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Coagulation/flocculation prior to low pressure membranes in drinking water treatment: a review

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Microfiltration (MF) and ultrafiltration (UF) consistently remove suspended material and pathogens from drinking water; however, membrane fouling inhibits their application by increasing operation and maintenance costs. Coagulation/flocculation is a commonly used pretreatment method for the reduction of membrane fouling; in this review it has been grouped into three typical configuration types: Type 1: coagulation + no/incidental flocculation, Type 2: coagulation + flocculation, and Type 3: conventional coagulation, based on operational conditions. The impact of each configuration on floc properties, membrane fouling, and organics removal has been reviewed in detail. Due to relatively high membrane resistance and low NOM reductions, configuration Type 1 may not be optimal for fouling control and organics removal when compared to Types 2 and 3. Configuration Type 2 led to the lowest cake layer and specific cake layer resistance for both MF and UF, while there is evidence that Type 3 results in the greatest reduction in fouling rate by reducing mass flux towards the membrane surface. As expected, with no coagulant results indicate that UF achieves greater organics removal when compared to MF, but with the addition of coagulant performance is similar for all configuration types. By highlighting the connection between coagulation/flocculation configuration types and membrane performance, the review provides insight for the design and operation of pretreatment for low pressure membrane filtration. In addition, understanding the impact of configuration types on floc properties aids in revealing the fouling mechanisms that dictate membrane performance. Knowledge gaps have been identified for guidance on future research.

Water impact

Membrane fouling inhibits the application of low pressure membranes by increasing operation and maintenance costs. Coagulation/flocculation is a commonly used pretreatment method to reduce fouling, which may be grouped into three typical configuration types. This review provides insight for the design and operation of coagulation/flocculation by highlighting the impact of each configuration on floc properties, membrane fouling, and organics removal.

1. Introduction

Microfiltration (MF) and ultrafiltration (UF) membranes, commonly referred to as low pressure membranes, are widely employed due to their ability to consistently remove suspended material and pathogens from drinking water.[1] However, membrane fouling remains a challenge, causing

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increased transmembrane pressure (TMP), flux deterioration, and greater frequency of required backwashing and chemical cleaning. Thus fouling reduces operating efficiency and membrane life, and ultimately increases the operating and maintenance costs of membrane filtration.[2] Membrane fouling is typically characterized as reversible or irreversible based on the impacts of cleaning practices. Hydraulically reversible fouling can be addressed hydraulically (*e.g.* backwashing), while hydraulically irreversible fouling can be addressed chemically (*e.g.* chemical cleaning). Chemically irreversible fouling cannot be removed and its gradual increase contributes to membrane 'ageing', or irreversible changes to membrane performance and characteristics associated with long-term foulant and cleaning agent exposure.[3]

Membrane fouling is highly impacted by natural organic matter (NOM).[4] Historically, hydrophobic humic substances (*i.e.* humic and fulvic acids) which constitute the majority of NOM present in surface waters[5] have been identified as the predominant NOM foulants.[6,7] However, there is increasing evidence that hydrophilic biopolymers (*i.e.* protein- and polysaccharide-like macromolecules) are the main contributor to membrane fouling.[8–12] As NOM is ubiquitous in source waters, identification of the NOM fractions responsible for membrane fouling is a primary concern for development of mitigation methods for drinking water treatment using low pressure membranes.

Coagulation/flocculation is commonly used prior to low pressure membrane filtration to reduce fouling, and has been reported to reduce pore blocking, decrease cake layer resistance, and increase backwash efficiency.[13] Previously published reviews regarding pretreatment,[1,14] as well as fouling and cleaning[15] for low pressure membranes, and an overview of coagulation/flocculation pretreatment for membrane treatment of drinking water and wastewater[16] can be found in the literature. Gao[1] summarized pretreatment methods (*e.g.* coagulation, adsorption, peroxidation, prefiltration) and operational conditions (*e.g.* running modes, rinsing modes, chemical cleaning, air scouring) for reducing fouling, while Huang[14] also reviewed pretreatment methods to address membrane fouling concluding that coagulation had been most successful. Shi[15] provided a summary of both conventional and non-conventional (*e.g.* electrical cleaning, ultrasonic) cleaning methods, as well as their impact on membrane materials though no results quantifying cleaning performance were included. Thus, the available review studies do not provide insight on the impact of coagulation/flocculation prior to low pressure membranes on floc properties, membrane fouling, and NOM removal. In particular, no comprehensive review exists on the impact of coagulation/flocculation configuration types on membrane performance.

The present review classified coagulation/flocculation pretreatment configurations from all of the reviewed literature into three typical types: Type 1: coagulation + no/incidental flocculation (*i.e.* coagulation with direct membrane filtration), Type 2: coagulation + flocculation (*i.e.* coagulation, flocculation, with direct membrane filtration), and Type 3: conventional coagulation (*i.e.* coagulation, flocculation, sedimentation, and membrane filtration). The impact of each configuration type on floc properties and membrane performance in terms of fouling and NOM removal was assessed. The number of studies considered in reviewing the impact of configuration type on floc properties, fouling performance, and DOC/TOC removal is summarized in Fig. 1. Published studies that counted towards fouling performance include at least one of total resistance, cake layer/specific cake layer resistance, specific hydraulic resistance, mean rate of TMP increase/flux decline, and flux recovery after hydraulic and chemical cleaning. This review highlights the connection between coagulation/flocculation configuration types and membrane performance, providing insight for the design and operation of pretreatment for low pressure membrane filtration. Summarized results are used to understand the impact of configuration types on floc properties, which dictate fouling mechanisms and membrane performance. Knowledge gaps have also been identified to provide guidance for future research.

2. Coagulation/flocculation-low pressure membranes

2.1. Overview of coagulation/flocculation theory

Coagulation and flocculation typically occur sequentially, governing the formation and properties of floc, and impacting the performance of downstream membranes. Initial aggregation of particles/NOM occurs by destabilization during coagulation, where mechanisms include charge neutralization, interparticle bridging, and sweep flocculation.[17] As particles in natural waters, including NOM, are negatively charged (pH 6.0 to 8.0), charge neutralization occurs when sufficient cationic metal hydroxides are adsorbed to reduce zeta potential to zero.[18,19] Sweep flocculation occurs when the concentration of a coagulant exceeds its solubility limit, precipitates, and enmeshes particles/NOM.[20] The preferred destabilization mechanism for membrane pretreatment is dependent on water quality, and may be different for each configuration type.

Following destabilization, particle-particle interactions (*i.e.* collisions) result in floc formation.[17,19] Fluid shear induced by mixing is the dominant flocculation mechanism when two colliding particles are >1 μm in diameter, while Brownian motion dominates when at least one particle is small (*i.e.* <1 μm in diameter) and differential sedimentation in all other cases.[21] Aggregation rates are highest when particles have been fully destabilized by coagulation, and are lower in the case of partial destabilization. The hydrodynamic conditions of coagulation/flocculation are typically described by mean velocity gradient, $\bar{G}$, and contact time, t, and will be different for each configuration type. In some of the published studies impeller speed (rpm) rather than $\bar{G}$ value is used to describe hydrodynamic conditions; however, despite the limitations of

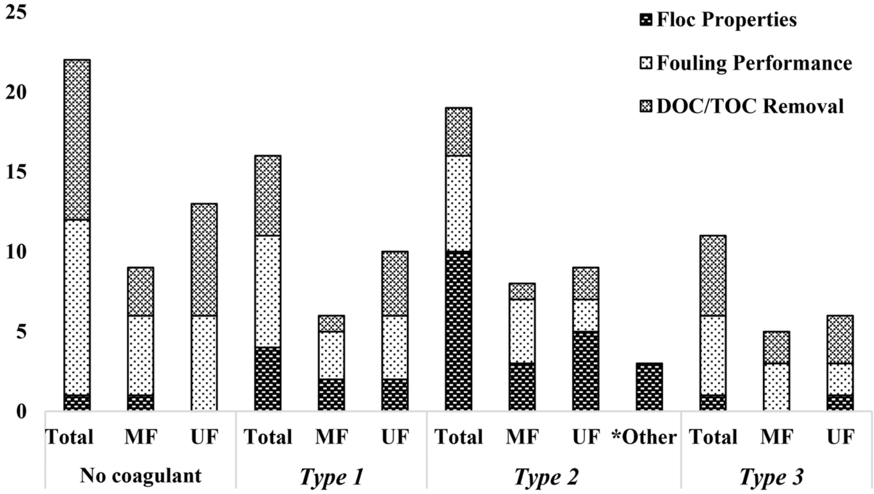

Fig. 1 Number of published studies considered for floc properties, fouling performance, and DOC/TOC removal based on configuration type.* Studies did not include membrane filtration.

$\bar{G}$ value, impeller speed is inadequate for quantifying the forces being applied to water during mixing, and hinders the reproducibility of study results. Finally, mass flux of floc towards the membrane surface results in fouling, where floc properties may be associated with the type of membrane fouling that predominates.

2.2. Typical configuration types

Coagulation/flocculation pretreatment for all of the reviewed literature could be classified into three typical configuration types: Type 1: coagulation + no/incidental flocculation; Type 2: coagulation + flocculation; and Type 3: conventional coagulation (Fig. 2). In this review, coagulation without flocculation/sedimentation is considered as coagulation + no/incidental flocculation. Coagulation followed by flocculation in the absence of sedimentation is commonly referred to as direct filtration,[22,23] while here it is characterized as coagulation + flocculation. Conventional coagulation, which includes sedimentation, has been applied prior to membrane filtration to remove aquatic constituents that cause fouling.[14] However, it has been observed that this may not be effective in removing the NOM fractions which contribute to irreversible fouling,[24] and that similar fouling performance may be realized without sedimentation.[25] In coagulation pretreatment without sedimentation, floc size is only required to grow beyond that of membrane pores (*i.e.* sub-micron), thus reduced coagulant dosages may be applicable.[26] Coagulation pretreatment without flocculation/sedimentation has been investigated for its potential to significantly reduce flocculation times and water treatment plant footprint.[27] A summary of coagulation/flocculation conditions for configuration Types 1, 2, and 3 from the literature is provided in Table 1.

2.2.1. Type 1: coagulation + no/incidental flocculation. For configuration Type 1, a coagulant is typically applied on a continuous basis followed by rapid or static mixing directly upstream of membrane filtration. This does not include a flocculation step, and contact time is typically <1 min,[9,23,27] though it may be slightly longer.[31] Two studies did not apply mixing after the addition of coagulant.[9,29] Incidental flocculation could result from the use of additional ancillary equipment, such as contact tanks[30] and recirculation lines,[32] or by extended contact time such as that which may be experienced in a membrane cell or tank.[33,41,43]

No standardized method for the optimization of coagulant dosage for configuration Type 1 has been presented in the literature. Coagulant dosages that have been considered range from 0.59 to 5.68 mg L^{-1} Al and 0.34 to 10.00 mg L^{-1} Fe. Several studies have examined the impact of coagulant dose on the basis of turbidity and NOM removal by charge neutralization and sweep flocculation.[22,29,30,32] Coagulant dosage has also been optimized for reducing membrane fouling. Pronk[9] reported increased fouling at an $FeCl_3$ dosage of 5 ppm (1.70 mg L^{-1} Fe) compared to 1 and 2 ppm (0.34 and 0.68 mg L^{-1} Fe), while Judd and Hillis[27] observed that at coagulant dosages <0.035 mM Fe^{3+} (1.96 mg L^{-1} Fe) fouling increased when compared to no coagulant addition. These results suggest there may be dose thresholds, above or below which fouling is exacerbated by the application of coagulant. Choi and Dempsey[32] examined the effect of alum and aluminum chlorohydrate (ACH) dosages ranging from 0.59 to 2.93 mg L^{-1} Al on membrane fouling. The authors suggested that low dose conditions (1.17 mg L^{-1} Al, pH 4.81), below those required for charge neutralization, could simultaneously reduce membrane fouling and coagulant costs. Konieczny[30] observed that $FeCl_3$ and $Al_2(SO_4)_3$ doses which were 20% lower than those determined by jar testing (3.0 mg L^{-1} Fe and 3.6 mg L^{-1} Al, respectively) resulted in the lowest drop in permeate flux.

Where mechanical mixing or inline mixing were applied, mean velocity gradient ($\bar{G}$) and contact time (t) after coagulation for Type 1 have been reported to range from 0 to 5.25 s^{-1}, and 360 to 720 s, respectively. Studies that did not

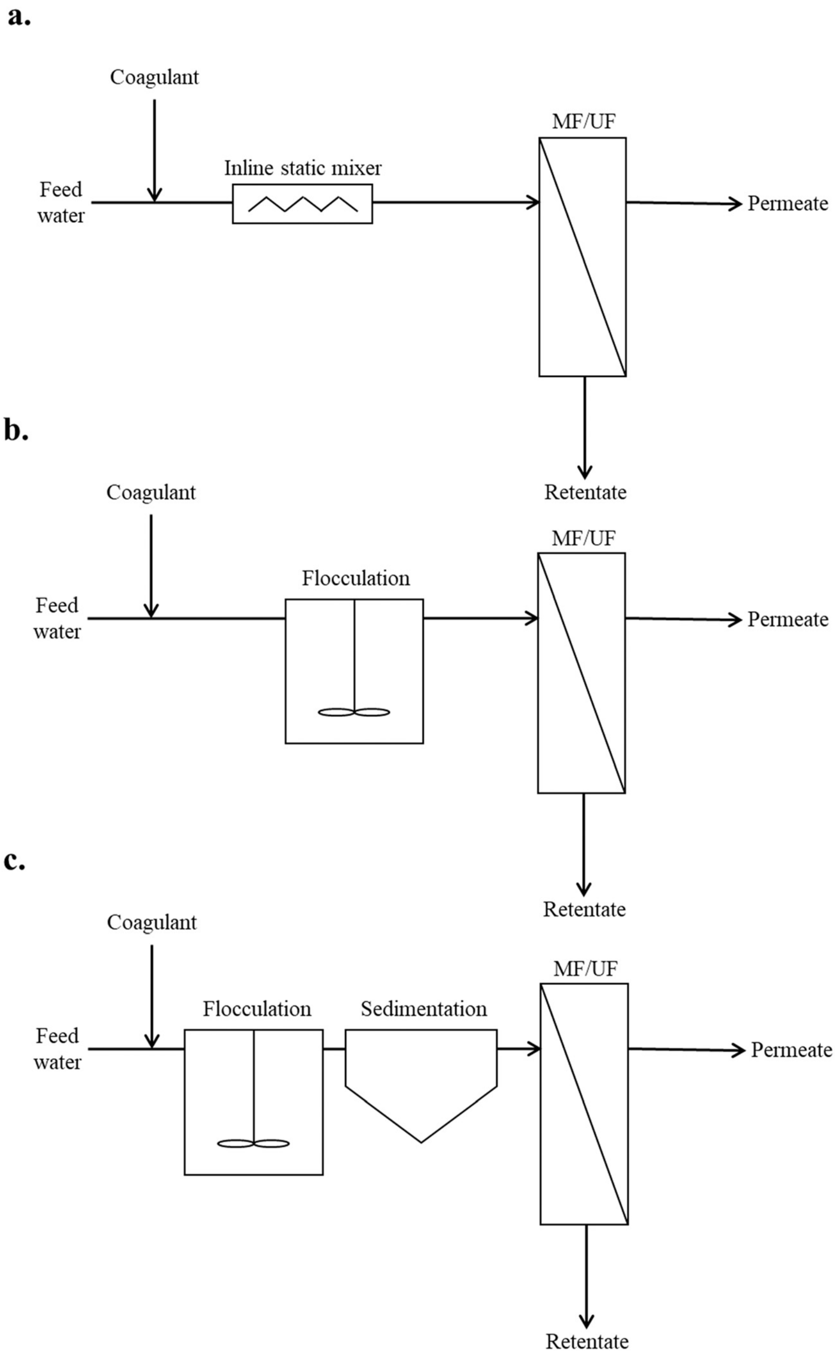

Fig. 2 Typical configuration types: a. Type 1: coagulation + no/incidental flocculation, b. Type 2: coagulation + flocculation, c. Type 3: conventional coagulation.

include mixing did not report a $\bar{G}$ value but contact time ranged from 20 to 120 s. Hydrodynamic conditions for coagulation generally are not well described in studies where static mixing is used.[27] For incidental flocculation conditions, it is difficult to characterize mixing in terms of $\bar{G}$ and t, and where these values are provided justification for their selection is not always stated directly.

2.2.2. Type 2: coagulation + flocculation. Configuration Type 2 includes flocculation, typically performed with mechanical mixing, followed directly by membrane filtration. Coagulant dosages that have been considered range from 6.75×10^{-4} to 39.96 mg L^{-1} Al and 22.00 mg L^{-1} Fe. As for Type 1, numerous studies considered coagulant dosages optimized for turbidity and NOM removal,[24,40,41,43,44] while others investigated a range of coagulant dosages[42] including those below what is considered optimum for NOM removal by sedimentation. Yao[33] observed that while a low coagulant dose led to higher total membrane resistance, when compared to an optimum dosage for humic acid and turbidity removal, irreversible fouling was lower. Wang[42]

Table 1 Summary of coagulation/flocculation conditions for typical configuration types

	Coagulation conditions			Flocculation conditions		Sedimentation	
Configuration	Dose (mg L^{-1} Me^+)	$\bar{G}$ (s^{-1})	t (s)	$\bar{G}$ (s^{-1})	t (s)	t (s)	Ref.
Type 1: coagulation + no/incidental flocculation	0.34–10.00	N/A[a] 100–150	N/A 180	N/A 0–5.25	20–120 360–720	N/A	9, 22, 25, 27–32
Type 2: coagulation + flocculation	6.75×10^{-4}–39.96	100–184	60–180	14.85–80	600–1800	N/A	22, 26, 28, 29, 33–43
Type 3: conventional coagulation	0.04–9.23	100–300 rpm	60–180	5.25–60	360–1740	900–3600	8, 22–24, 39, 44–48

[a] N/A = not available.

examined a range of alum, PACl, and ACH dosages (1.7 to 8.5 mg L^{-1} Al), reporting low (1.7 to 3.4 mg L^{-1}) and high (8.5 mg L^{-1}) coagulant doses to aggravate membrane fouling. Thus, the authors suggested that coagulant dosage for membrane pretreatment be maintained between these low and high dosage thresholds to minimize fouling by particles smaller than the membrane pore size, and to avoid increasing cake layer resistance from the accumulation of coagulant hydrolysis products. Considering coagulant dosages ranging from 1 to 300 μM Al (0.027 to 8.1 mg L^{-1} Al), Ma[26] identified critical concentrations of 5 and 10 μM Al (0.135 and 0.27 mg L^{-1} Al) where membrane flux drastically declined due to particle size. Ding[49] highlighted the importance of charge neutralization of meso-particles (20 nm to 0.5 μm) in minimizing irreversible MF fouling.

For configuration Type 2, $\bar{G}$ and t for coagulation have been reported to range from 100 to 184 s^{-1}, and 60 to 180 s, respectively. Flocculation $\bar{G}$ and t ranged from 14.85 to 80 s^{-1}, and 600 to 1800 s, respectively. Coagulation conditions have frequently been simulated using a jar test, where high mean velocity gradients ($\bar{G}$ ranging from 100 to 150 s^{-1}) were applied for a short duration (t ranging from 60 to 180 s).[22,23,28,41,42] Compared to configuration Type 1, $\bar{G}$ values during flocculation are approximately 3 to 15 times greater and contact times generally longer. Howe and Clark[23] incorporated a shorter contact time (240 s) to promote the development of pin-floc, which are floc with relatively small size when compared to those typically formed to promote sedimentation.

2.2.3. Type 3: conventional coagulation. In addition to flocculation, configuration Type 3 also includes sedimentation, where conditions are optimized for floc growth and the reduction of turbidity or NOM during settling. Coagulant dosages that have been considered range from 0.04 to 9.23 mg L^{-1} Al. Several studies selected coagulant dosage using jar tests, with optimum alum dosages for turbidity and NOM removal ranging from 10 to 15 mg L^{-1} (0.92 to 1.22 mg L^{-1} Al)[23] and 30 to 70 mg L^{-1} (2.76 to 6.44 mg L^{-1} Al),[22,23] respectively. Optimization of coagulant dose for biopolymer removal has also been considered.[8,50,51] Carroll[24] and Kabsch-Korbutowicz[46] used coagulant dosages optimized for NOM removal of 3.20 and 3.59 mg L^{-1} Al, respectively. Howe and Clark[23] examined membrane fouling following Type 3 for five different source waters with alum doses ranging from 0 to 50 mg L^{-1} (0 to 4.50 mg L^{-1} Al) representing low dose, optimum turbidity removal, and enhanced coagulation conditions. Generally, low dose conditions exacerbated fouling, while enhanced coagulation resulted in reductions. At an optimum dosage for turbidity removal, reductions in membrane fouling were observed for two of the source waters, no improvement for one, and increased fouling for the remaining two. The latter three cases coincided with poor NOM removal (<10%) since coagulant dosages were not optimized for this purpose, which was suggested to have contributed to increased fouling. This emphasizes the importance of NOM as a membrane foulant, and also the importance of differences in water quality when considering coagulant dosages for membrane pretreatment.

For configuration Type 3, $\bar{G}$ and t after coagulation have been reported to range from 5.25 to 60 s^{-1}, and 360 to 1740 s, respectively. Mixing intensities for coagulation were not reported in terms of $\bar{G}$, but impeller speeds ranged from 100 to 300 rpm, and t from 60 to 180 s. While the maximum flocculation time for Type 2 was 1800 s (30 min) that for Type 3 is slightly lower, but still expected to be relatively long to enhance floc development for subsequent settling. Settling times prior to membrane filtration ranged from 900 to 3600 s (15 to 60 min).[23,46,47] For Type 3, it was also demonstrated that tapered flocculation, or gradual reductions in mixing speed (65, 40, and 25 rpm for 17 min each), could promote the formation of larger floc size and greater removal of turbidity prior to membranes.[23]

2.3. Operational variations

While the conditions for each configuration described in section 2.2 provide an overview of the main alternatives, there are several operational variations that may be applied in each case. Yu[52] examined the addition of polyacrylamide (PAM) as a coagulant aid to reduce UF fouling, while Xu[53] investigated the use of titanium sulfate ($Ti(SO_4)_2$) and UF for treatment of waters containing algal organic matter. The application of natural coagulants in drinking water treatment, such as chitosan,[54,55] seed extract,[56] and starches,[57] has been investigated in order to address the sustainability of coagulation/flocculation. Encouraging results were obtained with respect to turbidity removal, as well as in reducing membrane fouling when applied in combination with Al-based coagulants. It was suggested that pre-coating metal hydroxides prior to a permeation cycle may reduce fouling and enhance the removal of organic matter by forming a permeable, easily removable fouling layer.[29,58] Pronk[9] suggested establishing a protective fouling layer would result in longer permeation cycle times, and examined the application of phased coagulation as an alternative. By applying 1 to 2 ppm $FeCl_3$ during the first 30 min of a 60 min permeation cycle, the authors observed similar total membrane fouling when compared to continuous coagulant addition. Phased coagulation would have the added benefit of significantly reducing coagulant costs.

3. Impact of configuration types on floc properties

3.1. Summary of results

Coagulation/flocculation, including coagulation conditions (*i.e.* coagulant dose, coagulant type, pH) and hydrodynamic conditions (*i.e.* $\bar{G}$ value, contact time) can impact floc properties such as floc size, growth rate, surface charge, structure, and strength[25,59] (Table 2). As the coagulation/flocculation conditions of each configuration type differ, it is anticipated that floc properties will also vary. Potential relationships between coagulation/flocculation conditions and floc properties for Types 2 and 3 have been highlighted for results compiled from several studies (Table 8 and Fig. 7 and 8). Due to lack of data, similar figures were not included for Type 1.

Floc structure is commonly characterized by fractal dimension, D_f, which may range from 1 to 3.[60] A D_f value of one represents a linear aggregate having a mass proportional to length, whereas a D_f value of three represents a uniform aggregate with mass approximately equal to the size cubed.[61] It has been suggested that as the value of D_f increases the number of particle–particle bonds within the floc also increases along with strength.[62] However, while floc formed under sweep flocculation conditions are compact, they are less dense and may be more susceptible to compression under pressure.[44]

3.2. Type 1: coagulation + no/incidental flocculation

Studies that have considered configuration Type 1 have investigated the impact of coagulant dose, type, and hydrodynamic conditions on floc size, surface charge, and D_f (Table 2). In general, floc sizes reported were smallest (~1 to 8 μm) when compared to other configuration types. The size range reported by Cho[28] (49 ± 5 to 63 ± 5 μm) is an exception since contact time (20 min to 8 h) was extended in an effort to determine the effects of hydrodynamic conditions on floc properties. After 20 s of rapid mixing (230 rpm) and applying low (1.12 to 1.67 mg L^{-1} Fe) and high (4.02 mg L^{-1} Fe) coagulant doses, Judd and Hillis[27] reported particle aggregation into 2 to 5 μm floc of <10% and 65%, respectively. Similar observations were made by Cho[28] following 3 min of rapid mixing where zeta potential was near zero, suggesting that charge properties develop early in the coagulation process.

For Type 1 hydrodynamic conditions, Amjad[22] reported that floc formed at low $\bar{G}t$ (1890) were at least an order of magnitude smaller, and D_f lower when compared to those at higher $\bar{G}t$ values (17 820 and 50 400). Cho[28] observed that D_f decreased over time (2.30 ± 0.02 to 1.92 ± 0.01) as floc structure became less dense. While this was with rapid mixing only, the contact time was relatively long (3 min to 8 h) and it is unknown if similar results would be observed at shorter times. As it has been suggested that floc strength is related to D_f, and since D_f increases with increasing particle collisions,[62] it is expected that floc formed by configuration Type 1 will have relatively low D_f values and may also have low strength.

3.3. Type 2: coagulation + flocculation

Studies that have considered configuration Type 2 examined the effect of coagulant dose, type, pH, and hydrodynamic conditions on floc size, growth rate, D_f, and strength (Table 2) (Fig. 7). The reported floc sizes (10 to 740 μm) generally increase with increasing coagulant dose. Coagulant doses below those required for charge neutralization may lead to insufficient hydrolytic coagulant species and poor floc formation.[33,42] Chakraborti[37] suggested that at a low alum dose (3.33 mg L^{-1}) that there was insufficient coagulant present to cause significant floc growth within the first 10 min (600 s) of flocculation. It has also been reported that at coagulant doses below those optimized for NOM removal by sweep flocculation, D_f values are also lower.[33,42]

With respect to coagulant type, Feng[41] observed that monomeric $AlCl_3$ resulted in larger floc size at pH 6.0 than polymeric coagulants. Similarly, at pH 6.0 and optimum dosages for humic acid removal (7 to 13 mg L^{-1}), Wang[64] reported that floc size was largest for aluminum chloride ($AlCl_3$) followed by polyaluminum chloride (PAC-1) and purified polyaluminum chloride (PAC-2), and that the corresponding growth rates were 0.59, 0.50, and 0.64 μm s^{-1}, respectively. The authors identified a lag time in floc formation where minimal growth occurred over the first 70 to 140 s of flocculation, followed by a growth period from 420 to 665 s. This lag is longer than that reported by Judd and Hillis,[27] where floc growth was observed within 20 s using an Fe-based coagulant. Wang[42] suggested that amorphous aluminum hydroxide ($Al(OH)_3$) produced by alum resulted in the formation of porous floc (*i.e.* low D_f), while aluminum polymers formed clusters and chains of small spheres with higher D_f. At pH 6.0, Feng[41] reported that $AlCl_3$ produced floc with higher D_f than PAC-1 and PAC-2; however, the variance in D_f values was <5%. Dong[43] also reported that monomeric coagulant species form the highest density floc around neutral pH.

The predominant coagulation destabilization mechanism at various pH levels affects floc size, growth rate, and D_f.

Table 2 Effect of coagulation/flocculation configuration types on floc properties

		Coagulation			Flocculation			Floc properties		
Coagulant/dosage	Feedwater	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G} \cdot t$	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G} \cdot t$	Size (μm)	D_f	Ref.
Type 1: coagulation + no/incidental flocculation										
Alum/70 mg L^{-1}	Synthetic (HA) 10 mg L^{-1} TOC pH 4.8–5.5	N/A[a]	N/A	12 600	5.25	360	1890	~1	1.95–2.12	22
Fe-Based/0–0.072 mM (Fe)	Reservoir 2.4 mg L^{-1} TOC pH ~5.4	230 rpm	20	N/A	N/A	N/A	N/A	2–5 (5–65%)[b]	N/A	27
3.18 mg L^{-1} (Al_2O_3)	River 2–3 mg L^{-1} DOC pH N/A	150	20 min–8 h	N/A	0	N/A	N/A	49–63	1.92–2.30	28
PACl/4.1/10.0 ppm (Al_2O_3)	River 2.3–2.9 ppm DOC pH 7.2–7.8	N/A	N/A	N/A	0	720	N/A	12–30	N/A	29
No coagulant	Latex particles 1–16 mg L^{-1} pH 3–12	140	10–220 min	N/A	N/A	N/A	N/A	77–371	2.13–2.64	63
Type 2: coagulation + flocculation										
AC/8 mg L^{-1} (Al) PACb/8 mg L^{-1} (Al) PACc/8 mg L^{-1} (Al)	Synthetic (HA) 5.35 mg L^{-1} DOC pH 4–8	200 rpm	90	N/A	40 rpm	900	N/A	235–295 130–250 130–210	2.509–2.533 2.493–2.534 2.448–2.482	41
$FeCl_3$/22 mg L^{-1} (Fe) PFC_{10}/22 mg L^{-1} (Fe) PFC_{22}/22 mg L^{-1} (Fe)	Synthetic (HA) 4.67 mg L^{-1} DOC pH 4–9	200 rpm	90	N/A	40 rpm	900	N/A	380–740 300–450 225–450	2.54–2.61 2.50–2.60 2.54–2.54	43
Alum/1.7 mg L^{-1} (Al) 3.4–8.5 mg L^{-1} (Al) PACl/2.6 mg L^{-1} (Al) 3.4–8.5 mg L^{-1} (Al) ACH/3.4 mg L^{-1} (Al) 3.4–8.5 mg L^{-1} (Al)	Synthetic (HA) 7.5 mg L^{-1} pH 8.3	100	60	6000	25	1200	30 000	20[c] 35–40 10 60–105 40 55–105	1.70 2.70–2.79 1.80 2.81–2.85 1.90 2.83–2.92	42
Alum/70 mg L^{-1}	Synthetic (HA) 10 mg L^{-1} TOC pH 4.8–5.5	N/A	N/A	12 600	14.85 42.00	1200	17 820 50 400	222 303	2.46–2.73 2.56–2.72	22
PACl/3.18 mg L^{-1} (Al_2O_3)	River 2–3 mg L^{-1} DOC pH N/A	150	3 min	27 000	45	20 min–8 h	N/A	90–447	1.89–2.29	28
PACl/0.025 mM (Al) 0.1 mM (Al)	Synthetic (HA) 5 mg L^{-1} pH 7.5	175	90	15 750	20	900	18 000	120 160	1.81 1.97	33
$AlCl_3$/7 mg L^{-1} PAC-1/13 mg L^{-1} PAC-2/8 mg L^{-1}	Synthetic (HA) 3.56–4.38 mg L^{-1} TOC pH 7.7	200 rpm	30	N/A	40 rpm	600	N/A	410 316 279	2.13–2.27 1.93–2.17 1.88–2.03	64
PACla/0.08 mM (Al) PAClb/0.08 mM (Al) PAClp/0.08 mM (Al)	Synthetic 3.0–3.13 mg L^{-1} DOC pH 7.8–7.9	200 rpm	1 min	N/A	40 rpm	15 min	N/A	375 255 360	2.37 2.35 2.37	34
PACl/4.1/10.0 ppm (Al_2O_3)	River 2.3–2.9 ppm DOC pH 7.2–7.8	N/A	N/A	N/A	58–350[d]	1800-3600	N/A	30–40	N/A	29
Alum/0.1 mmol L^{-1}	Synthetic (kaolin)	200 rpm	60	N/A	40 rpm	1500	N/A	595.7	2.45	35

Table 2 (continued)

Coagulant/dosage	Feedwater	Coagulation $\bar{G}$ (s^{-1})	Coagulation Time (s)	Coagulation $\bar{G}\cdot t$	Flocculation $\bar{G}$ (s^{-1})	Flocculation Time (s)	Flocculation $\bar{G}\cdot t$	Floc properties Size (μm)	Floc properties D_f	Ref.
	50 mg L^{-1} pH 7.50				60 rpm			380.1	2.52	
Alum/0.01 mM (Al)	Synthetic (kaolin)	184	60	11 040	23	1800	41 400	140	1.90–2.17	36
0.02 mM (Al)	57.8 g L^{-1} pH 4.4–7.0							170	1.92–2.42	
Alum/5.33 mg L^{-1}	Latex particles	100	60	6000	20	600	12 000	14	1.83[e]	37
	366 L^{-1}/pH 6.5				80		16 000	14	1.82	
3.33 mg L^{-1}	220 L^{-1}				20		12 000	14	1.91	
					80		16 000	14	1.94	
5.33 mg L^{-1}	220 L^{-1}				20		12 000	18	1.72	
					80		16 000	18	1.75	
3.33 mg L^{-1}	366 L^{-1}				20		12 000	12	1.84	
					80		16 000	16	1.91	
Type 3: conventional coagulation										
Alum/70 mg L^{-1}	Synthetic (HA)	N/A	N/A	12 600	5.25	360	1890	~1[f]	2.06–2.16	22
	10 mg L^{-1} TOC				14.85	1200	17 820	222[f]	2.53–2.71	
	pH 4.8–5.5				42.00	1200	50 400	303[f]	2.42–2.67	

[a] N/A = not available. [b] Particle size reported as percent of particles ranging from 2–5 μm at each coagulant dosage. [c] Given as range of highest volume averages across several dosage concentrations. [d] Lower values in range typical of Type 2, upper values Type 1; not distinguished in study, therefore data only presented for Type 2. [e] Reported as particle size distribution peak. [f] Floc sizes after flocculation, but before settling.

Results indicate that at acidic pH (4.0) larger steady-state floc size can generally be achieved when compared to more neutral or alkaline pH (6.0 to 8.0), while growth rates followed an opposite trend.[41,43] Low growth rates at acidic pH (4.0) were attributed to the predominance of charge neutralization by monomeric coagulant species, while at pH 6.0 and 8.0 monomeric coagulant species were rapidly transformed into polymers and solid precipitates. It was suggested that larger floc formed at lower pH because of charge neutralization and complexation, which involves stronger forces than sweep flocculation. Compared to more neutral and alkaline pH (>6.0), observations indicate that D_f is typically lower at acidic pH (4.0).[36,41,43] The formation of more compact floc at higher pH was attributed to sweep flocculation. Yu[36] applied breakage tests as an indicator of floc strength, where floc were exposed to a high $\bar{G}$ value (184 s^{-1}) and changes in size measured. It was observed that floc with low D_f produced smaller particle sizes (~38 to 58 μm, 41 to 60 μm) than floc with higher D_f (~76 to 102 μm, 130 to 133 μm), suggesting that D_f has a positive correlation with strength. Feng[41] and Dong[43] also reported rapid breakage for floc formed at pH 4.0 when compared to those formed at alkaline pH suggesting lower strength. Results indicate that sweep flocculation produces floc with higher D_f than charge neutralization, and that the D_f of floc formed by monomeric coagulants is higher at acidic pH and lower at alkaline pH when compared to polymeric coagulants.

Previous studies have examined the impact of $\bar{G}$ value and contact time on floc size and D_f. A positive correlation between contact time and floc size has been reported in multiple studies.[28,37] At a $\bar{G}$ value of 45 s^{-1}, Cho[28] observed that floc size increased (90 ± 9 to 447 ± 23 μm) as contact time increased from 20 min to 8 h, while D_f decreased (2.29 ± 0.04 to 1.89 ± 0.01). The observation of a decrease in D_f with contact time is unexpected, and while an explanation was not provided it may be related to the dramatic increase in floc size. Lower $\bar{G}$ values are expected to result in larger floc size,[35,64] while higher $\bar{G}$ values result in greater D_f than floc formed by Type 1.[22,35,37] Floc formed at higher $\bar{G}$ values are expected to be more compact due to increased particle collisions, floc breakup, and restructuring. As floc grow, cluster–cluster interactions become more important and smaller compact clusters have the chance to penetrate the pores of larger flocs.

3.4. Type 3: conventional coagulation

Only one study has considered the impact of configuration Type 3 on floc properties,[22] investigating the effect of hydrodynamic conditions on floc size and D_f (Table 2). After flocculation, floc sizes (~1 to 303 μm) are of similar magnitude as those of Type 2 conditions; however, neither floc size nor D_f were reported after sedimentation. Therefore, the reported floc sizes are expected to be larger than those that would have been present during subsequent membrane filtration, as larger floc would have been removed by settling. Amjad[22] reported that floc with higher D_f values were not preferentially removed by settling, and that the D_f values of floc that were settled and those that were not settled were not statistically different. While high D_f values may indicate more compact floc as mentioned in section 3, it is not necessarily an indication of higher density or floc that are more readily settled.

4. Impact of configuration types on membrane fouling

4.1. Overview of membrane fouling theory

As discussed in section 3, coagulation/flocculation configuration types dictate floc characteristics, which combined with membrane operating conditions, including flux, transmembrane pressure (TMP), and dead-end *vs.* crossflow modes, influence membrane fouling.[33,65,66] Membrane fouling depends on the mass flux of floc to the membrane surface as well as particle adsorption on the membrane surface and in membrane pores. Particle-membrane interactions initially dictate cake layer formation, followed by particle–particle interactions that may become more prominent once a cake layer has formed. Total membrane resistance, $R_{(t)}$, which increases with time, can be quantified using the relationships in eqn (1) and (2):[17]

$$R_{(t)} = R_{\mathrm{m}} + R_{\mathrm{c}(t)} = \frac{\Delta P}{\mu J} \tag{1}$$

$$R_{\mathrm{c}(t)} = R_{\mathrm{rev}(t)} + R_{\mathrm{irr}(t)} \tag{2}$$

where R_{m} is the intrinsic membrane resistance to flow, and $R_{\mathrm{c}(t)}$ is cake layer resistance at time t, ΔP is the TMP, μ is the dynamic viscosity, J is the membrane flux, $R_{\mathrm{rev}(t)}$ is the hydraulically reversible fouling resistance at time t, and $R_{\mathrm{irr}(t)}$ is the hydraulically irreversible fouling resistance at time t. $R_{\mathrm{c}(t)}$ is determined by eqn (3), with the empirical relationship for specific cake layer resistance, α, described by Carmen's equation (eqn (4)):[67]

$$R_{\mathrm{c}(t)} = \alpha M = \alpha J C_{\mathrm{b}} \tag{3}$$

$$\alpha = \frac{180(1-\varepsilon)}{\varepsilon^3 \rho_{\mathrm{p}} {d_{\mathrm{p}}}^2} \tag{4}$$

where M is the mass flux of foulant deposited onto the membrane surface, C_{b} is the bulk concentration of particles, ε is the cake porosity, ρ_{p} is the particle density, and d_{p} is the particle diameter. According to eqn (3), $R_{\mathrm{c}(t)}$ is the resistance due to mass of foulants accumulated on the membrane surface per unit area at time t, while eqn (4) indicates that α is the resistance per unit mass of foulant. Membrane fouling may also be quantified as specific hydraulic resistance, R'_{c}, which is the resistance per unit depth of foulant as expressed in eqn (5).[68]

$$\Delta P_{(t)} = J\mu\left(R_{\mathrm{m}} + \frac{R'_{\mathrm{c}}\,\varnothing_{\mathrm{s}} J}{\varnothing_{\mathrm{c}} - \varnothing_{\mathrm{s}}}\right) \tag{5}$$

where $\Delta P_{(t)}$ is the TMP at time t, $\varnothing_{\mathrm{s}}$ is the volume fraction of solids in the feedwater, and $\varnothing_{\mathrm{c}}$ is the volume fraction deposited in the cake layer. Eqn (5) was applied by Judd and Hillis[27] to quantify membrane fouling for a constant flux-variable pressure system, while Pikkarainen[38] applied it to a constant pressure-variable flux system.

4.2. Summary of results

This section discusses the impact of coagulation/flocculation configuration types on membrane fouling in terms of resistance (*i.e.* resistance to flow), fouling rate, as well as hydraulically and chemically reversible/irreversible fouling. For comparison with configuration types, results for membrane filtration with no coagulant addition have also been included. Published results on the impact of coagulation/flocculation configuration types on membrane resistance (*i.e.* total resistance, cake layer resistance, specific cake layer resistance, specific hydraulic resistance) for MF and UF are summarized in Tables 3 and 4, respectively. Results for membrane resistance are presented graphically in Fig. 3–5 and discussed for each configuration type in sections 4.3 to 4.5. As results are limited and experimental conditions (*e.g.* water matrix, coagulant dose, pH, hydrodynamic conditions) vary across studies, it is difficult to observe trends when comparing the configuration types. Amjad[22] is the only known study to compare the impact of all three configuration types on membrane fouling with similar experimental conditions, highlighting the need for further research to address current knowledge gaps. Potential relationships between coagulant dose and membrane resistance for MF and UF have been highlighted for configuration Types 1, 2, and 3 with results compiled from several studies (Fig. 9–11).

4.3. Type 1: coagulation + no/incidental flocculation

4.3.1. Resistance. Several authors reported a decrease in both total membrane resistance and specific hydraulic resistance with increasing coagulant dose, whereby thresholds were observed above or below which resistance increased[9,27,32] (Tables 3 and 4) (Fig. 9). Judd and Hillis[27] observed that with increasing coagulant dose the percentage of particles in the size range of 2 to 5 μm increased, which would be expected to reduce internal fouling and specific hydraulic resistance. At dosages <0.035 mM Fe (<1.95 mg L^{-1} Fe) formation of floc with diameter less than the membrane pore size resulted in higher specific hydraulic resistance than with no coagulant as well as increased internal fouling. Similar observations were reported by Choi and Dempsey[32] for total membrane resistance at a low coagulant dosage (0.59 mg L^{-1} Al) and alkaline pH (7.47 and 8.73). Applying increasing doses of $\mathrm{FeCl_3}$, Pronk[9] reported approximately equal total membrane resistances ($\sim 7.4 \times 10^{11}$ m^{-1}) during the first permeation cycle, then increasing resistances over the next three cycles due to the formation of a thicker, less permeable cake layer. The same authors observed similar membrane resistance for continuous *vs.* phased coagulation, where 2 ppm (0.68 mg L^{-1} Fe)

Table 3 Effect of coagulation/flocculation configuration types on membrane resistance for microfiltration

			Coagulation			Flocculation			Membrane resistance				
Coagulant/dosage	Feedwater	Membrane type	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G}\cdot t$	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G}\cdot t$	$R_{(t)}$ (m^{-1})	$R_{c(t)}$ (m^{-1})	α (m kg^{-1})	R'_c (m^{-2})	Ref.
Type 1: coagulation + no/incidental flocculation													
No coagulant	Surface water	MF	N/A[a]	N/A	N/A	N/A	N/A	N/A		N/A	N/A	N/A	69
	3 mg L^{-1} DOC	GVHP											
	pH 7.4	0.22 μm											
	Hydrophobic acids	(nominal)							3.20 × 10^{11}				
	Transphilic acids								2.00 × 10^{11}				
	Hydrophilic charged								1.80 × 10^{11}				
	Hydrophilic neutral								5.50 × 10^{11}				
3.18 mg L^{-1} (Al$_2$O$_3$)	River	MF	150	20 min	N/A	N/A	N/A	N/A	N/A	N/A	4.11 × 10^{12}	N/A	28
	2–3 mg L^{-1} DOC	GVWP		1 h							3.65 × 10^{12}		
	pH N/A	0.22 μm		2 h							3.10 × 10^{12}		
		(nominal)		4 h							2.50 × 10^{12}		
				8 h							1.86 × 10^{12}		
No coagulant	Reservoir	MF	230 rpm	180	N/A	N/A	N/A	N/A	N/A	N/A	N/A	2.0 × 10^{17}	27
Fe-Based/0.027 mM (Fe)	2.4 mg L^{-1} TOC	PES										1.5 × 10^{17}	
0.036 mM (Fe)	pH ~5.4	0.1 μm										6.0 × 10^{16}	
0.045 mM (Fe)		(mean)										4.0 × 10^{16}	
0.054 mM (Fe)												3.0 × 10^{16}	
0.072 mM (Fe)												5.0 × 10^{15}	
PACl/2 mg L^{-1} (Al)	River	MF	100	N/A	N/A	N/A	N/A	N/A		N/A	N/A	N/A	70
	1 mg L^{-1} DOC	PVDF							14.1 × 10^{11}				
	0.5 mg L^{-1} DOC	0.1 μm							14.1 × 10^{11}				
	2.0 mg L^{-1} DOC								16.0 × 10^{11}				
	1.6 mg L^{-1} DOC								18.5 × 10^{11}				
	1.0 mg L^{-1} DOC								16.2 × 10^{11}				
No coagulant	Latex particles	N/A	140	10–220 min	N/A	N/A	N/A	N/A	N/A	N/A	0.49–5.07 × 10^{12}[b]	N/A	63
	1–16 mg L^{-1}	Cellulose											
	pH 3–12	0.45 μm											
Type 2: coagulation + flocculation													
PACla/0.08 mM (Al)	Synthetic (HA)	MF	200 rpm	1 min	N/A	40 rpm	15 min	N/A	1.9 × 10^{11}	N/A	8.4 × 10^{12}	N/A	34
PAClb/0.08 mM (Al)	3.0–3.13 mg L^{-1} DOC	PVDF							3.4 × 10^{11}		1.7 × 10^{13}		
PAClp/0.08 mM (Al)	pH 7.8–7.9	0.22 μm							3.0 × 10^{11}		1.5 × 10^{13}		
		(mean)											
Alum/	Synthetic (HA)	MF	100	60	6000	25	1200	30 000	N/A	N/A			42
1.7 mg L^{-1} (Al)	7.5 mg L^{-1}	GVWP									9.00 × 10^{7}	1.25 × 10^{6}	
2.6 mg L^{-1} (Al)	pH 8.3	0.22 μm									6.50 × 10^{7}	0.95 × 10^{6}	
3.4 mg L^{-1} (Al)		(nominal)									2.50 × 10^{7}	0.50 × 10^{6}	
5.1 mg L^{-1} (Al)											2.75 × 10^{7}	0.70 × 10^{6}	
6.8 mg L^{-1} (Al)											3.00 × 10^{7}	0.90 × 10^{6}	
8.5 mg L^{-1} (Al)											3.00 × 10^{7}	1.10 × 10^{6}	
PACl/													
2.6 mg L^{-1} (Al)													
3.4 mg L^{-1} (Al)											12.5 × 10^{7}	1.70 × 10^{6}	
5.1 mg L^{-1} (Al)											6.00 × 10^{7}	1.05 × 10^{6}	
6.8 mg L^{-1} (Al)											6.00 × 10^{7}	1.40 × 10^{6}	
8.5 mg L^{-1} (Al)											6.00 × 10^{7}	1.60 × 10^{6}	
ACH/											7.00 × 10^{7}	2.00 × 10^{6}	

Table 3 *(continued)*

			Coagulation			Flocculation			Membrane resistance				
Coagulant/dosage	Feedwater	Membrane type	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G} \cdot t$	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G} \cdot t$	$R_{(t)}$ (m^{-1})	$R_{c(t)}$ (m^{-1})	α (m kg^{-1})	R'_c (m^{-2})	Ref.
2.6 mg L^{-1} (Al)											35.0×10^7	4.50×10^6	
3.4 mg L^{-1} (Al)											12.0×10^7	2.20×10^6	
5.1 mg L^{-1} (Al)											7.00×10^7	1.50×10^6	
6.8 mg L^{-1} (Al)											7.00×10^7	1.90×10^6	
8.5 mg L^{-1} (Al)											7.00×10^7	2.20×10^6	
PACl/3.18 mg L^{-1} (Al_2O_3)	River	MF	150	3 min	27 000	45	20 min	N/A	N/A	N/A	0.80×10^{12}	N/A	28
	2–3 mg L^{-1} DOC	GVWP					1 h				0.70×10^{12}		
	pH N/A	0.22 μm					2 h				0.60×10^{12}		
		(nominal)					4 h				0.25×10^{12}		
							8 h				0.20×10^{12}		
Al_2SO_4/0.11 mM	Natural	MF	300 rpm	60	N/A	30 rpm	30–90	N/A	N/A	0.49×10^{11}	N/A	1.10×10^{17}	38
0.22 mM	6.30–7.81 mg L^{-1} DOC	Cellulose								0.62×10^{11}		0.84×10^{17}	
0.33 mM	pH 6.9–7.1	acetate								0.73×10^{11}		0.71×10^{17}	
PACl/0.15 mM		0.22 μm								0.89×10^{11}		1.62×10^{17}	
0.30 mM										1.22×10^{11}		1.29×10^{17}	
0.45 mM										1.45×10^{11}		1.08×10^{17}	
$FeSO_4$/0.13 mM										1.03×10^{11}		1.58×10^{17}	
0.22 mM										0.76×10^{11}		0.78×10^{17}	
0.31 mM										2.01×10^{11}		0.69×10^{17}	
$FeCl_3$/0.09 mM										0.94×10^{11}		1.89×10^{17}	
0.18 mM										1.17×10^{11}		1.41×10^{17}	
0.27 mM										1.15×10^{11}		0.98×10^{17}	
Type 3: conventional coagulation													
Alum/10 mg L^{-1}	River	MF	100 rpm	3 min	N/A	30 rpm	20 min	N/A	N/A	N/A	3.5×10^{11}	N/A	44
	2.0–2.5 mg L^{-1} TOC	PVDF									4.0×10^{11}		
	pH 7–8	0.22 μm									4.5×10^{11}		
		(nominal)									4.5×10^{11}		
100 mg L^{-1}											2.5×10^{12}		
											3.0×10^{12}		
											3.4×10^{12}		
											4.0×10^{12}		

[a] N/A = not available. [b] Determined over a TMP range of 20–80 kPa.

Table 4 Effect of coagulation/flocculation configuration types on membrane resistance for ultrafiltration

			Coagulation			Flocculation			Membrane resistance				
Coagulant/dosage	Feedwater	Membrane type	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G}\cdot t$	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G}\cdot t$	$R_{(t)}$ (m^{-1})	$R_{c(t)}$ (m^{-1})	α (m kg^{-1})	R'_c (m^{-2})	Ref.
Type 1: coagulation + no/incidental flocculation													
No coagulant	Lake	UF	N/A[a]	N/A	N/A	N/A	N/A	N/A	N/A		N/A	N/A	12
	(biopolymers)	PES											
	pH N/A	150 kDa											
	0.22 mg L^{-1}	MWCO								2.4×10^{12}			
	0.24 mg L^{-1}									2.0×10^{12}			
	0.47 mg L^{-1}									3.6×10^{12}			
	0.50 mg L^{-1}									2.5×10^{12}			
	0.51 mg L^{-1}									3.1×10^{12}			
	0.54 mg L^{-1}									4.0×10^{12}			
	0.57 mg L^{-1}									3.0×10^{12}			
	0.62 mg L^{-1}									3.9×10^{12}			
	0.80 mg L^{-1}									5.3×10^{12}			
	0.89 mg L^{-1}									3.8×10^{12}			
	0.90 mg L^{-1}									5.7×10^{12}			
No coagulant	Synthetic	UF	N/A	N/A	N/A	N/A	N/A	N/A	N/A	N/A		N/A	71
	(HA/SA)	PES											
	10 mg L^{-1}	150 kDa											
	TOC	MWCO											
	pH 7.8												
	HA										1.50×10^{14}		
	+ 1 mM Ca^{2+}										$3.00–3.50 \times 10^{14}$		
	3 : 1 (HA/SA)										4.20×10^{14}		
	+ 1 mM Ca^{2+}										$2.20–2.25 \times 10^{15}$		
	1 : 1 (HA/SA)										5.70×10^{15}		
	+ 1 mM Ca^{2+}										$3.10–3.90 \times 10^{15}$		
	1 : 3 (HA/SA)										8.50×10^{14}		
	+ 1 mM Ca^{2+}										$3.90–4.00 \times 10^{15}$		
	SA										1.80×10^{15}		
	+ 1 mM Ca^{2+}										$4.00–4.60 \times 10^{15}$		
No coagulant	Synthetic (HA)	UF	60 rpm	3 min	N/A	N/A	N/A	N/A	12×10^{10}	N/A	N/A	N/A	32
Alum/	DOC N/A	PES							110×10^{10}				
0.59 mg L^{-1} Al	pH 4.81–8.73	100 kDa											
0.59 mg L^{-1} Al		MWCO							35×10^{10}				
1.17 mg L^{-1} Al									6×10^{10}				
1.76 mg L^{-1} Al									7×10^{10}				
2.34 mg L^{-1} Al									8.5×10^{10}				
2.93 mg L^{-1} Al									10×10^{10}				
No coagulant	Lake	UF	N/A	N/A	N/A	N/A	N/A	N/A	$9.7–15.0 \times 10^{11}$	N/A	N/A	N/A	9
$FeCl_3$/	3909 ppb TOC	PES							$7.4–8.2 \times 10^{11}$				
1.0 ppm	pH N/A	100 kDa											
2.0 ppm		MWCO							$7.4–8.9 \times 10^{11}$				
5.0 ppm									$7.4–10.2 \times 10^{11}$				
2.0/1.0 ppm[b]									$5.3–10.4 \times 10^{11}$				
Alum/	Synthetic (HA)	UF	N/A	N/A	12 600	5.25	360	1890	N/A	N/A	$3.17–5.93 \times 10^{13}$	N/A	22
70 mg L^{-1}	10 mg L^{-1} TOC	PES											
	pH 4.8–5.5	50 kDa											

Table 4 *(continued)*

			Coagulation			Flocculation			Membrane resistance				
Coagulant/dosage	Feedwater	Membrane type	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G} \cdot t$	$\bar{G}$ (s^{-1})	Time (s)	$\bar{G} \cdot t$	$R_{(t)}$ (m^{-1})	$R_{c(t)}$ (m^{-1})	α (m kg^{-1})	R'_c (m^{-2})	Ref.
		MWCO											
No coagulant PACl/ 4.1 ppm (Al_2O_3)	River 2.3–2.9 ppm DOC pH 7.2–7.8	UF PS 0.01 μm (nominal)	N/A	N/A	N/A	0	720	N/A	N/A	2.1–3.7 × 10^{10}	0.1–0.5 × 10^{13} 7.0–9.0 × 10^{13}	N/A	29
Type 2: coagulation + flocculation													
Alum/ 70 mg L^{-1}	Synthetic (HA) 10 mg L^{-1} TOC pH 4.8–5.5	UF PES 50 kDa MWC O	N/A	N/A	12 600	14.85 42.00	1200	17 820 50 400	N/A	N/A	3.17–4.16 × 10^{13} 3.10–5.10 × 10^{13}	N/A	22
No coagulant PACl/ 4.1 ppm (Al_2O_3)	River 2.3–2.9 ppm DOC pH 7.2–7.8	UF PS 0.01 μm (nominal)	N/A	N/A	N/A	 58–350[c]	 1800–3600	N/A	N/A	 1.8–5.4 × 10^9	0.1–0.5 × 10^{13} 6.0 × 10^{12}–1.1 × 10^{13}	N/A	29
Type 3: conventional coagulation													
Alum/ 70 mg L^{-1}	Synthetic (HA) 10 mg L^{-1} TOC pH 4.8–5.5	UF PES 50 kDa MWCO	N/A	N/A	12 600	5.25 14.85 42.00	360 1200 1200	1890 17 820 50 400	N/A	N/A	3.37–6.79 × 10^{13} 7.96–9.80 × 10^{13} 9.29–11.91 × 10^{13}	N/A	22
No coagulant Alum/ 0.5 mg L^{-1} 15 mg L^{-1}	Lake 2.13 ± 0.08 mg L^{-1} TOC pH 8.18 ± 0.2	UF PVDF 0.04 μm (nominal)	N/A	N/A	N/A	N/A	N/A	N/A	4.00–8.25 × 10^{11} 4.30–7.30 × 10^{11} 5.65–10.80 × 10^{11}	N/A	N/A	N/A	8
No coagulant Alum/ 0.5 mg L^{-1} 15 mg L^{-1}	Lake 4.25 ± 0.06 mg L^{-1} TOC pH 8.05 ± 0.12								8.60–13.40 × 10^{11} 4.05–7.80 × 10^{11} 2.05–7.80 × 10^{11}				
No coagulant Alum/ 0.5 mg L^{-1} 15 mg L^{-1}	River 5.99 mg L^{-1} TOC pH 8.1 ± 0.37								14.65–17.00 × 10^{11} 5.65–11.25 × 10^{11} 8.25–11.75 × 10^{11}				

[a] N/A = not available. [b] 2 ppm $FeCl_3$ dosed during first cycle and 1 ppm in subsequent cycles over first 30 min of a 60 min permeation cycle. [c] Lower values in range typical of Type 2, upper values Type 1; not distinguished in study, therefore data only presented for Type 2.

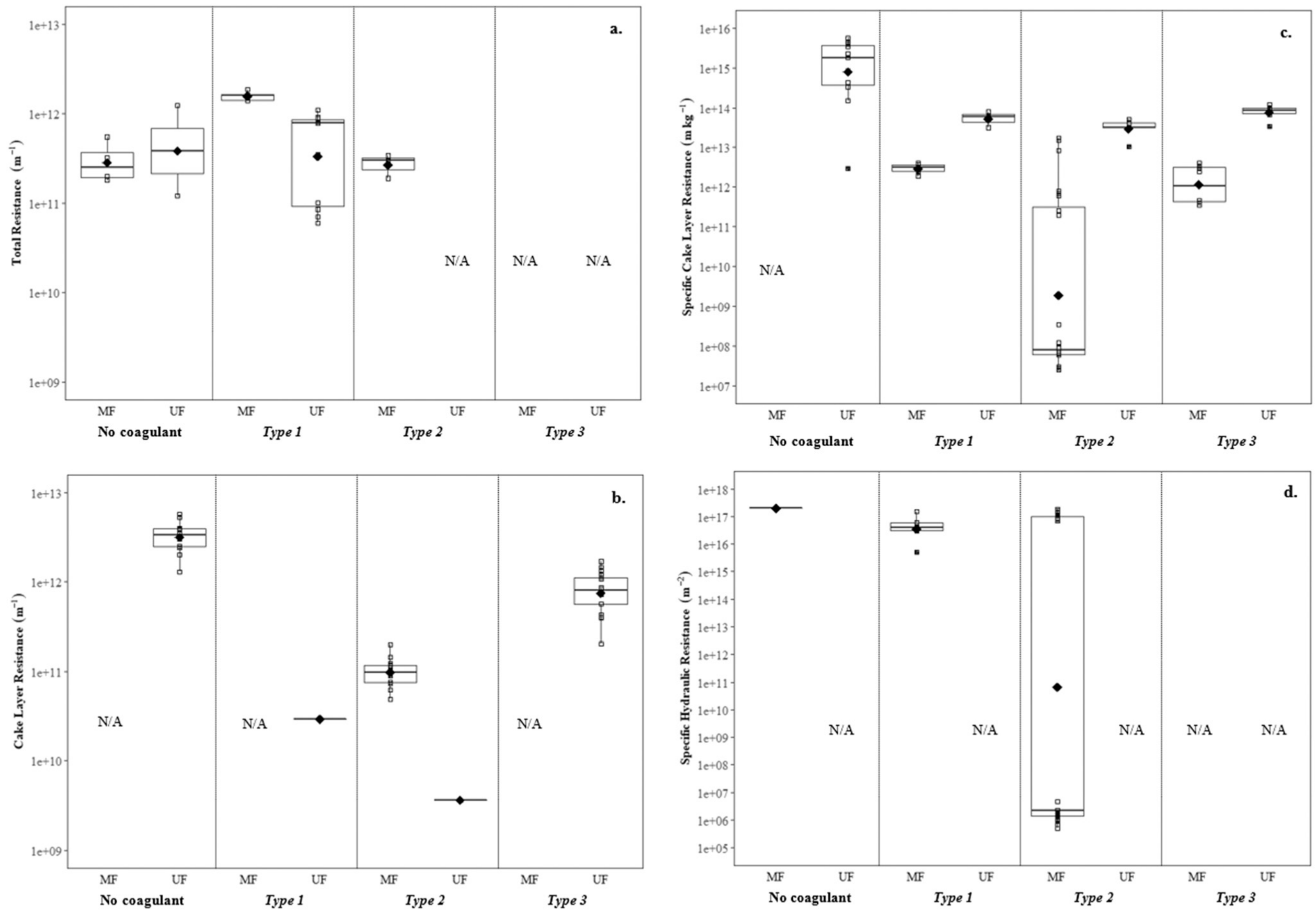

Fig. 3 Performance for a. total resistance, b. fouling resistance, c. specific cake layer resistance, and d. specific hydraulic resistance for coagulation/flocculation configuration types. Note: boxes represent the 25th to 75th percentiles, whiskers the lowest and highest values up to 1.5 times the inter-quartile range. N/A = not available.

coagulant was added for only the initial 50% of the duration of the first permeation cycle, followed by 1 ppm (0.34 mg L^{-1} Fe) for the initial 50% of the duration of subsequent cycles. The ability to achieve similar or lower total membrane resistance with phased coagulation has the potential to significantly reduce coagulant requirements.

There are no apparent trends in the results for total membrane resistance (Fig. 3a); however, both Pronk[9] and Choi and Dempsey[32] reported lower fouling for configuration Type 1 when compared to no coagulant addition. The limited results presented in Fig. 3b suggest that cake layer resistance for UF may be lower for configuration Type 1 when compared to no coagulant and Type 3, but higher when compared to Type 2. It was suggested that greater specific cake layer resistance for Type 1 could be due to the formation of smaller floc, which may form a less permeable cake layer when compared to the larger floc formed by configuration Type 2 (ref. 22 and 29) (Fig. 3c). As noted by Amjad,[22] specific cake layer resistance for Type 1 is likely to be lower when compared to that of Type 3 because of smaller floc that remain following settling (Fig. 3c). Specific cake layer resistance has been reported to decrease as rapid mixing time increased (20 min to 8 h) due to a decrease in the fractal dimension of floc.[28] Lower fractal values may not reduce resistance where compression occurs, although this may be negligible when operating under low or moderate pressures (*e.g.* ≤40 kPa). Amjad[22] observed that while cake layer thickness and resistance increased over the duration of a permeation cycle, porosity also increased.

4.3.2. Fouling rate. Results from multiple studies indicate that the duration of membrane permeation cycles (*i.e.* the time between required hydraulic and chemical cleaning) can be increased using coagulation/flocculation pretreatment by producing floc that is larger than the membrane pores[27,29] (Tables 5 and 6) (Fig. 4a and b). Low coagulant dosages may either exacerbate or reduce fouling rates (*i.e.* rate of TMP increase), impacting permeation cycle duration. Where partial charge neutralization occurs, incomplete aggregation may result in floc that are smaller than membrane pores thus increasing internal pore blocking.[27] However, where floc are sufficiently small, and foulant-membrane interactions limited, particles may also pass through the membrane filter, which would reduce fouling. Amjad[22] reported lower flux decline (~25.0% L^{-1}) over a permeation cycle for flocculation with low $\bar{G}t$ when compared to flux declines for medium

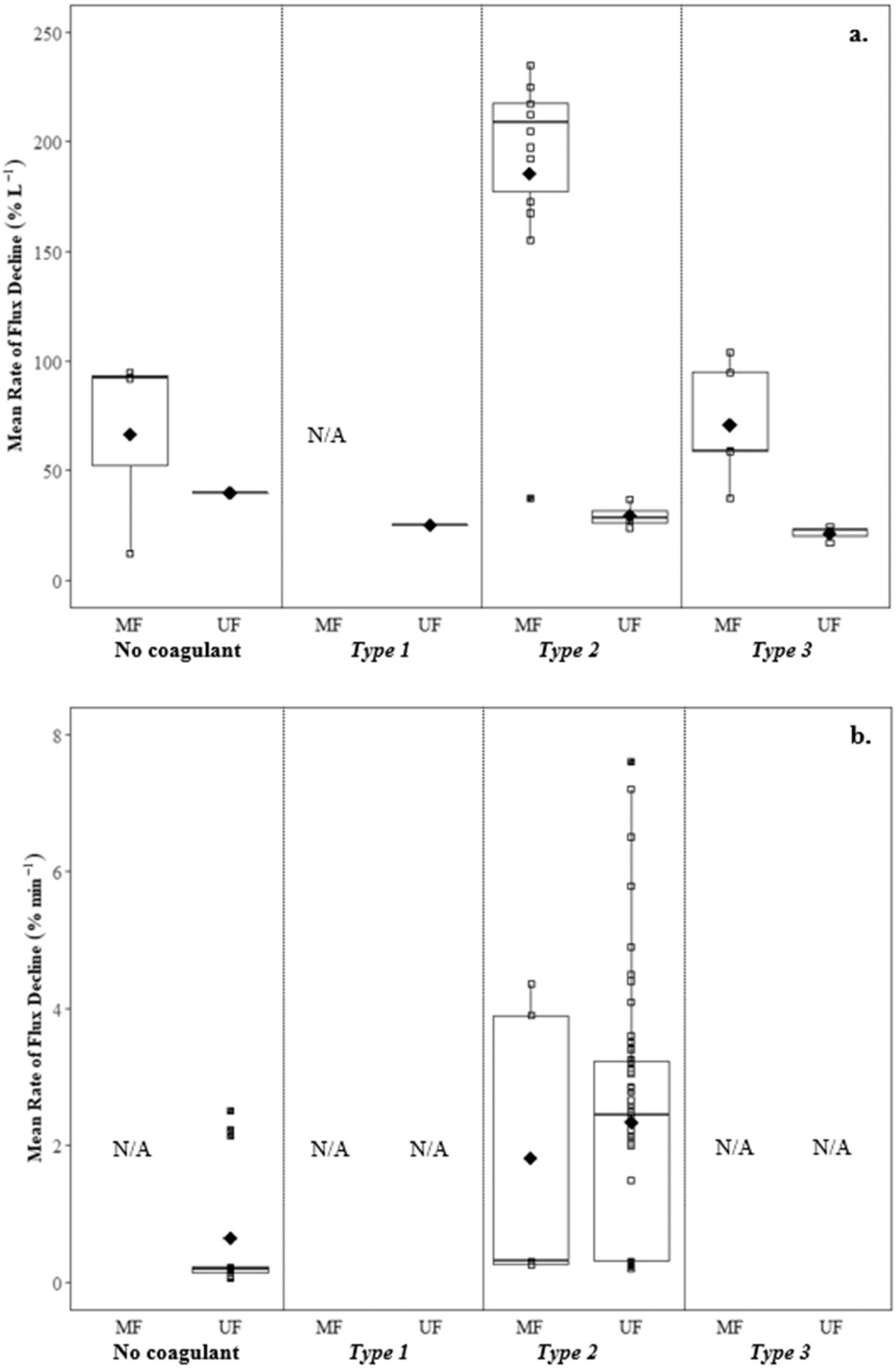

Fig. 4 Performance for mean rate of flux decline as a. % L^{-1}, and b. % min^{-1} for coagulation/flocculation configuration types. Note: boxes represent the 25th to 75th percentiles, whiskers the lowest and highest values up to 1.5 times the inter-quartile range. N/A = not available.

(~36.7% L^{-1}) and high $\bar{G}t$ (~36.7% L^{-1}, ~30.0% L^{-1}) (Table 6). Low flux decline with low $\bar{G}t$ coincided with the smallest floc size and lowest solids removal (60.3%).

4.3.3. Reversible/irreversible fouling. Membrane flux can be recovered to varying degrees by hydraulic and chemical cleaning. For configuration Type 1, hydraulically and chemically reversible fouling ranged from 9 to 99% and 0 to 100%, respectively (Table 7). The results in Fig. 5a and b indicate that for UF the mean value of hydraulically reversible fouling increased and that the mean value of chemically irreversible fouling decreased for Type 1 when compared to no coagulant. Regarding hydraulically reversible/irreversible fouling for UF, the mean values for Type 1 are similar to those reported for Type 3. The higher percentages of hydraulically reversible fouling reported by Judd and Hillis[27] (88 to 96%) when compared to those reported by Kimura *et al.*[70] (9 to 24%) may

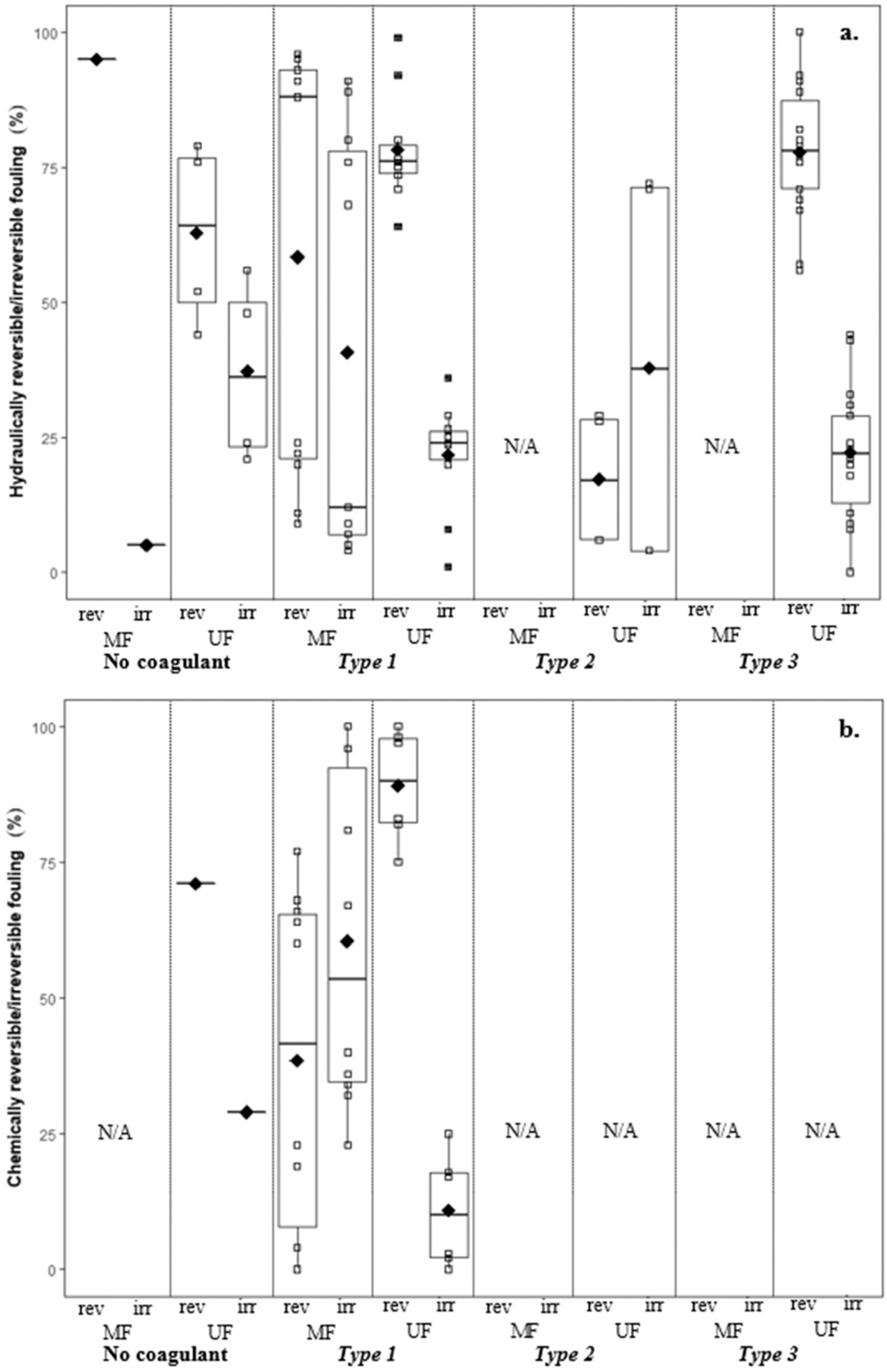

Fig. 5 Performance for flux recovery as a. hydraulically reversible (rev)/irreversible (irr), and b. chemically reversible/irreversible for coagulation/flocculation configuration types. Note: boxes represent the 25th to 75th percentiles, whiskers the lowest and highest values up to 1.5 times the inter-quartile range. N/A = not available.

be partially due to greater backwash frequency and intensity. Backwashes were conducted once every 10 min at 200 LMH in the former case, and once every 30 min at 94 LMH for the latter, which suggests that greater backwash frequency and intensity could increase the degree of hydraulically reversible fouling.

Several studies investigated the impact of coagulant dose on reversible/irreversible fouling. Applying a range of coagulant dosages at various pH levels, Choi and Dempsey[32] observed comparatively high hydraulically and chemically reversible fouling for a low coagulant dose (0.59 mg L^{-1}) (99% and 97%). This may have been due to partial charge neutralization, and formation of floc with a slightly negative charge that are more readily removed from the negatively charged membrane surface. Depending on coagulation conditions, results suggest that there may be a trade-off between greater membrane fouling and more frequent cleaning, as well as the recovery of permeability (*i.e.* increased fouling but higher permeability recovery). While specific hydraulic resistance decreased with increasing coagulant dose, Judd and Hillis[27] reported that the ratio of hydraulically reversible/irreversible resistance remained approximately the same at all coagulant dosages.

Table 5 Impact of coagulation/flocculation configuration types on transmembrane pressure for MF and UF

Coagulant/dosage	Feedwater	Membrane type	Backwash frequency	Permeation duration	Initial TMP	Mean rate of TMP increase	Ref.
Type 1: coagulation + no/incidental flocculation							
No coagulant	Reservoir	MF	1/10 min	13 000 s	N/A[a]	0.05 kPa min^{-1}	27
Fe-based/0.018 mM (Fe)	2.4 mg L^{-1} TOC	PES		8000 s		0.11 kPa min^{-1}	
0.027 mM (Fe)	pH ∼5.4	0.1 μm		5000 s		0.11 kPa min^{-1}	
0.036 mM (Fe)		(mean)		8000 s		0.05 kPa min^{-1}	
0.045 mM (Fe)				10 500 s		0.03 kPa min^{-1}	
0.054 mM (Fe)				5000 s		0.03 kPa min^{-1}	
0.072 mM (Fe)				21 000 s		0.01 kPa min^{-1}	
No coagulant	River	UF	1/60 min	9 h[b]	∼45–50 kPa	0.037 kPa min^{-1}	29
PACl/4.1 ppm (Al_2O_3)	2.3–2.9 ppm DOC	PS		26 h		0.013 kPa min^{-1}	
No coagulant	pH 7.2–7.8	0.01 μm	1/20 min	2 h[b]		0.167 kPa min^{-1}	
4.1 ppm (Al_2O_3)		(nominal)		37 h		0.009 kPa min^{-1}	
Type 2: coagulation + flocculation							
4.1 ppm (Al_2O_3)	River	UF	1/60 min	30 h[c]	∼45–50 kPa	0.011 kPa min^{-1}	29
4.1 ppm (Al_2O_3)	2.3–2.9 ppm DOC	PS	1/20 min	50 h[c]		0.007 kPa min^{-1}	
	pH 7.2–7.8	0.01 μm (nominal)					

[a] N/A = not available. [b] Crossflow mode. [c] Dead-end mode.

4.4. Type 2: coagulation + flocculation

4.4.1. Resistance. Typically, both cake layer and specific cake layer resistance for configuration Type 2 are expected to be lower than those for Type 1 due to the formation of larger floc[28,29,63] (Tables 3 and 4) (Fig. 3b and c). However, the impact of intra- and inter-aggregate porosities has been reported to be a contributing factor to cake layer resistance as well.[22,63] Amjad[22] reported similar resistance for configuration Types 1 and 2 despite the formation of larger floc, which would be expected to result in greater inter-aggregate porosity. This could be due to floc formed by Type 1 conditions being associated with lower D_f, which would result in higher intra-particle porosity despite smaller size. In addition, the authors suggested that while Type 2 results in the formation of larger floc with higher D_f when compared to Type 1, cake layer restructuring caused the specific cake layer resistance for Type 2 to decrease over time, eventually converging with that of Type 1. Wang[42] reported that smaller floc formed using monomeric alum coagulant resulted in the formation of a more porous cake layer than that of polynuclear PACl and aluminum chloro-hydrate (ACH), which may form gels and a tight cake layer structure. This could be attributed to the alum flocs having lower D_f, and greater intra-aggregate porosity. Where D_f values are similar, larger floc would still be anticipated to result in lower cake layer resistance.[28] For cake layers formed by particles with diameters of 400 and 40 μm, at a pressure of 40 kPa the reduction of inter- and intra-particle porosities were 67% and 88%, and 14%, 29%, respectively.[63] The significant reduction in inter-particle porosity indicates that the inter-particle porosity of particles has a greater effect on the overall cake layer porosity, which may be anticipated since inter-particle voids are expected to be larger when compared to intra-particle voids. Lee[72] suggested that the effect of pressure on intra-particle permeability is significant for small floc (*e.g.* 8.1 to 17 μm) while it has little influence on large floc (*e.g.* 40.6 to 48.5 μm).

Operating MF membranes in dead-end mode, Lee[44] reported that relative specific cake layer resistance ($\alpha_{\text{coagulation}}/\alpha_{\text{raw}}$) was lower when charge neutralization was the destabilization mechanism (<0.7) when compared to sweep flocculation (>1.0). Floc size distributions were similar for both mechanisms, thus it was suggested that the difference was due to floc compressibility, where floc formed under sweep flocculation were three times more compressible than floc formed by charge neutralization. That floc formed by sweep flocculation are more compressible appears counter-intuitive given that several studies have

Table 6 Impact of coagulation/flocculation configuration types on membrane flux for MF and UF

Coagulant/dosage	Feedwater	Membrane type	Coagulation/flocculation conditions	Volume filtered per cycle	Permeation time per cycle	Mean rate of flux decline	Ref.
Type 1: coagulation + no/incidental flocculation							
Alum/70 mg L^{-1}	Synthetic (HA) 10 mg L^{-1} TOC pH 4.8–5.5	UF PES 50 kDa MWCO	Rapid mix $\bar{G}\cdot t$ 12 600 + 1200 s slow mix (5.25 s^{-1})	~1.5 L	N/A[a]	25% L^{-1}	22
No coagulant	Synthetic (HA/SA) 10 mg L^{-1} TOC pH 7.8	UF PE 150 kDa	N/A	N/A			71
	HA	MWCO			6.5 h	0.14% min^{-1}	
	+1 mM Ca^{2+}				7 h	0.19% min^{-1}	
	3 : 1 (HA/SA)				7 h	0.07% min^{-1}	
	+1 mM Ca^{2+}				6.75 h	0.20% min^{-1}	
	1 : 1 (HA/SA)				7 h	0.08% min^{-1}	
	+1 mM Ca^{2+}				6.75 h	0.21% min^{-1}	
	1 : 3 (HA/SA)				7 h	0.10% min^{-1}	
	+1 mM Ca^{2+}				7 h	0.21% min^{-1}	
	SA				6.5 h	0.17% min^{-1}	
	+1 mM Ca^{2+}				6.5 h	0.22% min^{-1}	
Type 2: coagulation + flocculation							
No coagulant	River	MF	Stirred	900 mL	N/A	92.2% L^{-1}	24
Alum/3.2 mg L^{-1} (Al)	9.0 mg L^{-1} DOC pH 6.0	PP 0.2 μm (nominal)				58.9% L^{-1}	
No coagulant	Synthetic (HA)	MF	60 s rapid mix (100 s^{-1}) +	400 mL	N/A	<12.5% L^{-1}	42
Alum/1.7 mg L^{-1} (Al)	7.5 mg L^{-1}	GVWP	1200 s slow mix (25 s^{-1})			212.5% L^{-1}	
2.6 mg L^{-1} (Al)	pH 8.3	0.22 μm				172.5% L^{-1}	
3.4 mg L^{-1} (Al)		(nominal)				155% L^{-1}	
5.1 mg L^{-1} (Al)						167.5% L^{-1}	
6.8 mg L^{-1} (Al)						197.5% L^{-1}	
8.5 mg L^{-1} (Al)						197.5% L^{-1}	
PACl/1.7 mg L^{-1} (Al)						37.5% L^{-1}	
2.6 mg L^{-1} (Al)						217.5% L^{-1}	
3.4 mg L^{-1} (Al)						192.5% L^{-1}	
5.1 mg L^{-1} (Al)						205% L^{-1}	
6.8 mg L^{-1} (Al)						212.5% L^{-1}	
8.5 mg L^{-1} (Al)						217.5% L^{-1}	
ACH/1.7 mg L^{-1} (Al)						37.5% L^{-1}	
2.6 mg L^{-1} (Al)						235% L^{-1}	
3.4 mg L^{-1} (Al)						225% L^{-1}	
5.1 mg L^{-1} (Al)						212.5% L^{-1}	
6.8 mg L^{-1} (Al)						217.5% L^{-1}	
8.5 mg L^{-1} (Al)						225% L^{-1}	
PACl/3.18 mg L^{-1} (Al_2O_3)	River	MF	3 min–8 h rapid mix (150 s^{-1})	N/A	15 min	3.8–4.9% min^{-1}	28
	2–3 mg L^{-1} DOC pH N/A[a]	GVWP 0.22 μm (nominal)	3 min rapid mix (150 s^{-1}) + 3 min–8 h slow mix (45 s^{-1})			2.9–4.9% min^{-1}	
AC/8 mg L^{-1} (Al)	Synthetic (HA)	UF	90 s rapid mix (200 rpm) +	N/A	300 min	0.21–0.26% min^{-1}	41
PACb/8 mg L^{-1} (Al)	5.35 mg L^{-1} DOC	PES	900 s slow mix (40 rpm)		300 min	0.23–0.24% min^{-1}	

Table 6 *(continued)*

Coagulant/dosage	Feedwater	Membrane type	Coagulation/flocculation conditions	Volume filtered per cycle	Permeation time per cycle	Mean rate of flux decline	Ref.
PACc/8 mg L^{-1} (Al)	pH 4–8	100 kDa MWCO			275–300 min	0.20–0.26% min^{-1}	
No coagulant PACl/0.025 mM (Al) PACl/0.1 mM (Al)	Synthetic (HA) 5 mg L^{-1} pH 7.5	UF PES 100 kDa MWCO	90 s rapid mix (175 s^{-1}) + 900 s slow mix (20 s^{-1})	1000 mL	N/A	40% L^{-1} 27% L^{-1} 24% L^{-1}	33
FeCl$_3$/22 mg L^{-1} (Fe) PFC$_{10}$/22 mg L^{-1} (Fe) PFC$_{22}$/22 mg L^{-1} (Fe)	Synthetic (HA) 4.67 ± 0.25 mg L^{-1} DOC pH 4–9	UF PES 100 kDa MWCO	90 s rapid mix (200 rpm) + 900 s slow mix (40 rpm)	N/A	~14 000 s	0.21–0.30% min^{-1} 0.21–0.31% min^{-1} 0.31% min^{-1}	43
  AlCl$_3$/0.25–50 μM (Al) AlCl$_3$/0.25–50 μM (Al) AlCl$_3$/1–50 μM (Al) No coagulant AlCl$_3$/2.5–30 μM (Al) No coagulant AlCl$_3$/2.5–20 μM (Al) No coagulant AlCl$_3$/1–20 μM (Al) AlCl$_3$/0.025–30 μM (Al) AlCl$_3$/0.25–100 μM (Al) AlCl$_3$/0.25–30 μM (Al)	synthetic (HA/BSA) pH 6 10 mg L^{-1} HA 5 mg L^{-1} HA + 5 mg L^{-1} BSA 10 mg L^{-1} BSA pH 7 10 mg L^{-1} HA 5 mg L^{-1} HA + 5 mg L^{-1} BSA 10 mg L^{-1} BSA pH 8 10 mg L^{-1} HA 5 mg L^{-1} HA + 5 mg L^{-1} BSA 10 mg L^{-1} BSA	UF PES 50 kDa MWCO	60 s rapid mix (250 rpm) + 740 s slow mix (100 rpm)	N/A	600–700 s	 0.07–1.54% min^{-1} 3.20–3.90% min^{-1} 2.00–7.60% min^{-1} 2.23% min^{-1} 2.23–3.00% min^{-1} 2.30% min^{-1} 2.49–3.23% min^{-1} 2.50% min^{-1} 2.60–6.40% min^{-1} 2.12–3.05% min^{-1} 2.03–3.23% min^{-1} 2.14–2.83% min^{-1}	26
Alum/70 mg L^{-1}	Synthetic (HA) 10 mg L^{-1} TOC pH 4.8–5.5	UF PES 50 kDa MWCO	Rapid mix $\bar{G}\cdot t$ 12 600 + 1200 s slow mix (14.85 s^{-1}) Rapid mix $\bar{G}\cdot t$ 12 600 + 1200 s slow mix (42.00 s^{-1})	~1.5 L	N/A	36.7% L^{-1} 30.0% L^{-1}	22
Type 3: conventional coagulation							
No coagulant Alum/3.2 mg L^{-1} (Al)	River 9.0 mg L^{-1} DOC pH 6.0	MF PP 0.2 μm (nominal)	Settling	900 mL	N/A	92.2% L^{-1} 58.9% L^{-1}	24
No coagulant Alum/4 mg L^{-1} 12 mg L^{-1} 25 mg L^{-1} 50 mg L^{-1}	River 3.1 mg L^{-1} DOC pH 6.9–7.3	MF PP 0.2 μm (nominal)	Rapid mix + 30 min slow mix (60 s^{-1})	400–900 mL	N/A	~94.7% L^{-1} ~104% L^{-1} ~94.7% L^{-1} ~58.7% L^{-1} ~37.3% L^{-1}	23
Alum/70 mg L^{-1}	Synthetic (HA) 10 mg L^{-1} TOC pH 4.8–5.5	UF PES 50 kDa MWCO	Rapid mix $\bar{G}\cdot t$ 12 600 + 1200 s slow mix (5.25 s^{-1}) Rapid mix $\bar{G}\cdot t$ 12 600 + 1200 s slow mix (14.85 s^{-1}) Rapid mix $\bar{G}\cdot t$ 12 600 + 1200 s slow mix (42.00 s^{-1})	~1.5 L	N/A	22.7% L^{-1} 24.0% L^{-1} 17.3% L^{-1}	22

[a] Not available.

reported that floc formed by sweep flocculation have higher D_f than those formed by charge neutralization. However, the higher compressibility of floc formed by sweep flocculation may be due to their higher water content, while at the same time they are gelated, more compact, and less porous because of being predominantly made up of aluminum hydroxide precipitates.[44] Lee[44] suggested that floc formed by charge neutralization consist of aluminum cation and inorganic/organic complexes that are less compressible.

4.4.2. Fouling rate. Similar to configuration Type 1, several studies have reported that the rate of membrane flux decline can be reduced by configuration Type 2 (ref. 24 and 33) (Table 6). Evidence suggests that the fouling rate for Type 2 can be lower than that for Type 1, as longer permeation durations have been observed with both crossflow (1 m s^{-1} crossflow velocity) and dead-end modes for the same coagulation conditions[29] (Table 5). This is expected given the observation that configuration Type 2 produces larger floc, which are expected to result in a more permeable cake layer.[22,28,29] As such, it follows that as mixing time increases during both coagulation and flocculation, fouling rate decreases.[28] When compared to configuration Type 3, for medium and high $\bar{G}t$ followed by direct filtration Amjad[22] observed lower specific cake layer resistances for Type 2, but a greater rate of flux decline. This was attributed to a greater mass flux towards the membrane surface without settling.

Again, it was observed that a threshold coagulant dose exists below which fouling rate may increase. However, it may also be the case that low coagulant doses result in lower flux decline due to insufficient floc development and fewer particles being retained on the membrane.[42] Ma[26] observed critical doses of Al that resulted in dramatic flux reduction for water matrices containing humic acid (HA), bovine serum albumin (BSA), and a 1 : 1 mass ratio of HA/BSA, which were mainly induced by particle size. Flux decline varied slightly with pH for HA and significantly for BSA solutions, indicating that NOM type plays an important role in fouling. Dong[43] reported that at pH ranging from 7.0 to 9.0 flux declines were more dramatic than at pH ranging from 4.0 to 6.0. It was suggested that lower fouling rates at lower pH were due to either greater floc size or lower D_f. In addition, the better performance of $FeCl_3$ and polyferric chloride with basicity of 1.0 (PFC_{10}) at pH 6.0 and 7.0 was attributed to the predominance of monomeric and polymeric species.[43]

4.4.3. Reversible/irreversible fouling. Yao[33] observed lower overall fouling at a coagulant dosage optimized for turbidity and humic acid removal (0.1 mM Al), compared to fouling at a low dose (0.025 mM Al) (Table 7). However, at the low dose a greater proportion of fouling was hydraulically reversible, while hydraulically irreversible fouling was only slightly lower. Thus, it was suggested that the application of low dose coagulation be investigated further. As discussed in section 4.4.2., Amjad[22] observed lower specific cake layer resistances for configuration Type 2 than for other configurations, but greater rates of flux decline. Since porous cake layers are likely easier to remove by hydraulic backwashing, the combination with greater flux decline indicates that there may be a trade-off between greater hydraulic and chemical cleaning efficiency and higher cleaning frequency because of more rapid flux decline.

4.5. Type 3: conventional coagulation

4.5.1. Resistance. Floc that are removed by settling during Type 3 do not directly contribute to membrane fouling, instead it is the properties of the particles that remain after settling that contribute to membrane fouling. As discussed in section 3.4, there may be cases where the D_f of particles remaining after settling is similar to that of the settled floc. Amjad[22] observed similar cake layer fractal dimension, D_c, with (2.943 ± 0.007 to 2.987 ± 0.005) and without settling (2.931 ± 0.001 to 2.977 ± 0.003). Despite similar D_c, specific cake resistance was higher when floc was removed by settling than without floc removal (Fig. 3c). The authors suggested that this was due to the remaining particles being smaller and packing into less porous cake. The same authors also observed effective cake porosity to typically increase with permeation time, a phenomenon attributed to breakup and restructuring of floc within the cake layer.[72–74]

4.5.2. Fouling rate. Carroll[24] reported no change in the rate of flux decline when examining configuration types with or without settling (Table 6). Comparing both configuration types, the portion of the fouling rate associated with floc could be separated from that associated with dissolved NOM. The authors suggested that dissolved NOM, which remained following settling, was the main contributor to membrane fouling. When comparing low, medium, and high $\bar{G}t$ conditions followed by settling, Amjad[22] reported lower rates of flux decline with settling (ranging from 17.3 to 24.0% L^{-1}) than without settling (ranging from 25.0 to 36.7% L^{-1}), likely due to the reduction of solids in the feedwater when measured gravimetrically. Multiple studies have reported similar flux declines when evaluating raw water and water following coagulation with an optimum coagulant dosage for turbidity removal,[23,24] suggesting NOM as being the main contributor to flux decline. At higher coagulant dosages more suitable for NOM removal (3.2 to 4.0 mg L^{-1} Al^{3+}), reported flux declines were much lower.

4.5.3. Reversible/irreversible fouling. Limited information exists regarding both hydraulically and chemically reversible/irreversible fouling for configuration Type 3. However, since the particles remaining in feedwater following settling are expected to be small, it has been suggested that they will form a less porous cake layer that may be more difficult to remove during hydraulic and chemical cleaning.[22] Thus, while the rate of flux decline may be slower with Type 3, the resulting cake layer properties may have important implications for the reversibility of fouling. Additionally, as NOM has been identified as an important contributor to membrane fouling,[8–11] the potential to achieve greater removals of NOM prior to membrane treatment by enhanced coagulation and settling could reduce irreversible fouling.

Wray and Andrews[8] reported different results for the impact of coagulant dose on hydraulically reversible/irreversible

Table 7 Membrane fouling after hydraulic/chemical cleaning for coagulation/flocculation configuration types

Coagulant/dosage	Feedwater	Membrane type	Hydraulic/chemical cleaning conditions	Reversible/irreversible fouling (%)				Ref.
				Hydraulically reversible	Hydraulically irreversible	Chemically reversible	Chemically irreversible	
Type 1: coagulation + no/incidental flocculation								
PACl/2 mg L^{-1} (Al)	River	MF	1 per 30 min for 30 s at					70
	1 mg L^{-1} DOC	PVDF	94 LMH/NaClO (24 h)	22[a]	78	23–60	40–67	
	0.5 mg L^{-1} DOC	0.1 μm	NaOH (24 h)	11	89	0–64	36–100	
	2.0 mg L^{-1} DOC		HCl (24 h)	9	91	4–66	34–96	
	1.6 mg L^{-1} DOC			24	76	4–68	32–96	
	1.0 mg L^{-1} DOC			20	80	19–77	23–81	
No coagulant	Reservoir	MF	1 per 10 min at 200 LMH and 2 bar	95	5	N/A[b]	N/A	27
Fe-based/0.018 mM (Fe)	2.4 mg L^{-1} TOC	PES		95	5			
0.027 mM (Fe)	pH ~5.4	0.1 μm		96	4			
0.036 mM (Fe)		(mean)		91	9			
0.045 mM (Fe)				93	7			
0.054 mM (Fe)				88	12			
0.072 mM (Fe)				93	7			
No coagulant	Lake	UF	1 per 90 min for 60 s at 0.65 bar and	75–77	23–25	N/A	N/A	9
$FeCl_3$/	3909 ppb TOC	PES	forward flush for 60 s at 0.3 L min^{-1}	64–83	17–36			
1.0 ppm	pH N/A	100 kDa						
2.0 ppm		MWCO		68–84	16–32			
5.0 ppm				70–83	17–30			
2.0/1.0 ppm[c]				69–73	27–31			
No coagulant	Synthetic (HA)	UF	N/A/NaOH then DI water permeation	79	21	71	29	32
Alum/0.59 mg L^{-1} Al	DOC N/A	PES	for 30 min each at 150 LMH	75	25	98	2	
0.59 mg L^{-1} Al	pH 4.81–8.73	100 kDa		99	1	97	3	
1.17 mg L^{-1} Al		MWCO		92	8	83	17	
1.76 mg L^{-1} Al				64	36	100	0	
2.34 mg L^{-1} Al				76	24	82	18	
2.93 mg L^{-1} Al				80	20	75	25	
Type 2: coagulation + flocculation								
No coagulant	Synthetic	UF	1 per 350 mL with 100 mL ultrapure water	44–52[d]	48–56	N/A	N/A	33
PACl/0.025 mM (Al)	5 mg L^{-1} HA	PES		28–29	71–72			
PACl/0.1 mM (Al)	pH 7.5	100 kDa		6	4			
		MWCO						
Type 3: conventional coagulation								
No coagulant	Lake	UF	1 per 30 min for 10 min at 30 LMH	76–100	0–24	N/A	N/A	8
Alum/0.5 mg L^{-1}	2.13 ± 0.08 mg L^{-1} TOC	PVDF		57–79	21–43			
15 mg L^{-1}	pH 8.18 ± 0.2	0.04 μm		56–79	21–44			
No coagulant	Lake	(nominal)		76–89	11–24			
Alum/0.5 mg L^{-1}	4.25 ± 0.06 mg L^{-1} TOC			71–89	11–29			
15 mg L^{-1}	pH 8.05 ± 0.12			71–80	20–29			
No coagulant	River			77–91	9–23			
Alum/0.5 mg L^{-1}	5.99 mg L^{-1} TOC			69–92	8–31			
15 mg L^{-1}	pH 8.1 ± 0.37			67–82	18–33			

[a] Determined after physical wiping. [b] N/A = not available. [c] 2 ppm $FeCl_3$ dosed during first cycle and 1 ppm in subsequent cycles over first 30 min of a 60 min permeation cycle. [d] Reported as normalized fouling.

fouling for various source waters. It was reported that for Lake Ontario, the addition of 15 mg L^{-1} alum increased reversible fouling, and greater variability in the hydraulic reversibility of fouling was observed at this higher coagulant dose. In contrast, for Lake Simcoe and Otonabee River waters the addition of alum reduced reversible fouling at dosages of both 0.5 and 15 mg L^{-1}. As mentioned in section 2.2.3., this emphasizes the importance of differences in water quality when considering coagulant dosages for membrane pretreatment.

5. Performance of configuration types related to removal of organic matter

5.1. Summary of results

Removals of dissolved and total organic carbon (DOC/TOC) for coagulation/flocculation configuration types are summarized in Fig. 6 (Table 9). For comparison with configuration types, results for membrane filtration with no coagulant addition have also been included. While less results are available for configuration Type 1 regarding floc properties, when compared to Types 2 and 3 there are more results for Type 1 regarding DOC/TOC removal. This may be because the analysis of floc properties for configuration Type 1 is more difficult due to smaller floc size.

5.2. Type 1: coagulation + no/incidental flocculation

A number of studies have investigated the impact of coagulant dosage, coagulant type, and hydrodynamic conditions on the removal of organics and particulates for configuration Type 1. Reported DOC/TOC removal ranges from 42% to 75%, with a mean of 56% for MF, and 16% to 62%, with a mean of 40% for UF (Fig. 6). The mean removal for MF is greater than that reported for no coagulant (9%), below the mean value for configuration Type 2 (61%), and above the mean for configuration Type 3 (38%). The mean removal for UF is approximately the same as that reported for no coagulant (40%), and below the mean values for both configuration Types 2 (47%) and 3 (47%). With no coagulant, results indicate that UF achieves greater DOC/TOC removal when compared to MF, but with the addition of coagulant performance is similar for all configuration types. In general, results suggest that the removal of DOC/TOC increases with increasing coagulant doses.[25,27] There is evidence that charge neutralization, sweep flocculation, and low dose conditions at pH <5.0 are favorable for the removal of DOC/TOC, while low dose conditions at pH 7.0 to 9.0 lead to poor removal.[30,32] Fe-Based coagulants may perform better than Al-based coagulants for biopolymer removal, possibly due to faster generation rates of hydrolysis products and precipitates.[75] Guigui[25] noted that at approximately neutral pH (5.5 to 7.5), there was a greater variance in DOC removal for high (16 to 42%) compared to low (27 to 33%) coagulant doses.

Amjad[22] reported poor solids removal (60.3%, as measured gravimetrically) at low $\bar{G}t$ (1890) for configuration Type 1 when compared to higher $\bar{G}t$ for Types 2 and 3, likely because of incomplete coagulation and dissolved solids not being retained on the membrane surface. Similarly, Howe and Clark[23] reported turbidity reductions after MF of 57% for rapid mixing alone compared to ≥89% for configuration Type 3. Again, this may be expected due to the formation of smaller floc, resulting in lower retention on the membrane surface.

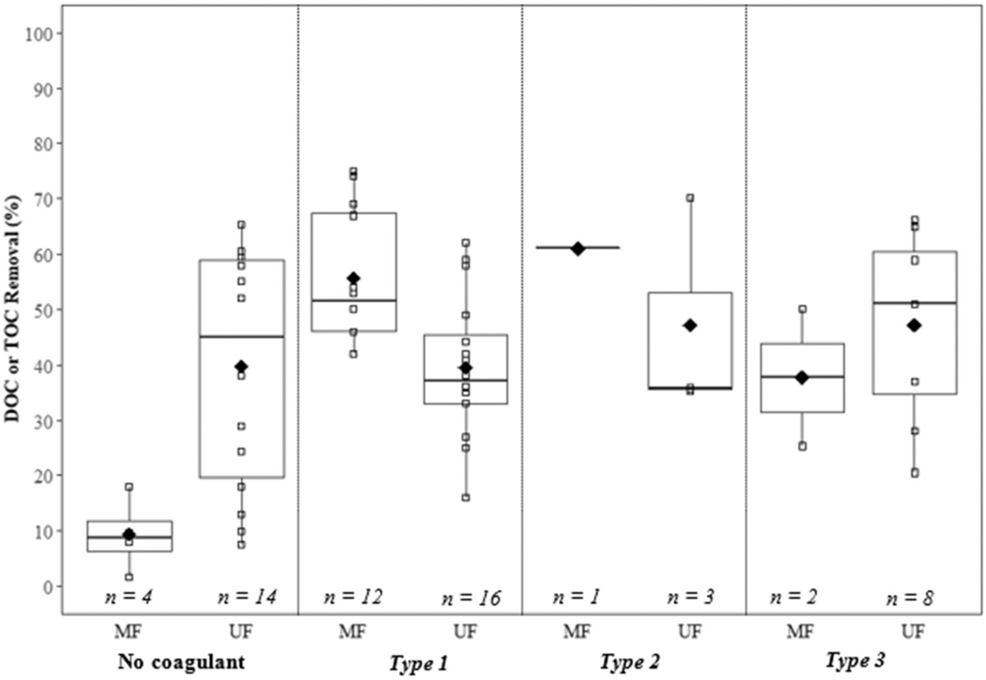

Fig. 6 Published performance for % DOC and TOC removal for coagulation/flocculation configuration types. Note: boxes represent the 25th to 75th percentiles, whiskers the lowest and highest values up to 1.5 times the inter-quartile range. *n* = number of studies cited.

5.3. Type 2: coagulation + flocculation

As for configuration Type 1, studies for configuration Type 2 have examined the impact of coagulant dosage, type, and hydrodynamic conditions on the removal of DOC/TOC and particulates. The reported DOC/TOC removal is 61% for MF, and ranges from 35% to 70%, with a mean of 47% for UF (Fig. 6). The reported removal for MF is greater than that reported for no coagulant, as well as configuration Types 1 and 3, while the me removal reported for UF is greater than that reported for both no coagulant and configuration Type 1, and approximately the same as Type 3. Comparing the removal of TOC by MF for no coagulant addition with that obtained with charge neutralization (10 mg L^{-1}, pH 5.0) and sweep flocculation (30 mg L^{-1}, pH 7.5) mechanisms, Lee[44] observed the greatest reduction in TOC for charge neutralization. The greater TOC removal may be attributed to the charge of soluble organics being less electronegative at pH 5.0, thus improving particle agglomeration. At pH 8.3 with sweep flocculation as the predominant coagulation mechanism, Wang[42] reported UV_{254} removals of ≥90% for alum, PACl, and ACH doses of ≥1.7, 2.6, and 3.4 mg L^{-1} Al, respectively. Yao[33] observed greater reductions in both DOC and UV_{254} using an alum dosage optimized for turbidity removal compared to a low dosage. With a coagulant dosage optimized for DOC and UV_{254} removal, Zhang[40] reported greater removals of both DOC and UV_{254} for coagulation–UF compared to UF with no coagulant addition.

Considering medium and high $\bar{G}t$ (17 820 and 50 400) for configuration Type 2, Amjad[22] observed nearly 100% solids removal following UF. This solids removal was greater than the removal reported for configuration Type 1, and indicated effective aggregation of humic acid. Howe and Clark[23] observed similar turbidity reductions (57%) after rapid mixing with 4 min flocculation (65 rpm) followed by MF or UF. Despite the addition of flocculation, turbidity removal was the same as that for 30 s rapid mixing only.

5.4. Type 3: conventional coagulation

Several studies have considered DOC/TOC removal following configuration Type 3; however, the majority of results reported in the literature are for removal following sedimentation only. DOC/TOC removal after sedimentation ranged from 22% to 51% with a mean of approximately 37% (Table 9). Reported DOC/TOC removal ranges from 25.3% to 50% for MF, with a mean of 38%, while reported DOC/TOC removal ranges from 20.5% to 66.2% for UF, with a mean of 47% (Fig. 6). The mean removal for MF is greater than that reported for no coagulant addition, and lower than that reported for configuration Types 1 and 2, while the mean removal reported for UF is greater than that reported for both no coagulant addition and configuration Type 1, and approximately the same as Type 2. Using alum and PAC, Kabsch-Korbutowicz[46] observed TOC removals to increase by 24.1% and 14.8% for configuration Type 3-UF compared to conventional coagulation with settling alone. Dixon[47] used Type 3-UF to treat river water with PACl dosages optimized for UV_{254} removal and enhanced coagulation, and reported similar DOC and UV_{254} removals after conventional coagulation with settling and Type 3-UF for both coagulant doses. If the removal of organic matter following settling does not increase it could be attributed to dissolved organics and colloids passing through the membrane.[24]

Amjad[22] observed that for hydrodynamic conditions of low (1790), medium (17 820), and high (50 400) $\bar{G}t$, solids removals following settling were approximately 19%, 76%, and 82%, respectively. Subsequent removals by UF were 41.0%, 22.2%, and 15.5% totaling approximately 60% (similar to low $\bar{G}t$ without settling), and nearly 100% for the latter two conditions. Solids removals indicate that medium and high $\bar{G}t$ result in the formation of larger, more settlable floc than low $\bar{G}t$ that are more readily removed by UF.

6. Discussion and conclusion

Results indicate that configuration Type 1 (coagulation + no/incidental flocculation) leads to the formation of small floc with low D_f. There is evidence that this causes the formation of less porous cake layers with greater resistance than those formed by configuration Type 2 (coagulation + flocculation) (Fig. 3), and results in lower removal of solids and turbidity (section 5.2.). As a result, configuration Type 1 may not be optimal for fouling control and DOC/TOC removal when compared to Types 2 and 3; however, more evidence is required. When considering fouling rate, reversibility, and performance in terms of DOC/TOC removal, floc size has been identified as an important factor (sections 4.2. and 5.2.). While a wide range of coagulant doses have been examined for configuration Type 1, it may be beneficial to achieve a balance between thresholds where internal fouling is minimized and DOC/TOC removal targets are achieved, while limiting additional fouling from the accumulation of hydrolytic coagulant products on the membrane surface (section 2.2.1.). Results of several studies indicate that such a balance may be realized at low dose conditions. While characteristically short contact times (≤2 min) appear adequate for developing floc sizes greater than those of membrane pores, the effects of hydrodynamic conditions on floc properties are not clear, perhaps because of difficulty in accurately simulating these conditions at bench-scale. In addition, more information is needed comparing the impact of NOM type on floc properties and subsequent membrane fouling. It is expected that small floc with low D_f would form a cake layer with higher resistance. However, there is also evidence that floc formed at low dosages and acidic pH conditions can result in permeable cake layers with high hydraulic and chemical recoveries (section 3.3), though it is not clear if this would occur for configuration Type 1 without flocculation. Phased coagulation, whereby coagulant is applied during only a portion of the permeation cycle (*e.g.*

50%), is an operational variation that should be investigated further, as it has been reported that fouling and coagulant requirements may be reduced simultaneously (section 4.2.1.).

Application of configuration Type 2 may provide greater control of floc properties. As for configuration Type 1, a range of coagulation doses and destabilization mechanisms have been examined, and as expected for increased particle collisions and contact time, reported floc sizes were generally larger and D_f higher (section 3.3.). Floc formed at acidic pH (≤5.0) have a slower growth rate, but reach larger steady-state size, which could lead to the formation of a more porous cake layer. Results also suggest that floc formed at acidic pH have lower D_f. During mixing floc with lower D_f experienced greater breakage than those formed by the sweep flocculation mechanism with higher D_f. The apparent correlation between D_f and the degree of floc breakage indicates that a correlation also exists between D_f and floc strength. However, it has been reported that floc formed by sweep flocculation are more compressible, which can reduce cake layer permeability due to hydraulic pressure during membrane operation. Future research efforts should be directed towards more clearly distinguishing shear strength and compressibility of floc. While some studies reported that larger floc resulted in lower cake layer resistance, others reported similar resistances to those observed without flocculation (section 4.3.1.). Amjad[22] explained that despite forming cake with higher intra-particle permeability, the higher D_f of floc could compensate for this with lower inter-particle permeability. Finally, without settling, mass flux of floc towards the membrane surface may be comparatively high. While the larger floc may form a more porous and easily removed cake layer, rapid accumulation of material could increase cake layer thickness causing rapid flux decline (sections 4.4.2. and 4.5.2.). A trade-off may exist between cleaning efficiency and frequency. There is evidence that Type 2 conditions result in greater NOM removal when compared to Type 1 (section 5.3.).

For configuration Type 3 (conventional coagulation), settling prior to membrane filtration results in lower mass accumulation on the membrane surface, thus lower fouling rate, but higher specific cake layer resistance (section 4.5.). While the summarized results for DOC/TOC removal (Fig. 6) do not suggest a clear trend when comparing configuration types, Amjad[22] reported greater solids removal for configuration Type 3 when compared to Type 1 and similar solids removal when compared to Type 2. In some cases, it was reported that the rate of flux decline was similar with and without settling, which was attributed to poor removal of NOM despite effective reduction of turbidity (section 4.5.2.). In order to reduce membrane fouling using configuration Type 3, the application of enhanced coagulation for increased NOM removal may be required. Limited information exists regarding the cleaning efficiency of membranes incorporating conventional coagulation (section 4.5.3.). While cake layers are anticipated to be thin, they may also be more compact. In addition, the accumulated foulants may include a lower concentration of hydrolytic coagulant products, which could result in greater irreversible fouling.

This review compiled the results of 36 studies on the impact of coagulation/flocculation pretreatment on floc properties and membrane performance. Despite the significant number of published studies, no clear guidance can yet be obtained to optimally design coagulation/flocculation pretreatment for membrane filtration.

Appendix

Table 8 Potential relationships between coagulation/hydrodynamic conditions, floc properties, and resistance for typical coagulation/flocculation configurations

	Type1: coagulation + no/incidental flocculation						Type 2: coagulation + flocculation						Type 3: conventional coagulation					
	Floc size		Fractal dimension		Resistance		Floc size		Fractal dimension		Resistance		Floc size		Fractal dimension		Resistance	
	Ref.	Cp	Ref.	Cp	Ref.	Cp	Ref.	Cp	Ref.	Cp	Ref.	Cp	Ref.	Cp	Ref.	Cp	Ref.	Cp
Coagulation conditions																		
Coagulant dose	\	\	\	\	+(9)	\	+(33, 36, 42)	+	+(33, 36, 42)	+	\	+	\	\	\	\	\	\
pH	\	\	\	\	■	■	+(36, 41, 43)	\	+(43)	\	■	■	\	\	\	+	■	■
Hydrodynamic conditions																		
$\bar{G}t$	\	\	\	\	■	■	+(22)	\	\	\	■	■	+(22)	\	\	\	■	■
Floc properties																		
Floc size	■	■	\	\	\	\	■	■	\	\	\	\	■	■	\	\	+(22)	\
Fractal dimension	■	■	■	■	\	\	■	■	■	■	\	\	■	■	■	■	\	\

Ref = observations from referenced studies. Cp = observations based on review of results compiled from several references. + = potential positive correlation. - = potential negative correlation. \ = inconclusive. ■ = not applicable.

Table 9 DOC and TOC removal for typical coagulation/flocculation configurations

Coagulant/dosage	Feedwater	Membrane type	DOC/TOC removal (%)	Ref.
No coagulant				
No coagulant	River 1.53 ± 0.41 mg L^{-1} DOC pH N/A[a]	MF PVDF 0.1 μm (nominal)	1.66 (DOC) 9.66 (DOC)	45
No coagulant	Reservoir 2.4 mg L^{-1} TOC pH ~5.4	MF Hydrophilic PES 0.1 μm (mean)	8 (TOC)	27
No coagulant	River 10.8 ± 0.8 mg L^{-1} DOC pH 8.14 ± 0.09	MF PVDF 0.1 μm (nominal)	18 (DOC)	39
No coagulant	Synthetic (HA,SA,BSA) 4 mg L^{-1} TOC pH 7.0±0.3	UF N/A 150 kDa MWCO	52.1 (TOC) 60.5 (TOC) 65.3 (TOC)	76
		UF PES 100 kDa MWCO	57.9 (TOC) 59.4 (TOC) 65.4 (TOC)	
No coagulant	River 2–6 mg L^{-1} DOC pH 7	UF CA 150 kDa MWCO	10 (DOC)	31
		UF PAN 200 kDa MWCO	18 (DOC)	
No coagulant	River 5.341–6.29 mg L^{-1} DOC pH 7.1–7.5	UF PVDF 150 kDa MWCO	7.5 (DOC)	48
No coagulant	Synthetic 9.43 mg L^{-1} TOC pH 5–10	UF PES 30 kDa MWCO	38.0 (TOC) 37.0 (TOC)	46
No coagulant	Canal 5.4 ppm DOC pH 5.5–7.5	UF Cellulose N/A	13 (DOC)	25
No coagulant	Natural 3.521 ± 1.423 mg L^{-1} DOC pH 7.1–7.3	UF PVDF 0.01 μm (nominal)	24.3 (DOC)	40
No coagulant	River 6.53 mg L^{-1} TOC pH 7.8–8.0	UF PES N/A	55.1 (TOC)	30
Type 1: coagulation + no/incidental flocculation				
Fe-based/0.018 mM (Fe)	Reservoir 2.4 mg L^{-1} TOC pH ~5.4	MF Hydrophilic PES 0.1 μm (mean)	50 (TOC)	27
0.027 mM (Fe)			46 (TOC)	
0.036 mM (Fe)			69 (TOC)	
0.045 mM (Fe)			42 (TOC)	
0.054 mM (Fe)			54 (TOC)	
0.072 mM (Fe)			75 (TOC)	
$FeCl_3$/5 ppm Fe	Canal 5.4 ppm DOC pH 5.5–7.5	UF Cellulose N/A	33 (DOC)	25
5 ppm Fe			36 (DOC)	
5 ppm Fe			27 (DOC)	
10 ppm Fe			42 (DOC)	
10 ppm Fe			33 (DOC)	
10 ppm Fe			16 (DOC)	
Alum/0.59–2.93 mg L^{-1} Al	Synthetic DOC N/A pH 4.81–8.73	UF PES 100 kDa MWCO	35 (TOC)	32

Table 9 *(continued)*

Coagulant/dosage	Feedwater	Membrane type	DOC/TOC removal (%)	Ref.
$FeCl_3$/2.8 mg L^{-1} Fe	River	UF	44.2 (TOC)	30
$Fe_2(SO_4)_3$/2.8 mg L^{-1} Fe	6.53 mg L^{-1} TOC	PES	59.1 (TOC)	
$Al_2(SO_4)_3$/3.6 mg L^{-1} Al	pH 7.8–8.0	N/A	57.9 (TOC)	
1 mg L^{-1} Al	River	UF	25 (DOC)	31
5 mg L^{-1} Al	2–6 mg L^{-1} DOC	CA	35 (DOC)	
7 mg L^{-1} Al	pH 5.5–7.5	150 kDa MWCO	38 (DOC)	
5 mg L^{-1} Al		UF	62 (DOC)	
5 mg L^{-1} Al		PAN	49 (DOC)	
5 mg L^{-1} Al		200 kDa MWCO	41 (DOC)	
Type 2: coagulation + flocculation				
PACl/15 mg L^{-1}	River	MF	61 (DOC)	39
	10.8 ± 0.8 mg L^{-1} DOC	PVDF		
	pH 8.14 ± 0.09	0.1 μm (nominal)		
PACl/0.025 mM (Al)	Synthetic	UF	35.32 (DOC)	33
0.1 mM (Al)	5 mg L^{-1} HA	PES	70.2 (DOC)	
	pH 7.5	100 kDa MWCO		
Alum/0.06 mM Al	Natural	UF	35.8 (DOC)	40
	3.521 ± 1.423 mg L^{-1} DOC	PVDF		
	pH 7.1–7.3	0.01 μm (nominal)		
Sedimentation				
$Al_2(SO_4)_3$/3.59 mg L^{-1} Al	Synthetic	UF	42.1 (TOC)	46
PAC10WA/3.59 mg L^{-1} Al	9.43 mg L^{-1} TOC	PES	44.1 (TOC)	
$NaAlO_2$/3.59 mg L^{-1} Al	pH 5–10	30 kDa MWCO	22.0 (TOC)	
PACl/14 and 28 mg L^{-1}	River	UF	38 (DOC)	47
	4.5 mg L^{-1} DOC	PVDF	51 (DOC)	
	pH N/A	0.02 μm (nominal)		
PACl/6 mg L^{-1}	Synthetic	UF	36 (DOC)	77
	4.104 ± 0.043 mg L^{-1} DOC	PES	31 (DOC)	
	pH 8.12	100 kDa MWCO	32 (DOC)	
			41 (DOC)	
			35 (DOC)	
			37 (DOC)	
Type 3: conventional coagulation				
Al-based/20 mg L^{-1}	River	MF	25.3 (DOC)	45
	1.53 ± 0.41 mg L^{-1} DOC	PVDF		
	pH N/A	0.1 μm (nominal)		
PACl/15 mg L^{-1}	River	MF	50 (DOC)	39
	10.8 ± 0.8 mg L^{-1} DOC	PVDF		
	pH 8.14 ± 0.09	0.1 μm (nominal)		
$Al_2(SO_4)_3$/3.59 mg L^{-1} Al	Synthetic	UF	66.2 (TOC)	46
PAC10WA/3.59 mg L^{-1} Al	9.43 mg L^{-1} TOC	PES	58.9 (TOC)	
$NaAlO_2$/3.59 mg L^{-1} Al	pH 5–10	30 kDa MWCO	37.0 (TOC)	
$Al_2(SO_4)_3$/3.59 mg L^{-1} Al		UF	65.0 (TOC)	
PAC10WA/3.59 mg L^{-1} Al		Cellulose	51.0 (TOC)	
$NaAlO_2$/3.59 mg L^{-1} Al		30 kDa MWCO	28.0 (TOC)	
PACl/14 and 28 mg L^{-1}	River	UF	51 (DOC)	47
	4.5 mg L^{-1} DOC	PVDF		
	pH N/A	0.02 μm (nominal)		
Alum/4 mg L^{-1}	River	UF	20.5 (DOC)	48
	5.341–6.29 mg L^{-1} DOC	PVDF		
	pH 7.1–7.5	150 kDa MWCO		

[a] N/A = not available.

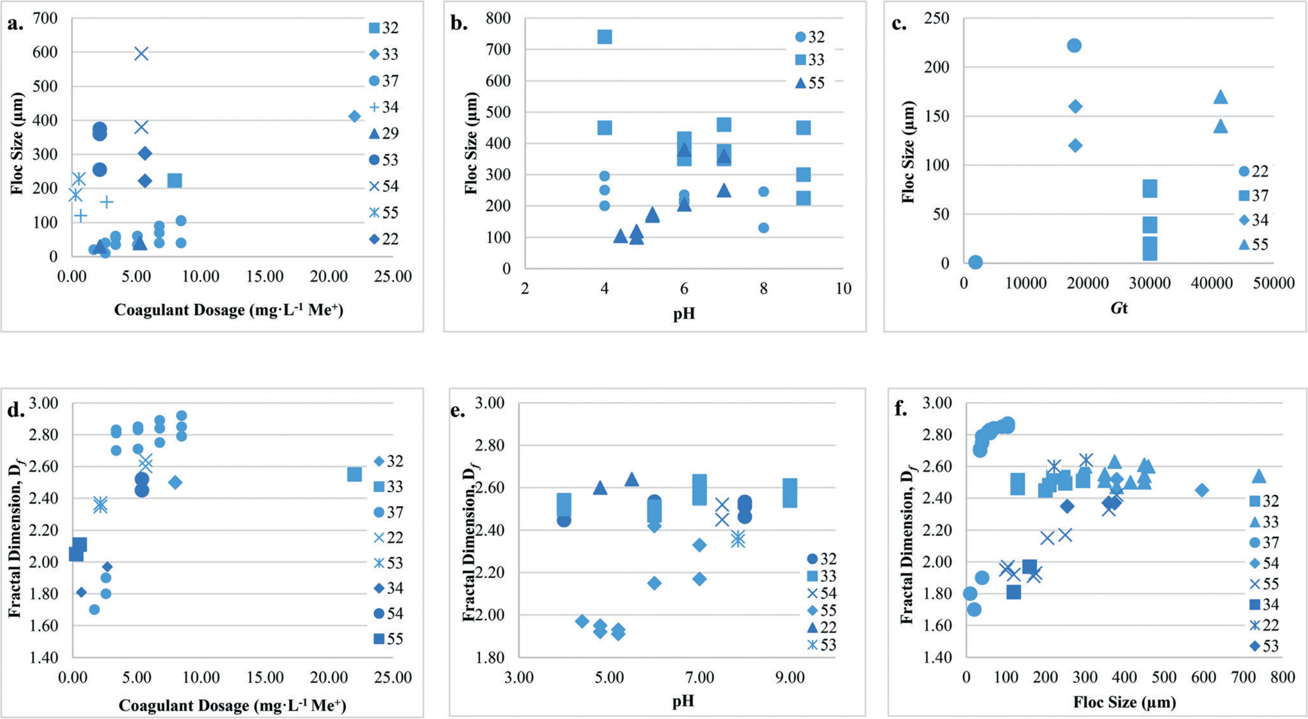

Fig. 7 Type 2: coagulation + flocculation a. floc size *vs.* coagulant dosage, b. floc size *vs.* pH, c. floc size *vs.* $\bar{G}t$, d. fractal dimension *vs.* coagulant dosage, e. fractal dimension *vs.* pH, and f. fractal dimension *vs.* floc size.

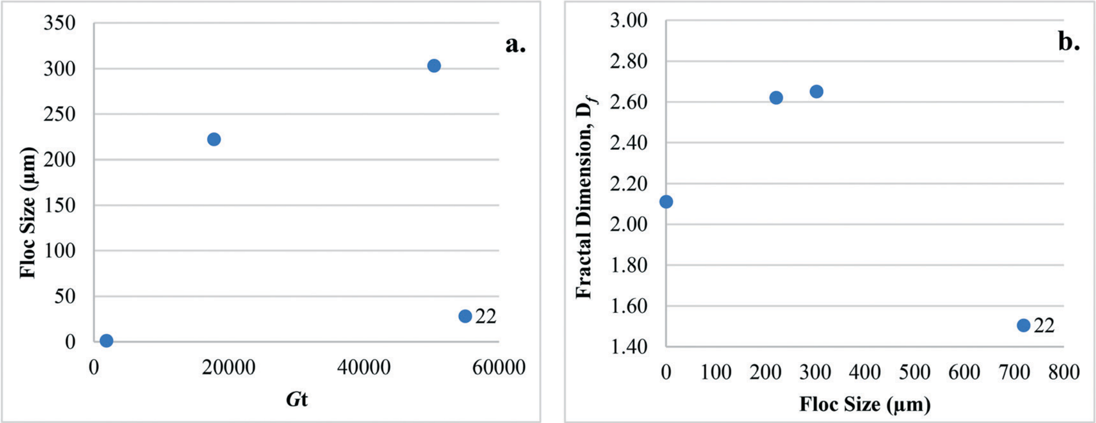

Fig. 8 Type 3: conventional coagulation a. floc size *vs.* $\bar{G}t$, b. fractal dimension *vs.* floc size.

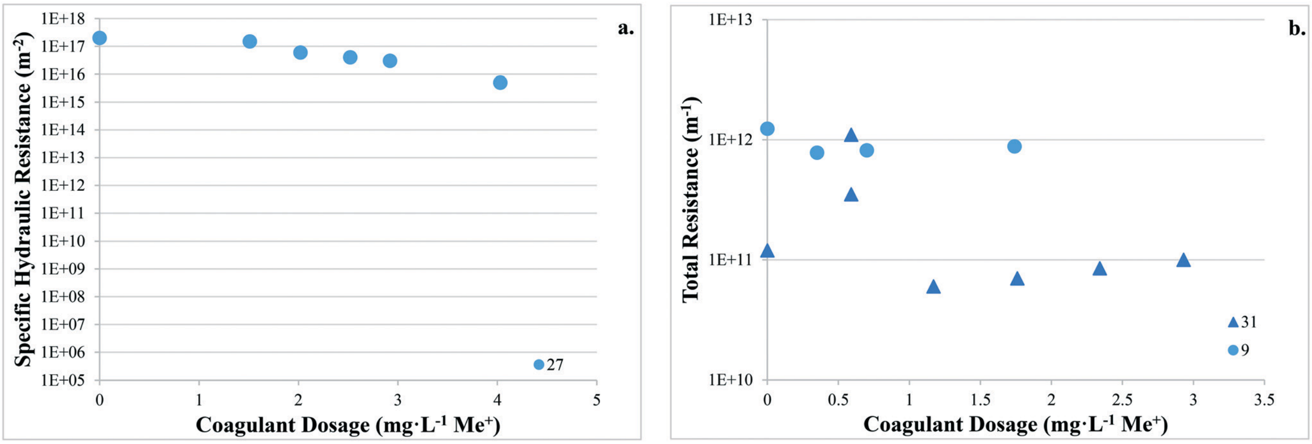

Fig. 9 Type 1: coagulation + no/incidental flocculation a. specific hydraulic resistance *vs.* coagulant dosage (MF), and b. total resistance *vs.* coagulant dosage (UF).

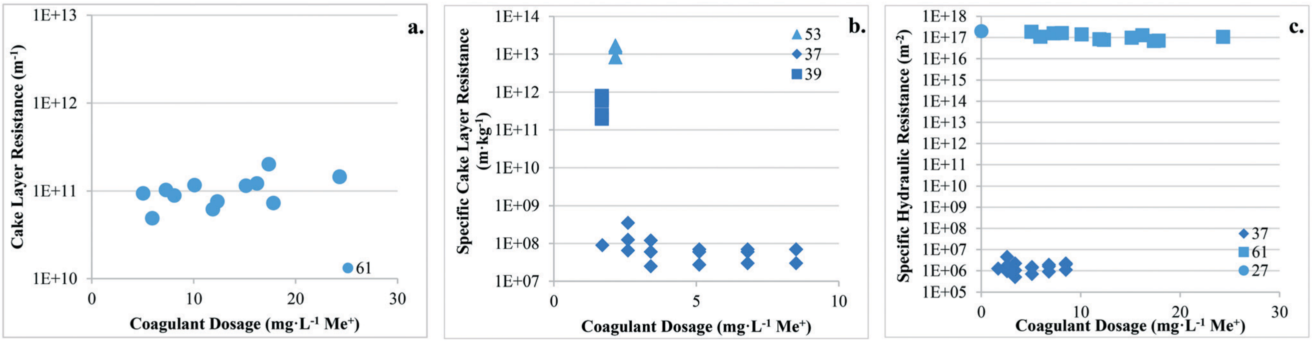

Fig. 10 Type 2: coagulation + flocculation a. cake layer resistance *vs.* coagulant dosage (MF), b. specific cake layer resistance *vs.* coagulant dosage (MF), and c. specific hydraulic resistance *vs.* coagulant dosage (MF).

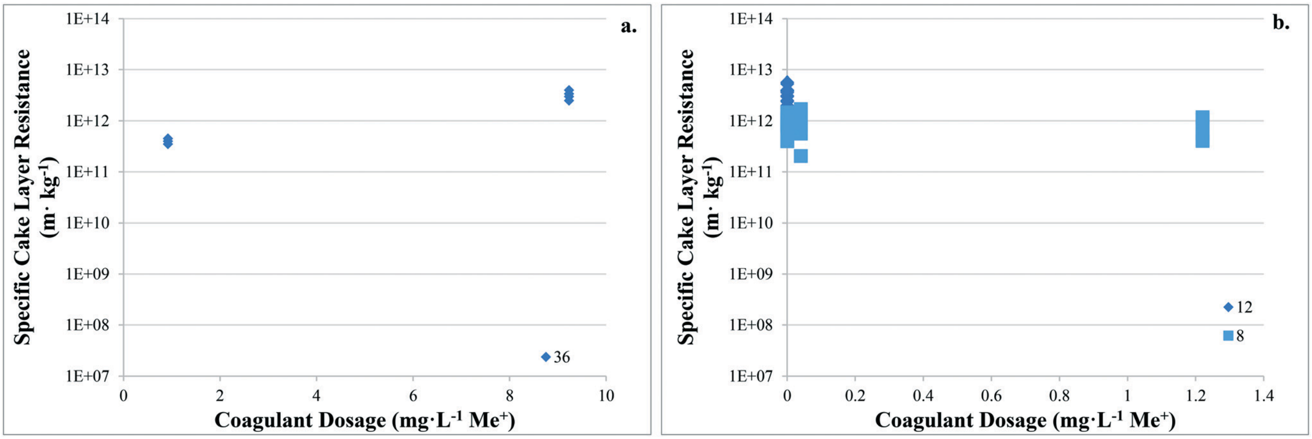

Fig. 11 Type 3: conventional coagulation a. specific cake layer resistance *vs.* coagulant dosage (MF), b. specific cake layer resistance *vs.* coagulant dosage (UF).

Conflicts of interest

There are no conflicts of interest to declare.

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