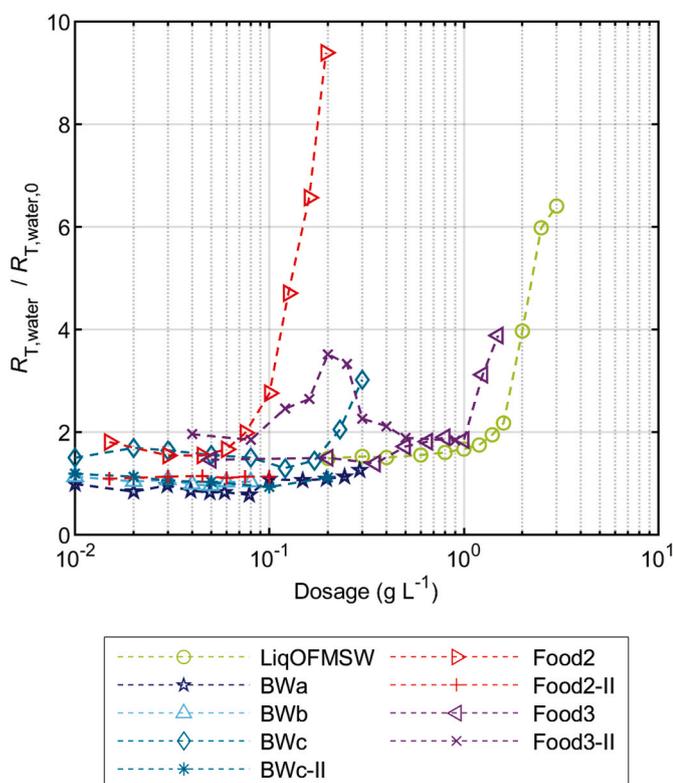


Fig. 5. Resistance to water filtration after each step ( $R_{T,water}$ ) during FE dosage-step test in the AnDFCm installation for sludges from different sources. The  $R_{T,water}$  was normalised by resistance to water filtration before starting each test ( $R_{T,water,0}$ ). For Food3-II, the membrane was chemically cleaned with  $2 \text{ g L}^{-1}$  of citric acid during 1 h between the FE dosages  $0.2 \text{ g L}^{-1}$  and  $0.25 \text{ g L}^{-1}$ .

### 3.4. Correlation analysis

Kendall's correlation (Table 4) revealed that  $D_{opt}$  had a statistically high significant correlation ( $p < 0.01$ ) with:  $\Delta R_{20(1.5,60)}$  ( $\tau = 0.89$ ), CST ( $\tau = 0.79$ ) and cCOD ( $\tau = 0.86$ ). These correlations were all positive ( $\tau > 0$ ), indicating that a sample with higher  $\Delta R_{20(1.5,60)}$  (lower filterability), CST or cCOD required higher FE dosages to mitigate fouling. Moreover,  $D_{opt}$  was statistically significantly correlated with SMP-PS ( $\tau = 0.61$ ,  $p < 0.05$ ), contrary to SMP-PR and SMP-HS. Furthermore,  $D_{opt}$  had a non-significant correlation with  $\text{NH}_4\text{-N}$ , TON, TP, conductivity, pH and  $D_{50}$ . In addition, there was a significant correlation

Table 3

Optimal and critical FE dosages for:  $\Delta R_{20}$  decrease ( $D_{opt}$  and  $D_{crit}$ ), csCOD removal ( $D_{opt,csCOD}$  and  $D_{crit,csCOD}$ ), sCOD removal ( $D_{opt,sCOD}$  and  $D_{crit,sCOD}$ ), and CST decrease ( $D_{opt,CST}$  and  $D_{crit,CST}$ ).

Sludge name	Dosage range (g L <sup>-1</sup> )	$D_{opt}$ (g L <sup>-1</sup> )	$D_{opt,csCOD}$ (g L <sup>-1</sup> )	$D_{opt,sCOD}$ (g L <sup>-1</sup> )	$D_{opt,CST}$ (g L <sup>-1</sup> )	$D_{crit}$ (g L <sup>-1</sup> )	$D_{crit,csCOD}$ (g L <sup>-1</sup> )	$D_{crit,sCOD}$ (g L <sup>-1</sup> )	$D_{crit,CST}$ (g L <sup>-1</sup> )
LiqOFMSW	0.2–3.0	1.16	1.88	0.76	3.00	2.5	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>
BWa	0.01–0.30	0.06	0.03	ND <sup>c</sup>	ND <sup>c</sup>	NA <sup>d</sup>	NA <sup>d</sup>	ND <sup>c</sup>	ND <sup>c</sup>
BWb	0.01–0.10	0.03	0.02	0.09	0.03	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>
BWc	0.01–0.30	0.03	0.02	0.03	0.06	0.12	0.12	0.12	NA <sup>d</sup>
BWc-II	0.01–0.20	0.02	0.02	0.03	0.05	0.10	0.10	0.05	NA <sup>d</sup>
Food1	0.11–1.00	0.38	ND <sup>c</sup>	ND <sup>c</sup>	0.40	0.68	ND <sup>c</sup>	ND <sup>c</sup>	NA <sup>d</sup>
Food2	0.02–0.20	0.03 <sup>a</sup>	0.06	0.06	0.09	0.10	NA <sup>d</sup>	0.10	NA <sup>d</sup>
Food2-II	0.02–0.10	NA <sup>b</sup>	0.06	0.06	0.10	NA <sup>d</sup>	NA <sup>d</sup>	0.08	NA <sup>d</sup>
Food3	0.05–1.50	0.15	0.21	0.30	0.07	1.0	1.0	0.5	NA <sup>d</sup>
Food3-II	0.04–0.90	0.23	0.17	0.29	0.06	NA <sup>d</sup>	NA <sup>d</sup>	0.5	NA <sup>d</sup>

Abbreviations: ND, not determined; NA, not applicable.

<sup>a</sup> Optimal dosage determined using  $\Delta R_{20} = \Delta R_{20(0.5,60)}$  instead of  $\Delta R_{20} = \Delta R_{20(1.5,60)}$ .

<sup>b</sup> Optimal dosage was not estimated because  $\Delta R_{20}$  decrease was 100% immediately after the lower dosage tested.

<sup>c</sup> The corresponding characteristic was not measured during this dosage-step test.

<sup>d</sup> Critical dosage could not be estimated because no adverse effect was observed.

between  $D_{opt}$  and TSS ( $>0.7 \mu\text{m}$ ) but not between  $D_{opt}$  and scCOD ( $<1 \mu\text{m}$ ); the correlation with TSS could be caused by the fraction of colloidal organic matter in the range of 0.7 and  $1 \mu\text{m}$ , which is retained in the filter during the measurement. Consequently, the variables selected for empirical models were  $\Delta R_{20(1.5,60)}$ , CST, cCOD and SMP-PS.

### 3.5. Empirical models

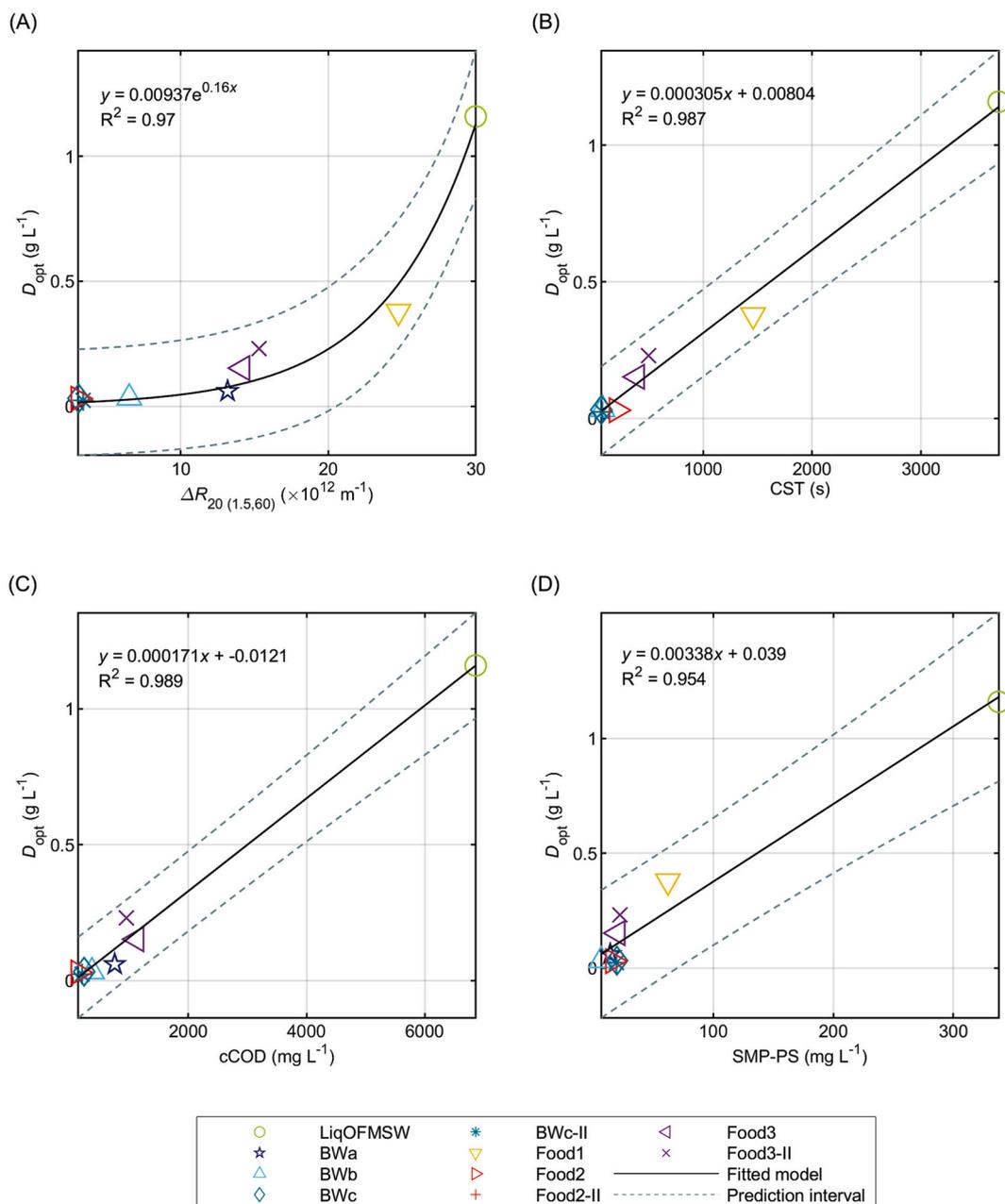
The scatterplots, presented in Appendix A, Figure A6, suggest that the empirical model for  $\Delta R_{20(1.5,60)}$  was exponential:  $D_{opt} = a \exp(b\Delta R_{20(1.5,60)})$ , where  $a$  and  $b$  are parameters; whereas for CST, cCOD and SMP-PS linear models were adopted:  $D_{opt} = ay + b$ , where  $y$  represents CST, cCOD or SMP-PS variables. Fig. 6 displays the experimental data, the empirical models, and the prediction interval for new observations.

LiqOFMSW considerably influenced the calibration of the empirical models. Figure A5 in Appendix A shows the empirical models calibrated excluding LiqOFMSW, and Table A3 compares the predicted dosages with the models calibrated including (Fig. 6) and excluding (Figure A5) LiqOFMSW. The predictions using different empirical models slightly differed. However, all predicted optimal dosages are in the same order of

Table 4

Kendall ( $\tau$ ) correlation coefficient between raw sludge characteristics and optimal dosage of flux enhancer for filterability improvement ( $D_{opt}$ ). Significant correlation at levels 0.01 (\*\*) and 0.05 (\*).

Variable	$\tau$
$\Delta R_{20(1.5,60)}$	0.89**
CST	0.79**
scCOD	0.43
cCOD	0.86**
SCOD	0.57
TP	0.20
TON	0.44
$\text{NH}_4\text{-N}$	0.50
pH	0.31
Conductivity	0.55
TSS	0.56*
SMP-PR	0.62
SMP-HS	-0.08
SMP-PS	0.61*
$D_{10}$	0.22
$D_{50}$	0.28
$D_{90}$	0.17



**Fig. 6.** Experimental data and fitted model of the optimal FE dosage versus raw sludge characteristics: (A) filterability expressed as  $\Delta R_{20(1.5,60)}$ , (B) capillary suction time, (C) concentration of colloidal organic matter, and (D) concentration of soluble polysaccharides. The grey-dashed lines represent the prediction interval for new observations; and  $R^2$  is the coefficient of determination of the fitted model.

magnitude as the measured  $D_{opt}$  and below the measured critical dosages  $D_{crit}$ . Therefore, all models showed their suitability for predicting the dosage required for filterability improvement of new sludge samples, especially as a reference dosage to avoid overdosing. Nevertheless, further research with sludges characterised by a poor filterability could improve the predictive value of the models.

#### 4. Discussion

This research paper analysed the effect of polymer dosage on the various sludge characteristics and water matrix parameters. The terminology ‘optimal required dosage’ or  $D_{opt}$  was introduced for the minimum required polymer dosage to exert a maximum improvement in sludge filterability; a dosage beyond  $D_{opt}$  had only a marginal additional effect. Moreover, a further increase in polymer dosage negatively

impacted the sludge filterability. The dosage at which this occurred was defined as  $D_{crit}$  or the ‘critical flux enhancer dosage’. The applied FE dosages exerted a different impact on the specific sludge characteristics and water matrix parameters of the various investigated sludges. As such, correlations were identified between  $D_{opt}$  and  $D_{crit}$  of the various sludges and some of these characteristics and parameters.

##### 4.1. Effect of sludge characteristics on optimal FE dosage

###### 4.1.1. Colloidal and soluble organic matter

The concentration of soluble and colloidal organic matter play a major role in fouling of membrane bioreactors [1,7,8]; high concentrations increase fouling by pore blocking, cake layer formation, and decrease in cake layer porosity. Electrostatic repulsion between the negatively charged colloidal particles prevents them from

agglomerating, whereas the intermolecular attraction promotes bonding. Cationic polymers promote colloidal agglomeration by bridge flocculation, where polymer segments are adsorbed onto the surface of different colloid particles linking them together. Alternatively, agglomeration is promoted by electrostatic patch mechanisms, where the polymer is adsorbed onto the particle surface creating positively charged patches that interact, upon collision, with the negative surface of other colloidal particles to form flocs. Moreover, excessive cationic polymer dosages can cause particle redispersion, also designated as deflocculation or restabilisation, by saturation of bridging sites or charge reversal due to complete surface coverage [9].

Accordingly, in Fig. 4A, csCOD decreased with increasing dosages of FE with a saturation behaviour which corresponds to complete colloidal surface coverage. The csCOD increased at dosages above  $D_{crit}$  due to redispersion of colloidal particles. The dosage at which complete coverage is attained depends on the colloidal surface area available. Regarding sludge filterability,  $D_{opt}$  was defined as the dosage at which the sludge filterability reaches saturation. Since the available colloidal surface is directly related to cCOD,  $D_{opt}$  was positively correlated with cCOD ( $r = 0.86$ ,  $p < 0.01$ ). Therefore, samples with higher concentrations of colloidal organic matter require higher FE dosages to mitigate fouling.

Soluble organic matter can be positively or negatively charged or can be neutral, namely: the overall charge of a protein depends on the pH and its isoelectric point; polysaccharides can be negatively charged (containing carboxylic groups or sulfuric ester groups), positively charged (with protonated free amino groups) or neutral; humic substances are negatively charged (containing carboxylic and phenolic groups). Therefore, the interaction with the cationic polymer is not straightforward, as it is for the exclusively colloidal organic matter. Consequently, sCOD did not correlate significantly with  $D_{opt}$  ( $p > 0.05$ ).

A linear correlation between the optimum cationic polymer concentration and concentration of humic substances has been reported [9]. In our present research, the SMP-HS was not significantly correlated with  $D_{opt}$  ( $p > 0.05$ ), but the variation of SMP-HS between samples was small (Table 2), with the exception of LiqOFMSW. The high optimal dosage required for LiqOFMSW was possibly due to the high concentration of SMP-HS that bind with the FE, thus decreasing the availability of FE to flocculate the colloidal material.

#### 4.1.2. Supracolloidal organic matter concentration and size

Supracolloidal organic matter in sludge matrices, further referred to as flocs, usually present an overall negative surface charge [20,33]. Thus, cationic FE may adsorb onto the flocs, thereby decreasing FE availability for flocculation of colloidal material. Consequently, bigger quantities of FE would be required at higher flocs concentrations. Nevertheless, colloidal material can be incorporated into flocs by interaction with the FE adsorbed or by entrapment between aggregated flocs. Therefore, flocs present opposite effects on the flocculation of colloidal material, and thus on filterability improvement. Consequently, we did not observe a significant correlation between sCOD and  $D_{opt}$ .

For suspensions with uniform particle size, higher optimal dosages are required for smaller particles due to its larger specific surface. Nevertheless, good flocculation performance can be obtained at different dosages for samples with various particles sizes [9]. Therefore, in our samples, which had flocs of a wide range of sizes (~1–1000  $\mu\text{m}$ , Appendix A), there was no significant correlation between the  $D_{opt}$  and the floc size distribution, represented by  $D_{10}$ ,  $D_{50}$  and  $D_{90}$  in Table 4.

#### 4.2. pH and electrical conductivity

When using coagulants as FE, pH is a crucial variable. However, cationic polymers can be used within a wide pH range, without the need for pH adjustment. Research has shown an effect of pH on the optimal required dosage, which was mainly attributed to changes in the surface

charge of the material to be flocculated [9]. Contrarily, we did not observe a relationship between pH and  $D_{opt}$ ; however, the pH range of the samples was narrow: 6.9–8.1 (Table 2). Similar pH values are observed in most membrane bioreactors where approximately neutral pH is required to favour biological processes. Therefore, pH is not expected to influence the required dosage of cationic polymer.

Electrical conductivity is correlated with ionic strength and it is a fast measurement to estimate the concentration of ions in solution. High ionic strength could positively influence aggregation of colloidal material by reducing the electrostatic repulsion of particles of the same charge. However, it might also harm flocculation by reducing the extension of the polymer chains; the size of polymer chains are very important for effectively bridging colloidal material [9]. Additionally, the adsorption of FE onto the negatively charged material can be reduced by polymer neutralisation by anions, or by decreased available adsorption sites due to adsorption of cations onto the colloidal material [34]. Therefore, ionic strength has opposite effects on flocculation of colloidal material. Accordingly, we did not observe a correlation between conductivity and  $D_{opt}$ .

#### 4.3. Side effects of FE and the risk of overdosing

Dosages of FE above the  $D_{crit}$  caused an adverse effect on filterability, csCOD and sCOD in several sludges. Various researchers have observed this negative effect on different response variables [10,16,18,20,22,23]. Excessive FE dosage can impair filtration performance due to increased concentration of colloidal and soluble organic matter by deflocculation. Additionally, the interaction between the cationic FE, that remains unbound in the bulk liquid, and the membrane may result in intense fouling, by FE adsorption over the membrane surface and inside the pores [24]. This was shown, in Fig. 5, by high  $R_{T,water}$  after filtering sludge with excessive dosages of FE. Additionally, in Appendix A, Section A3, we studied the effect of unbounded FE on membrane filtration by filtering demineralised water with different concentrations of Adifloc KD451. The FE deteriorated the filtration performance, measured as an increase in  $R_T$ , upon contact with the membrane but only at high concentrations, which are unlikely to be present in AnMBRs dosed with FE.

The high  $R_{T,water}$  measured after filtering sludge with excessive dosages of FE, suggests that the excess polymer caused irreversible fouling, which refers to the fouling that needs to be removed by chemical cleaning. Irreversible fouling increases the frequency of chemical cleaning, decreasing the lifespan of the membranes. Therefore, it is crucial to avoid overdosing FE to the AnMBR.

Furthermore, we observed that a fraction of the unbounded Adifloc KD451 passed through the membrane and contaminated the permeate (results shown in Appendix A, Section A3). Research had shown that PolyDADMAC, which is the chemical compound in Adifloc KD451, can be a precursor of *N*-nitrosodimethylamine (NDMA), which is a suspected human carcinogen produced during chloramination or ozonation [35, 36]. Therefore, determining the  $D_{opt}$  and  $D_{crit}$  for each sludge individually is crucial to avoid adverse effects on filterability and permeate contamination by unbounded FE.

Comparison of optimal and critical FE dosages determined with different response variables.

The optimal and critical dosages determined in terms of filterability improvement ( $\Delta R_{20}$  decrease) were compared with those obtained based on csCOD removal, sCOD removal and CST decrease. The CST showed the worst results:  $D_{opt,CST}$  was most different from  $D_{opt}$ , and CST decrease proved unable to detect critical dosages ( $D_{crit,CST}$ ). For most samples  $D_{opt,CST}$  was higher than  $D_{opt}$ , probably because the FE increases sCOD which deteriorates dewaterability, here measured as CST, but does not have a direct effect on filterability. Thus, more FE is needed to compensate for the increased formation of sCOD increasing CST. Furthermore, during dead-end filtration, which is the principle of CST measurements, a sludge cake is deposited on top of the filter and can act

as a first filtration step. At high FE dosages, exceeding saturation, this sludge cake might adsorb both the unbounded FE and the positively charged organic matter saturated with FE, and thereby protecting the filter. Such protective layer is not likely to be critical in cross-flow filtration mode systems, such as in the AnDFCm installation and membrane bioreactors, due to the shearing effect of the cross flow. Therefore, high FE dosages caused a detrimental effect on filterability and not in CST. Consequently, required FE dosages which are assessed using CST measurements may lead to overdosing, with possible adverse effects in fouling and permeate quality.

Better results, compared to CST decrease, were obtained with sCOD removal as response variable: the difference with  $D_{opt}$  was lower and the adverse effect for determining  $D_{crit,sCOD}$  was observed in several sludges. Nevertheless, the  $D_{crit,sCOD}$  was only equal to  $D_{crit}$  in 3 samples (and differed in 5), and  $D_{opt,sCOD}$  was generally higher than  $D_{opt}$ . This overestimation of the optimal required dosage might be attributed to possibility that part of the soluble material that is removed by the FE remains as colloidal material, which could have a similar adverse effect on filterability as the original soluble material. Similarly, Koseoglu et al. (2008) found that SMP removal, where SMP was measured as SMP-PS plus SMP-PR, and improvement in filtration performance do not always correlate.

The variable csCOD removal exhibited the best results in terms of optimal and critical dosages, likely because csCOD comprised both soluble and colloidal material, which interacted with the FE and affect filterability. However, the difference between  $D_{opt}$  and  $D_{opt,csCOD}$ , which was on average 47%, remained significant.

Therefore, we recommend the use of cross-flow filtration modes, such as the one applied in the AnDFCm installation, to determine the optimal dosage of FE. If such methods are not available, as is the case in most full-scale AnMBR and MBR plants, we recommend the use of csCOD removal as an alternative variable, instead of measurements based on dead-end filtration mode or based on soluble organic matter removal.

#### 4.4. Guidelines for dosing FE in a continuous AnMBR

When using Adifloc KD451 as FE in a new sludge sample,  $D_{opt}$  can be estimated with the empirical models in Fig. 6 by measuring any of the following sludge characteristics:  $\Delta R_{20(1.5,60)}$ , CST, cCOD or SMP-PS. Furthermore, the estimated  $D_{opt}$ , further called  $D_{opt,simulated}$ , could be used to design an experiment to determine  $D_{opt}$  more precisely for each specific sludge. We recommend to use the following dosages in the experiment: 0, 0.2, 0.4, 0.6, 0.8, 1, 1.1, 1.5, 2.2.5 and 3-folds  $D_{opt,simulated}$ , where the triplicates at  $D_{opt,simulated}$  are applied to estimate the variability of the test. After each dosage, the sludge filterability should be determined or, alternatively, the csCOD.

For the application of FE to a full-scale AnMBR, avoiding FE overdose is important since it can cause: i) reversible fouling increase caused by deteriorated sludge filterability, ii) irreversible fouling increase caused by the interaction of unbounded FE with the membrane, and iii) permeate contamination by part of the unbounded FE that can pass through the membrane. Thus,  $D_{crit}$  should be used as a limiting dosage, meaning that the concentration of FE in the reactor should be below  $D_{crit}$  during the entire operational period. Different dosing strategies can be applied, for example: continuously dosing FE to the reactor to sustain a concentration equal to  $D_{opt}$  while compensating for FE losses with the sludge wastage; or in a fed-batch mode, where the FE is dosed as a pulse to achieve  $D_{opt}$ , whenever the sludge has poor filterability, or high cCOD, CST or SMP-PS.

Moreover, the effectiveness of the FE can be largely affected by the degree of mixing during flocculation, which is governed by velocity gradients and time of flocculation [9]. Therefore, when applying FE to a full-scale AnMBR, special attention should be given to the mixing conditions.

The annual cost of dosing FE for fouling control in membrane bioreactors is subject to the volume of mixed liquor, price of FE, optimal required dosage, and frequency of dosage. As an example, we estimated the cost for the AnMBRs where the sludges with highest and lowest  $D_{opt}$  (i.e. LiqOFMSW and BWc-II) were collected. We assumed a dosing strategy where the frequency of dosage is adjusted to compensate for sludge wastage, resulting in a frequency of 1/SRT. The price of the Adifloc KD451 depends on the purchase amount and location. Nevertheless, the price used here was 6 € kg<sup>-1</sup>, which was given by the supplier as a base price. The AnMBR where BWc-II was collected operated at 70 d SRT, 2.35 m<sup>3</sup> mixed liquor volume, and  $D_{opt}$  was 0.02 g L<sup>-1</sup>, and thus the FE cost would be 1.5 € y<sup>-1</sup> or 0.63 € m<sup>-3</sup> y<sup>-1</sup>, which is negligible. However, the AnMBR where LiqOFMSW was collected operated at 8 d SRT, 42 m<sup>3</sup> mixed liquor volume, and  $D_{opt}$  was 1.16 g L<sup>-1</sup>, and thus the FE cost would be 13,337 € y<sup>-1</sup> or 318 € m<sup>-3</sup> y<sup>-1</sup>. Therefore, the costs of FE dosing can vary considerable for different AnMBRs and should be considered in the economic evaluation of each treatment plant.

## 5. Conclusions

The main findings are summarised as follows:

- The optimal and critical dosages differed considerably between the various AnMBR sludge samples:  $D_{opt}$  ranged from 0.02 to 1.16 g L<sup>-1</sup> and  $D_{crit}$  from 0.10 to 2.5 g L<sup>-1</sup>.
- $D_{opt}$  presented a linear relationship with CST ( $R^2 = 0.975$ ), cCOD ( $R^2 = 0.983$ ), and SMP-PS ( $R^2 = 0.936$ ); and  $D_{opt}$  had an exponential relationship with  $\Delta R_{20}$  ( $R^2 = 0.950$ ), which is inversely related to sludge filterability. The empirical models derived can be potentially used to predict  $D_{opt}$  in new sludge samples, or as guidance for experimental design for  $D_{opt}$  determination.
- $D_{opt}$  had non-significant correlations with scCOD, floc size, TP, TON, NH<sub>4</sub>-N, SMP-PR and SMP-HS.
- Excessive FE dosages had an adverse effect on sludge filterability, increased irreversible fouling in the AnDFCm installation and possibly promoted permeate contamination by the unbounded FE. Therefore, overdosing must be avoided when applying FE to full-scale AnMBRs.
- The  $\Delta R_{20}$ , measured following the AnDFCm protocol, was a reliable variable to determine the optimal and critical FE dosages. If the AnDFCm or an alternative cross-flow filtration measurement is not available, csCOD removal could be used as an alternative variable to estimate the optimal FE dosage, since it provided better results than scCOD removal and CST decrease.

## Author contributions

MO designed and performed the experiments, analysed the data and wrote the first draft of the manuscript. ML-F, HS and JvL supervised the research. All authors provided critical feedback and helped shape the research, analysis and manuscript.

## CRediT authorship contribution statement

**Magela Odriozola:** Conceptualization, Methodology, Formal analysis, Investigation, Visualization, Writing - original draft, Writing - review & editing. **Maria Lousada-Ferreira:** Supervision, Project administration, Funding acquisition, Writing - review & editing. **Henri Spanjers:** Supervision, Project administration, Writing - review & editing. **Julius B. van Lier:** Supervision, Funding acquisition, Writing - review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.memsci.2020.118776>.

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