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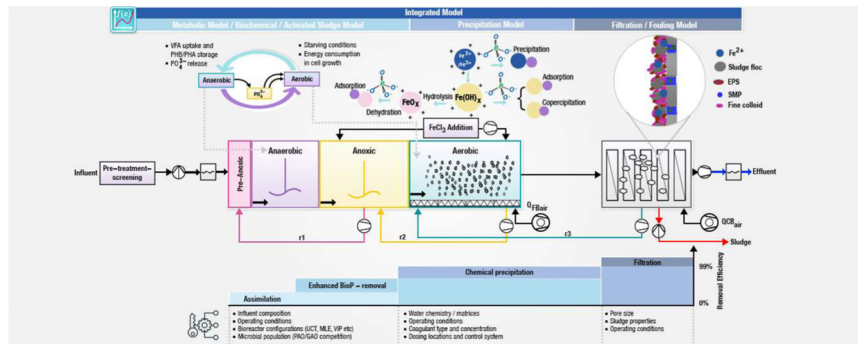
Modeling, simulation and control of biological and chemical P-removal processes for membrane bioreactors (MBRs) from lab to full-scale applications: State of the art

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- P-removal employing combined EBPR and chemical precipitation are successful in MBRs.
- Bio-kinetic models appeared to be successful in simulating the P-removal in MBRs.
- Specificities of MBR functioning require peculiar parameters for these models.
- EBPR instability at full-scale due to in-comprehension of micro-organisms role.
- P-removal modeling studies at full/super-large scale MBRs are required.



ABSTRACT

Phosphorus (P) removal from the domestic wastewater is required to counter the eutrophication in receiving water bodies and is mandated by the regulatory frameworks in several countries with discharge limits within 1-2mgPL⁻¹. Operating at higher sludge retention time (SRT) and higher biomass concentration than the conventional activated sludge process (CASP), membrane bioreactors (MBRs) are able to remove 70–98% phosphorus without addition of coagulant. In full-scale facilities, enhanced biological phosphorus removal (EBPR) is assisted by the addition of metal coagulant to ensure >95% P-removal. MBRs are successfully used for super-large-scale wastewater treatment facilities (capacity >100,000 m³d⁻¹). This paper documents the knowledge of P-removal modeling from lab to full-scale submerged MBRs and assesses the existing mathematical models for P-removal from domestic wastewater. There are still limited studies involving integrated modeling of the MBRs (full/super large-scale), considering the complex interactions among biology, chemical addition, filtration, and fouling. This paper analyses the design configurations and the parameters affecting the biological and chemical P-removal in MBRs to understand the P-removal process sensitivity and their implications for the modeling studies. Furthermore, it thoroughly reviews the applications of bio-kinetic and chemical precipitation models to MBRs for assessing their effectiveness with default stoichiometric and kinetic parameters and the extent to which these parameters have been calibrated/adjusted to simulate the P-removal successfully. It also presents a brief overview and comparison of seven (7) chemical precipitation models, along with a quick comparison of commercially available simulators. In addition to advantages associated with chemical precipitation for P-removal, its role in changing the relative abundance of the microbial community responsible for P-removal and denitrification and

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the controversial role in fouling mitigation/increase are discussed. Lastly, it encompasses several coagulant dosing control systems and their applications in the pilot to full-scale facilities to save coagulants and optimize the P-removal performance.

Contents

1.	Introduction	0
1.1.	Phosphorus removal regulation context	0
1.2.	Phosphorus in wastewater and its removal by treatment processes.	0
1.3.	Scientific context	0
2.	Biological phosphorous removal processes and its modeling	0
2.1.	Enhanced biological P-removal (EBPR) in MBRs	0
2.2.	MBR configurations and treatment sequence for P-removal	0
2.3.	Factors affecting the BioP-removal in MBR	0
2.3.1.	Feed characteristics	0
2.3.2.	MLSS concentration and P-removal in MBR	0
2.3.3.	Microbial communities and P-removal in MBRs	0
2.3.4.	Sludge retention time and P-removal in MBRs	0
2.3.5.	Hydraulic retention time and P-removal in MBRs.	0
2.3.6.	Effect of temperature on P-removal in MBRs.	0
2.3.7.	Dissolved oxygen concentration and P-removal in MBR.	0
2.4.	Mathematical modeling of MBRs	0
2.5.	Modeling and simulation of BioP-removal in MBRs	0
2.5.1.	Calibration of models	0
2.5.2.	MBR simulation studies	0
2.5.3.	Wastewater simulation software	0
3.	Chemical P-removal modeling and control	0
3.1.	Phosphorus precipitation process	0
3.2.	Factors affecting chemical precipitation for phosphorous	0
3.2.1.	Influent chemistry and composition.	0
3.2.2.	Influence of operational conditions	0
3.2.3.	Type of coagulant, molar ratio, and dosing location	0
3.2.4.	Mixing conditions.	0
3.3.	Chemical P-removal and membrane fouling	0
3.4.	Modeling chemical precipitation of phosphorous	0
3.4.1.	Equilibrium-based models	0
3.4.2.	Models based on empirical kinetics	0
3.4.3.	Models based on combined chemical equilibrium-kinetic approaches	0
3.4.4.	Models based surface complexation and chemical equilibrium.	0
3.5.	Application of chemical precipitation models to MBRs	0
4.	Control systems for P-removal	0
5.	Knowledge gaps and future research directions	0
6.	Conclusion	0
	IWA standard nomenclature for model parameters.	0
	Credit authorship contribution statement	0
	Declaration of competing interest.	0
	Acknowledgments	0
	References	0

Nomenclature

ANO	autotrophic nitrite-oxidizing bacteria;
AOB	ammonia oxidizing bacteria;
A/O	anoxic-oxic;
A ² /O	anaerobic–anoxic-oxic;
AnMBR	anaerobic MBR;
ASMs	activated sludge models;
ATP	adenosine triphosphate;
AFO	amorphous ferric oxyhydroxide;
ASF	active site factor
ANN	artificial neural network;
BOD	biological oxygen demand;
BEMR	bioelectrical membrane bioreactor;
BNR	biological nutrient removal;

BAF	biologically aerated filter
CASP	conventional activated sludge process;
CPP	chemical precipitation process;
C/N	carbon to nitrogen ratio;
COD	chemical oxygen demand;
DAF	dissolved air floatation;
DNN	deep learning neural network;
DNN-GP	dual N/denitrification (Gao-PAOs)
DO	dissolved oxygen;
DPAOs	denitrifying PAOs;
eFAST	extended fourier amp. Sensitivity test
EPS	extra polymeric substances;
EU	European Union;
EEM	excitation emission matrix;

EQI	effluent quality index;
EBPR	enhanced biological P-removal;
F/M ratio	food to micro-organism ratio;
FTIR	Fourier transformed infrared spectrum;
GAOs	glycogen accumulating organisms;
GFC	gel-filtration chromatography;
HRT	hydraulic retention time;
HF	hollow fiber;
HSG	hochschulgruppe;
HFO	hydrous ferric oxides;
KOH	DO half-saturation coefficient;
MeP	metal phosphate
IWA	International Water Association
MeOH	metal hydroxide
MBR	membrane bioreactor
MLSS	mixed liquor suspended solids
MT	multi-tubular
MABR	membrane aerated bioreactor
MMBR	microalgae membrane bioreactor
MLE	modified Luzack-Ettinger
ML	mixed liquor
NOB	nitrite-oxidizing bacteria
OHO	ordinary heterotrophic organisms
OH ⁻	hydroxyl radical
OCI	operating cost index
PO ₄ ³⁻	orthophosphate
PI	proportional integral
PID	proportional-integral-derivative
PFC	polymeric ferric chloride
PAOs	phosphate accumulating organisms
PHA	polyhydroxyalkanoates
PHB	poly-hydroxybutyrate
PSD	particle size distribution
RM	real municipal wastewater
RIS	resistance in series
sMBR	submerged MBR
SCM	surface complexation model
SRT	sludge retention time
SMP	soluble microbial products
SAM	sequencing anoxic-anaerobic membrane
SEM	scanning electron microscope
SM	synthetic municipal wastewater
SRC	standardized regression coefficient
SCADA	supervisory control & data acquisition
TP	total phosphorous
TSS	total suspended solids
TMP	transmembrane pressure
TUDP	Technical University of Delft phosphorus
UCT	University Cape Town
VIP	Virginia institute process
VFA	volatile fatty acids
WWTPs	wastewater treatment plants(s)
WEF	Water Environment Federation

1. Introduction

1.1. Phosphorus removal regulation context

Rapid urbanization, coupled with industrial and agricultural sector growth, has increased nutrients in the effluent of wastewater treatment plants (WWTPs) and thus in the receiving water bodies. Higher nutrients concentrations exceeding the minimum permissible limits, among which phosphorus, is largely responsible for eutrophication. It deteriorates the aquatic environment due to excessive algal growth

and depleting oxygen, making water habitat unsuitable for marine life. Depending upon the flow and fraction of extraneous domestic wastewater, phosphorous (P) concentrations range between 1 and 20 mgP_{tot}L⁻¹, and vary seasonally from region to region (Gray, 2004; Henze et al., 2008; Sayi-Ucar et al., 2015). Release of phosphorus above 0.1–0.2 mgP_{tot}L⁻¹ in running water and 0.005–0.01 mgP_{tot}L⁻¹ in stagnant water is highly assistive for eutrophication (Omwene et al., 2018; Rahman et al., 2016). Therefore, majority of the countries have set their national regulatory discharge limits in the range of 1–2 mgP_{tot}L⁻¹ with few exceptions such Japan and Belarus (Table A.1). Generally, effluent discharge limits are set based upon total phosphorus (TP) concentration. Since eutrophication is predominantly caused by the bioavailable fraction of phosphorus (phosphate, PO₄³⁻), it is necessary to determine the permissible values of these bioavailable forms (Preisner et al., 2020). Keeping this in view, it is anticipated that many countries will introduce more stringent regulatory measures in the upcoming years, and the existing WWTPs might require upgradation to meet the required discharge limits.

1.2. Phosphorus in wastewater and its removal by treatment processes

The primary sources of phosphorus in the domestic wastewater are households (van Puijenbroek et al., 2019) with human excreta contributing about 30–50% while Gomes de Quevedo and da Silva Paganini (2016) reported that detergents are the main contributors with 20–80% of the phosphorus load. Generally, TP in wastewater is classified based upon its physical characteristics into soluble (can pass through 0.45 mm filter) and particulate fractions (Gu et al., 2011). Different analytical approaches have been developed for better characterizing the phosphorus including colorimetry, digestion, acid hydrolysis and particle size distribution (Gu et al., 2011; van Nieuwenhuijzen et al., 2004). In order to better choose the technology, unit operations and appropriate configurations for TP removal from the wastewater, a comprehensive understanding of the phosphorus fractions is essential (Gu et al., 2011; Yu et al., 2021). In a detailed fractionation approach, TP can be studied under a total of 17 fractions. Among these, 6 can be directly measured from wastewater samples while the remaining 11 could be calculated from the results of 6 different analyses (Gu et al., 2011; Reynolds & Davies, 2001) as depicted in Fig. 1. There is no standardized and universally accepted approach for classifying the phosphorus fractions and there is urgent need to address this issue along with the standardizing notations of each fraction and corresponding testing method (Rosario et al., 2021). For convenience, TP is generally classified into three major classes i.e. orthophosphate (PO₄³⁻), polyphosphate (Poly-P) and organic phosphate. According to Rossle and Pretorius (2001), orthophosphate (PO₄³⁻) is the most abundant in domestic wastewater and constitute about 70–90% in the raw and settled wastewater while other fractions are limited to 10–30% of the total phosphorus.

The phosphorus concentration in raw municipal wastewater tends to change due to lifestyle, urbanization, and industrial development. It has been estimated that around 3 million tons (Mt)/annum is lost as human waste, and only 1.3 Mt /year is treated by the WWTPs (Van Vuuren et al., 2010). Different technologies target a particular fraction of the phosphorus via chemical, biological, or combined treatment processes (Gu et al., 2011). Majority of the commercially available technologies can remove about 30% of the BioP, but high costs remain a major challenge (Ramamahayam et al., 2014). Furthermore, Phosphorus is a limited and non-renewable mineral resource. Globally, around 20 Mt/annum of phosphorus is mined, and due to this increasing consumption, phosphorus availability is projected to hit the low availability limits by the next 50–100 years, and the peak is expected to occur by 2034 (Azizi, 2018; Cordell et al., 2009). This emphasizes the recovery of phosphorus from the wastewater, which is estimated to be sufficient to meet 15–20% of the global phosphorus demand (Cordell et al., 2009). Keeping this view, Sweden has introduced regulatory criteria to recover 75% of the phosphorus from the WWTPs (Bashar et al., 2018).

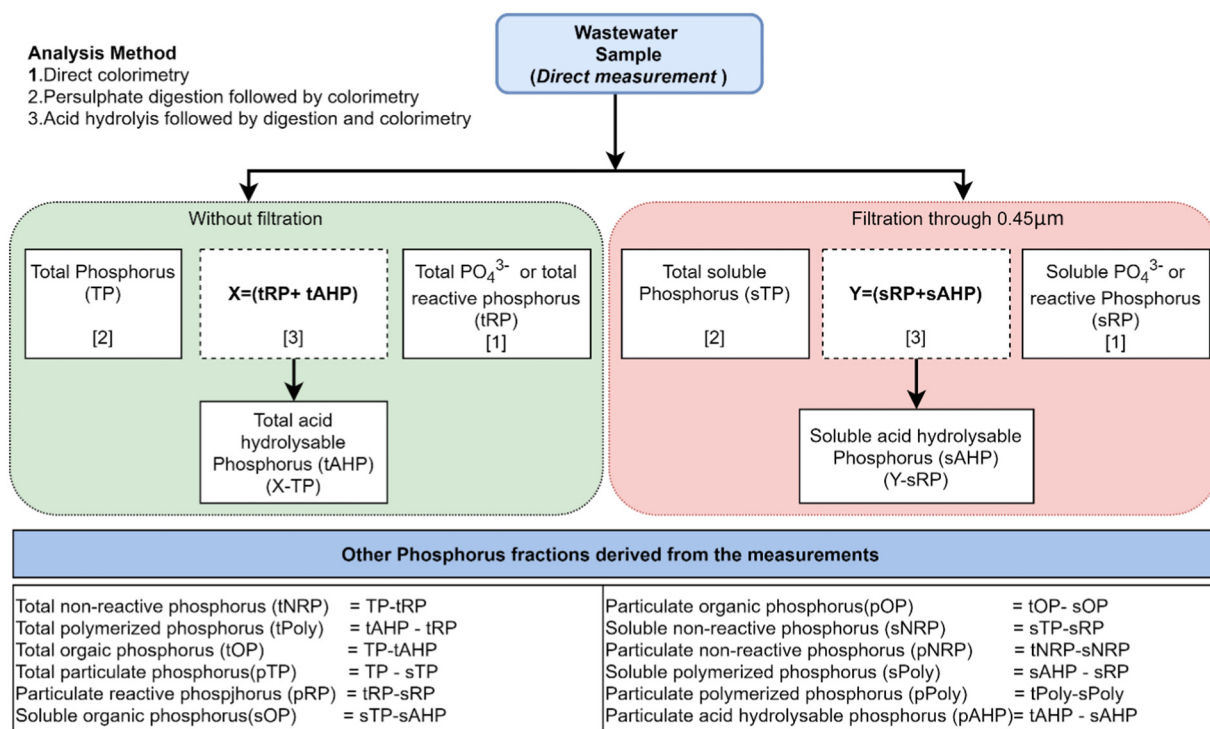


Fig. 1. Fractions of phosphorus in wastewater (modified from Gu et al., 2011).

Phosphate removal before discharging can be accomplished using various treatment methods such as physio-chemical, biological, and combinations thereof (Hai et al., 2018; Henze et al., 2008). Biological treatment is commonly used for domestic wastewater treatment at large-scale WWTP. Generally, BioP-removal efficiencies varied from 58.2% to 93.9% for full-scale EBPR facilities (Bunce et al., 2018; Zhang et al., 2011) and this large variations is mainly due to the varying high COD/P ratio (28.6–196.2) in the domestic influents. While, for bench scale studies the P-removal efficiencies are reported up to 99% (Bunce et al., 2018; Denisova et al., 2020).

In the last two decades, MBRs have extensively been adopted in wastewater treatment for municipal and industrial applications (Judd, 2010; Krzeminski et al., 2017). The MBR system is superior over conventional activated sludge process (CASP) regarding its compactness (up to 50% less footprint), robustness, better and consistent permeate quality with excellent solids retention (less sludge production), increased volumetric loading due to operation at higher sludge retention time (SRT), independence from hydraulic retention time (HRT), better disinfection, biomass enrichment and flexibility of the process (Hai et al., 2018; Judd, 2010; Xiao et al., 2019). According to Meng et al. (2012), there were more than 2500 MBRs operating globally by 2013 and number was growing with a growth rate of 10.5%. As of today, globally there are around 62 super-large scales (>100,000 m³d⁻¹) MBR facilities contributing to the treatment of more than 10 million m³.day⁻¹ of wastewater (Xiao et al., 2019). Given its commercial success, the global size of the MBR market was valued at US\$ 3.09 billion in 2020 and is forecasted to reach US\$ 5.48 billion by 2028 at a compound annual growth rate of 7.02% (Emergen, 2021). The present article aims to establish a state of the art of the modeling, simulation and control of super-large scale MBR for P-removal.

1.3. Scientific context

Literature screening in the current work found that the earlier reviews on P-removal are focused on either identifying the factors affecting P-removal in wastewater (Mulkerriens et al., 2004), micro-organisms and pathways involved (Ahmed, 2012), or emerging technologies (Guo

et al., 2014). Similarly, very few reviews are dedicated to assessing the applicability of the ASMs (modified/unmodified) for nutrient removal in full-scale CASP/MBRs (Fenu et al., 2010; Naessens et al., 2012a, 2012b; Ng and Kim, 2007). The Scopus database was explored with key terms for the P-removal in MBRs and for modeling studies conducted from 2000 onwards. A total of 240 publications were found, including 18 publications related to P-removal modeling in MBRs. Each of these publications was further analyzed, as depicted in Fig. 2, constituting the basis of this review article. Furthermore, also based on Scopus data, 442 patents have been granted in the domain of P-removal in submerged MBR and 82% of these are registered in the US. They are reported in Fig. 2. Their number evolution is similar to the one of publications.

Most of these reviews are focused on CASP and/or general P-removal in wastewater, and none of these reviews is comprehensive enough to address all aspects of biological and chemical P-removal and its modeling in MBRs. Modeling approaches have been used to optimize MBRs ranging from the lab scale to full scale (Ferrero et al., 2011; Verrecht et al., 2008). However, very few publications have been dedicated to assessing and optimizing full scale MBR plants for P-removal in particular. There is no published study devoted to modeling and optimizing the super-large scale MBR for P-removal.

Based on the articles considered in this review, the MBRs are able to achieve 70 – 99 % P-removal with optimum operational settings and by maintaining sufficient substrate for the phosphorus accumulating organisms (PAOs) for their normal growth (Abegglen et al., 2008; Ersu et al., 2010; Silva et al., 2012; Smith et al., 2014). Although, better performance is reported for the MBR systems as compared to CASP but P-removal in MBRs is still difficult (especially in wastewater with low F/M ratio) because of its functional specificities (e.g., longer SRT and decoupled HRT). It is also expensive as compared to CASP due to excessive recirculation between aerobic/anoxic and anaerobic zones (mixing in the anaerobic zone) requiring extra energy and thus additional operational cost (Daigger et al., 2010; Lee et al., 2009; Sun et al., 2013). A recent study conducted at full-scale MBR facilities estimated P-removal specific energy consumption as high as 71.0 kWh(kgP_{tot})⁻¹ (Wang, 2020). The cost of P-removal in MBR is higher than CASP mainly

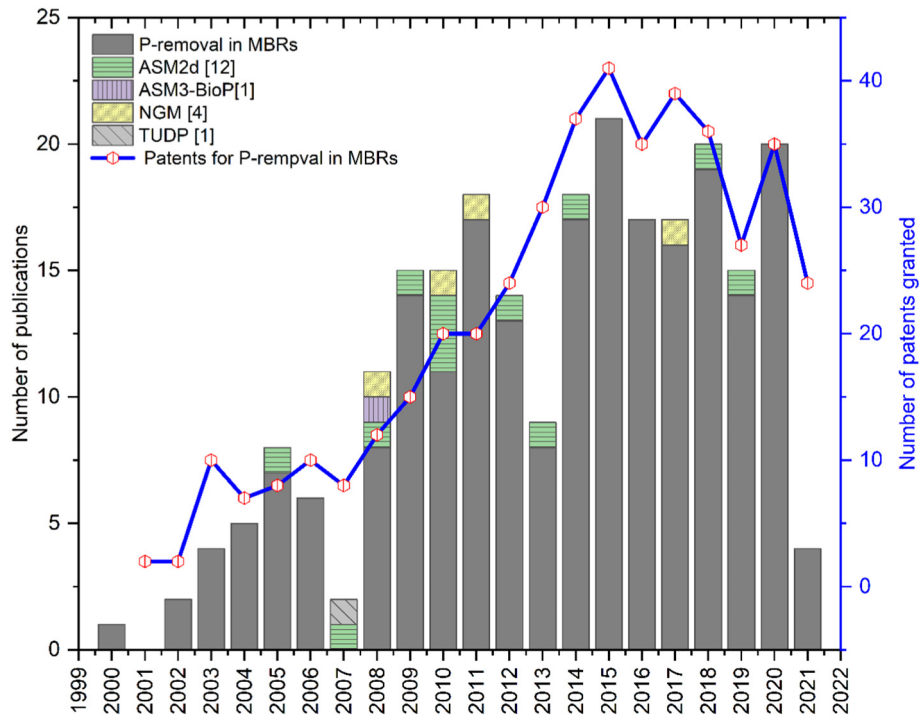


Fig. 2. Number of publications related to P-removal and modeling in MBRs.

due to higher aeration. The average removal cost of 1 kg P-removal in MBR varies from 39.8 to 480 US\$ depending on influent load, plant configuration, aeration intensity and removal efficiencies. While, in CASP it costs between 13.30 and 101 US\$/kg (Arif et al., 2020; Bashar et al., 2018; Iglesias et al., 2017). Generally, the super-large-scale MBR plants are operated as per the manufacturers' recommendations, and there has been limited research on their optimization for the nutrient removals.

This review focuses on assessing the applications of the existing mathematical models for biological and chemical P-removal in CASP and their applicability in MBRs ranging from pilot to full-scale. Section 2 includes a brief summary of the BioP-removal process in MBR compared to CASP, followed by an in-depth discussion on the role of various reactor configurations and their P-removal performance along with detailed discussion on factor affecting the P-removal in MBRs. Section 2.5 dedicated for assessing the applicability of the existing mathematical models for biological phosphorus removal. It also presents a summary of the MBRs specificities to be considered for their modeling. Furthermore, section 3 presents chemical P-removal process, factors affecting the chemical P-removal and its modeling using the existing models. The role of coagulants as a medium for chemical P-removal and fouling development or mitigation is also discussed in this section. Section 4 is dedicated to the role of various control systems used to optimize the P-removal in CASPs and MBR and corresponding operational cost reduction. Finally, the article concludes along with knowledge gaps and future research directions.

2. Biological phosphorous removal processes and its modeling

2.1. Enhanced biological P-removal (EBPR) in MBRs

P-removal process involves inducing phosphate incorporation into total suspended solids (TSS) and removing them via precipitation or filtration (Kim and Chung, 2014). Usually, a P_{tot}/TSS fraction of 2–5% is essential for BioP-removal in CASP, while this can be as high as 6–10% in MBR (Abegglen et al., 2008; Adam et al., 2002; Choi et al., 2011). This incorporation is generally accomplished through biological and chemical processes and combinations (Wilfert et al., 2015). No sharp limits

exist between these methods to differentiate their performance when used in conjunction. For example, it has been observed that during the EBPR process, a significant part of the compound is chemically precipitated by the action of metal ions that were natural constituents of the wastewater (Wang et al., 2014). A carefully designed enhanced biological P-removal (EBPR) process may remove over 85–99% of phosphorus (i.e. Adam et al., 2003; Ramasahayam et al., 2014) and could even reduce the phosphorous concentration to less than 0.1 mgPL^{-1} with the addition of external carbon source (Gnirss et al., 2003; Henze et al., 2008). The chemical precipitation processes can enhance the treatment efficiency of the biological WWTPs (Adam et al., 2003; Henze et al., 2008). However, it has the disadvantages of high chemical costs, chemical handling, storage requirements, increased chemically enriched sludge production, and subsequent sludge handling and disposal costs (Bunce et al., 2018; Chae et al., 2015). Stringent regulatory pressure and tight discharge limits have forced the WWTPs manufacturers and operators to develop and deploy efficient technologies to reduce the phosphorous discharge into water bodies (Gu et al., 2011; Vohla et al., 2011; Wadood and Sarmad, 2012).

MBRs can sufficiently remove the nutrient from the domestic wastewater and usually provide better results than CASP. Combining the EBPR with the MBR process reduces the reactor volume while achieving similar P-removal performance (Adam et al., 2003; Judd, 2010). MBR with 100% solids retention and chemical precipitation features could be a useful technology for phosphorus recovery, and further research should be dedicated to exploring the opportunities for P recovery. The understanding of the MBR functioning for the nutrient removal (P and N) is essential to design and optimize the P-removal through model based approaches. The experimental studies about the P-removal performance (along with the N removal) help to understand the process knowledge and to prepare a set of guidelines for a better adaptation of the available phosphorous models for the MBR.

2.2. MBR configurations and treatment sequence for P-removal

Traditionally, MBR is configured as submerged/immersed or side stream. In a submerged/immersed (sMBR) system, a membrane module

is installed within the bioreactor, and the treated effluent is withdrawn via negative pressure/vacuum. Air scouring is typically used to counter the cake layer formation, which can further be assisted through additional means such as filtration-relaxation sequencing, aeration sequencing and backwashing (Burman and Sinha, 2018; Hai et al., 2018). While, for the side stream configuration, the membrane is external to the bioreactor, and cake layer formation is mainly inhibited due to cross-flow-velocity (Hai et al., 2018; Judd, 2010). Novel configurations of the sMBR are based on an anti-fouling mechanism, such as a fixed-film membrane aerated biofilm reactor (MABR), bio-electrochemical MBR (BEC-MBR) or electrically-induced MBRs, microalgae membrane bioreactor (MMBR) which are yet being explored at the lab/pilot scale have not been included in this review. This review only focuses on sMBR along with the reactor configurations adopted for P-removal.

Several reactor configurations involving plug flow or strict compartmentalization, such as modified Ludzack-Ettinger (MLE), anoxic-oxic (A/O), anaerobic-anoxic-oxic (A²/O), University Cape Town (UCT), modified UCT, Virginia institute process (VIP), and Bardenpho/modified Bardenpho with multiple stages processes, have been tried and tested for nutrient removal (Ahn et al., 2003; Ersu et al., 2008; Ersu et al., 2010; Hai et al., 2018; Monclús et al., 2010; Vaiopoulou et al., 2007; Zhang, 2020). The BioP-removal performance in these configurations mainly depends upon how the anaerobic reactor is maintained and protected from the nitrates interferences by controlling the internal recirculation. Table B.1 provides a brief description of each of the BioP-removal system configuration along with its advantages and disadvantages (Daigger et al., 2010; Fleischer et al., 2005; Hai et al., 2018). The roles of various zones involved in these configurations and dominating fractions of the microbial communities in these zones is summarized in Table B.2. The following section is dedicated to assess the performance of different configurations for BioP-removal in MBRs with capacity ranging from 0.01 to 45,000 m³.d⁻¹.

Recycling oxygen-saturated mixed liquor from the membrane zone to the unaerated zone (anoxic/anaerobic) negatively impacts the denitrification process. Thus, the abundance of nitrates reduces the P-removal performance since the occurrence of anaerobic conditions is less likely at higher recirculation ratios. An increase in the recirculation ratio leads to higher effluent NO₃-N and PO₄³⁻ concentrations (Fleischer et al., 2005). This is due to the fact that denitrification and P-removal processes compete for the same carbon source as demonstrated by another study (Maere et al., 2011). Different recirculation arrangements and rates from the MBR tank to upstream reactors result in lower/higher MLSS upstream of the membrane tank, also responsible for nutrients removal (Dold et al., 2010). In the last 2 decades, several attempts have been made to improve the BioP-removal efficiency of MBR by modifying the conventional configurations and developing the novel ones with focus on: i) reduced nitrates interferences ii) reduced recirculation iii) simultaneous denitrification and EBPR and iv) reduced overall footprint of the reactor.

Early studies of Adam et al. (2003) and of Lesjean et al. (2003) evaluated the effect of pre and post denitrification on P-removal in UCT configuration. A lab-scale study of Adam et al. (2003) found similar P-removal (99% with coagulant addition) performances in both cases while lower recirculation was required in post-denitrification mode from MBR to aerobic zone. Similarly, the pilot-scale studies of Lesjean et al. (2003, 2005) found identical P-removal performances in both pre- and post-denitrification modes without external carbon source addition. The recirculations in the study of Lesjean et al. (2003) were same as of Adam et al. (2003) and in both studies precipitation was used to lower the effluent phosphorus below 0.1 g·m⁻³. Another pilot scale study (Fleischer et al., 2005) compared the nutrient removal performance of three configurations with multi-stage Bardenpho process. The 5-stage configuration was comparatively effective in BioP-removal, however it was unable to meet the required effluent discharge standard without addition of coagulant. The recirculation of oxygen-saturated mixed-liquor from the MBR tank to an anaerobic zone

through the aerobic zone proved successful in improving P-removal. However, the proposed configuration involved multiple recirculation streams associated with higher flow rates, resulting in increased footprint and recirculation cost. In another study dedicated to evaluating different MBR configurations, Ersu et al. (2008) evaluated five different configurations (three modified A²/O and two A/O). The modified A²/O configuration with 300% recirculation of the MBR sludge (3 times the influent flow) to anaerobic zone and 100% permeate recirculation to anoxic resulted in higher P-removal efficiency (> 88%). However, recirculation of the permeate may not be an economical choice since energy consumption in MBR is already slightly higher than in CASPs (Krzeminski et al., 2017; Xiao et al., 2019). Another study (Lee et al., 2009) evaluated the UCT and modified UCT (with step feed to anoxic zone) configurations with weak domestic wastewater (low F/M ratio) and found that modified UCT-configuration provided better P-removal (> 70%) than UCT (40%). The authors concluded that feeding weak wastewater directly to the anoxic zone provided enough substrate to the denitrifiers to remove nitrates, and this resulted in improved P-removal performance of the MBR. Based upon the findings of these two full scale facilities with 4–5 stages Bardenpho process, Daigger et al. (2010) provided guidelines for designing MBR configurations to achieve the P-removal below regulatory level including: i) membrane recirculation flow should be directed to the aerobic zone, ii) intense mixing at the inlets of the anaerobic and anoxic zones, iii) consistent internal recirculation flow rates to maintain the desired MLSS distribution and iv) carefully controlled metal salt addition in proportion to the phosphorus remaining after biological removal. The study of Holba et al. (2012) presented a comparative assessment of three MLE configurations. The highest P-removal efficiency was achieved with 2-stage MLE having single internal recirculation from aerobic to anoxic zone and with addition of coagulant. The study of Corsino et al. (2020) evaluated the nutrient removal performance of pre-denitrification A/O configuration with recirculation of 500% (5 times of the influent flow) from aerobic to anoxic tank. The authors found that the novel layout is capable to remove 97% of the phosphorus with addition of external carbon source and without addition of coagulant.

Several efforts have been devoted to develop an innovative MBR with intermittent aeration and recirculation to provide anaerobic and anoxic conditions for proliferation of PAOs and to maximize the P-removal. The study of Ahn et al. (2003) proposed an innovative modification to MLE process by introducing an intermittently sequencing anoxic/anaerobic zone to alternate the anoxic conditions for denitrification and anaerobic conditions for phosphorus release. Anoxic conditions were imposed for 1 h after every 3 h, and anaerobic conditions lasted for two hours with no recirculation. During the anoxic stage, a recirculation ratio of 600% was maintained. This modified MLE improved the P-removal efficiency (93%) without coagulant addition and at much lower recirculation rates as compared to conventional MLE. Similarly, Zhang et al. (2006) investigated a cyclic aerobic, anoxic/anaerobic reactor for improved nutrient removal while reducing the number of recirculation streams, complexity and footprint of the system. The proposed configuration successfully removed 90% of the phosphorus without addition of coagulant and carbon source and even at low COD/TKN ratio with anaerobic-aerobic cycle time of 40 min and 120 min respectively. In another study, Ahmed et al. (2007) evaluated the impact of internal recirculation (IR) in sequencing anoxic/anaerobic membrane bioreactor (SAM). The authors found highest BioP-removal performance (55%) when the IR rate set to 2.5 times of the influent flow. The study of Yuan et al. (2008) introduced an innovative MBR with alternating anoxic and anaerobic environments for improved denitrification and P-removal. The proposed reactor consisted of a continuously aerated MBR and an alternating anaerobic and anoxic zone in two separate bioreactors. Controlling the recirculation flow from aerobic zone by control valves to either of these two bioreactors, anoxic and anaerobic conditions were implemented in two single tanks alternately. The authors found higher P-removal (> 94%)

without addition of carbon source and coagulant. However, the P-removal efficiency was found highly correlated to recirculation cycle time which may vary depending upon the influent load.

MLE and 4-stage Bardenpho configurations offer limited to moderate BioP-removal due to absence of the anaerobic zones essential for growth and proliferation of the PAOs responsible for phosphate consumption and therefore coagulant addition is required to eliminate the phosphorus below the regulatory limits. While, A/O or Pho-redox configuration provided moderate to high P-removal performances depending upon, operational conditions, influent characteristic, introduction of sequencing anaerobic/anoxic conditions and controlled intermittent recirculation. A²/O, 5-stage Bardenpho (with anaerobic as influent receiving zone), step-feed Bardenpho, UCT, MUCT and VIP configurations offer high to excellent BioP-removal performances. Table 1 depicts that the majority of the full-scale MBR facilities are equipped with MLE configuration and similar results have been documented by Pellegrin et al. (2015). This is due to fact that the CASPs with MLE configuration were upgraded to MBRs (Itokawa et al., 2014).

Bardenpho is the second widely used configuration at full-scale MBR facilities. Whereas, UCT and MUCT configurations are widely used for research studies at bench and pilot scales mostly with synthetic wastewater. This presents an opportunity to conduct more studies in this domain to fully assess the capabilities of innovative MBR configurations for simultaneous carbon and nutrient removal as required by the MBRs in majority of the cases in full-scale applications. Apart from the design configurations, P-removal in MBR is also affected by several other factors discussed in detail in Section 2.3.

2.3. Factors affecting the BioP-removal in MBR

In order to improve the P-removal mechanism as well as its prediction by mathematical model, a comprehensive understanding of the parameters affecting the BioP-removal is essential. There are limited numbers of MBR specific publications documenting the effect of SRT, HRT, feed composition and availability of VFA, DO concentrations, various recirculations, bacterial community (including PAOs/GAOs), alkalinity, pH, temperature, and other associated process and design parameters for BioP-removal. It is established that the BioP-removal in CASPs and the modified CASPs (such as MBR) are critically sensitive to various parameters such as SRT (Mulkerriens et al., 2004; Tchobanoglous et al., 2002; Wang et al., 2015). Since MBR is an extended version of the CASP replacing the secondary clarifiers by the membranes to retain 100% suspended solids, thus providing superior P-removal than CASP (Cho et al., 2005). Factors affecting the P-removal process in MBRs have been analyzed considering the various configurations of the WWTPs ranging from lab to large-scale applications from the reviewed publications.

2.3.1. Feed characteristics

COD is a limiting factor for the BioP-removal in the MBR process. It serves as a substrate for the microbial community (specifically PAOs) and can impede the removal efficiency if not available in the required concentration (Kapagiannidis et al., 2012; Park et al., 2019). For MBRs, a minimum COD/P ratio of 40–45 is required for effective P-removal (du Toit et al., 2007). Low PO₄³⁻ concentration in the influent resulted in higher P-removal efficiency in MBR. It was also found that increasing

Table 1
P-removal performance of various MBR configurations.

Configuration	Plants assessed	Wastewater type	Capacity (m ³ .d ⁻¹)	Influent TP (g·m ⁻³)	BOD/TKN ratio	SRT (days)	HRT (hours)	Coagulant addition	P-removal (%)	Reference	
2-stage MLE	1	Municipal	0.50	4.2	17.75***	40–45	16	YES	83	Holba et al., 2012	
	7	Municipal	125–2140	2.7–4.1	1.1–3.5	n.a.	10–40	NO	50–68	Itokawa et al., 2014	
	2	Municipal	1000–4200	2.90	0.1–1.1	n.a.	10–40	YES	65–92	Itokawa et al., 2014	
	5	Municipal	1100–16,000	2.8–4.9	1.7–2.6*	37–45	8–9.2	YES	89–97	Chae et al., 2015	
	1	Municipal	7000	7.0	4.30*	n.a.	n.a.	NO	60	Gabarrón et al., 2015	
2-stage MLE (SAM)	1	Synthetic	0.01	3.66	6.52*	70	n.a.	NO	93	Ahn et al., 2003	
3-stage MLE (cascade)	1	Municipal	6520	3.9	7.90*	14–21	3.5–5	NO	78	Fenu et al., 2010	
	1	Municipal	60	7.8	10.86***	30–75	42	YES	45.9	Holba et al., 2012	
	1	Municipal	30	10.6–18.3	8.9***	30–55	34	YES	27	Holba et al., 2012	
A2/O	1	Synthetic	0.264 (1)	11.4	6.92	25	14	NO	59.1	Ersu et al., 2008	
	1	Synthetic	0.264 (2)	11.4	6.92	25	14	NO	88.1	Ersu et al., 2008	
	1	Synthetic	0.264 (5)	11.4	6.92	25	14	NO	58.7	Ersu et al., 2008	
	1	Synthetic	0.083	5.5	5.25*	n.a.	10	YES	90–95	Banu et al., 2009	
	1	Municipal	~45,000	3.3	7.87**	25	17	YES	93.9	Wang et al., 2018	
4-stage Bardenpho	2	Municipal	500–2400	8.0	7.20**	18	n.a.	NO	74	Yilmaz et al., 2020	
	1	Municipal	75	5.5	3.8	27	27	YES	99.45	Fleischer et al., 2005	
5-stage Bardenpho	1	Municipal	42,000	5.4	6.5*	14	12.6	YES	99.07	Daigger et al., 2010	
Step-feed Bardenpho	1	Municipal	33,000	8	5.95	10	8.9	YES	99.13	Daigger et al., 2010	
A/O (Sequencing)	1	Synthetic	0.024–0.072	4	3.4–28	n.a.	7.3–22	NO	90	Zhang et al., 2006	
A/O (sequencing)	1	Synthetic	0.096	6	10***	20	9.6	NO	94.1	Yuan et al., 2008	
A/O (SAM)	1	Synthetic	0.01	5.7	7.09	50	8	NO	55	Ahmed et al., 2007	
A/O/Phoredox	1	Municipal	30	20	5.1–10.5	n.a.	72	NO	70–90	Abegglén et al., 2008	
A/O (modified)	1	Synthetic	0.264 (3)	11.4	6.92	25	14	NO	44.6	Ersu et al., 2008	
A/O (modified)	1	Synthetic	0.264 (4)	11.4	6.92	25	14	NO	42.7	Ersu et al., 2008	
A/O (Modified)	1	Synthetic	0.103	11–12.4	11–13.3	35–40	14.4–24	NO	97	Corsino et al., 2020	
UCT	2	Municipal	0.01	8.4–10.5	10–14	15	18–21	YES	99	Adam et al., 2003	
	2	Municipal	0.01–0.122	3–7.5	4.5–5.5	15–26	11–18	YES	90	Lesjean et al., 2005	
	1	Synthetic	lab-scale	8.8	7.97*	35	5	NO	39.8	Lee et al., 2009	
	1	Municipal	3–4.2	3.63 [◇]	5.7	15–22.6	n.a.	NO	80	Monclús et al., 2010	
	1	Municipal	0.616	4	1.93	n.a.	15.4	NO	60	Cosenza et al., 2013	
	1	Municipal	19–26	5.39	4.67	20	7	NO	94.1	Smith et al., 2014	
	1	Synthetic	0.012	4.7	7.07*	25	18	NO	82	Sun et al., 2019	
	1	Synthetic	0.012	4.63	6.5*	25	18	YES	95	Sun et al., 2019	
	MUCT [step feed]	1	Synthetic	lab-scale	8.2	7.46*	37	5	NO	72.5	Lee et al., 2009
	MUCT (sequencing)	1	Synthetic	0.011	8.2–11.3	4.3–5.2*	15	11–13	NO	75.2	Zhang et al., 2009
Modified VIP	1	Municipal	32,000	6.2	5.11	7.8	n.a.	YES	93.87	Daigger et al., 2010	

* COD/TN ratio.

** BOD/TN ratio.

*** COD/NH₄-N, [◇]PO₄³⁻.

κ the influent PO_4^- from 20 to 80 mgL^{-1} increased the sludge production from 2 to 6.2% (Choi et al., 2011). A recent A^2/O MBR study has investi-gated that high P-removal (82.6%) can be achieved with a low C/P ratio

(29) and relatively high nitrate (5.6 $\text{NO}_3\text{-N}$ mgL^{-1}) concentration (Falahati-Marvast and Karimi-Jashni, 2020) by the combination of assimilation and EBPR processes. Sometimes despite having a favorable COD/P ratio, P-removal is still compromised due to overloading and consequent decay of PAOs due to limited aeration (Abegglen et al., 2008). In the study of Oehmen et al. (2007) it was found that a COD/P ratio lower than 50 mgCOD/mgP favors the growth of GAOs, and the same was experienced by Monclús et al. (2010) in their UCT type MBR pilot plant.

Theoretically, 7–12 mg of VFAs are required to remove 1 mg of PO_4^- through EBPR in CASP, and this VFA/PO_4^- can be high as 20 depending upon the soluble fraction of COD in municipal wastewater (Al-Atar, 2007; Janssen et al., 2002; Monti et al., 2007; Mulkerrens et al., 2004). Typically municipal wastewater is VFA deficient, while MBR operated at high MLSS/higher SRT would require an external carbon source (ethanol) in the anaerobic zone or side stream P-recovery unit for effective P-removal (Monti et al., 2007).

Typically, a BOD/P ratio of 20 is considered a minimum requirement for better P-removal efficiencies in MBR studies dealing with municipal wastewater (Adam et al., 2003). However, this could go as higher as 45 and are still favorable for biological P-removal (Fleischer et al., 2005). It has been validated under a study (Wang, 2020) conducted to evaluate the nutrient removal performance of 11 full-scale MBR facilities located in China wherein the BOD/P ratio was found in the range of 12.22–36.94, and eight MBR facilities performed good P-removal due to the BOD/P ratio > 20. However, an external carbon source was still essential for total nitrogen removal.

In addition to COD/P or BOD/P, it was investigated that a low COD/N ratio between 5.3 and 7.3 resulted in satisfactory P-removal (Wang et al., 2015), while in a more recent study, it is found that low C/N ratio decreased P-removal (Mannina et al., 2020). A lower BOD/TKN ratio may result in poor denitrification and remaining nitrates, thus negatively impacting the overall nutrient removal capabilities of MBR. External addition of carbon source in these systems become essential (Fleischer et al., 2005). Several studies with PAOs have also confirmed that elevated concentrations of nitrite negatively affect phosphorus uptake activities of PAOs under both aerobic and anoxic conditions (Saito et al., 2004). When there is limited nitrate availability, ordinary heterotrophic organisms (OHOs) outcompete PAOs for nitrate, and there is a low impact on the EBPR process.

From all these considerations, it may be concluded that the optimum range of COD/P is around 40 and C/N in between 5.3 and 7.3. MBR is adapted to treat wastewater with high P and N contents with COD/TN/TP ratio in the range of 100/28.5/1.16–100/11/0.87 compared to conventional limit of 100/5/1 (Ersu et al., 2008). This is probably explained by the fact that the total bacteria and nitrogen-cycling groups in the MBR sludge are different than CASP (Wan et al., 2011).

2.3.2. MLSS concentration and P-removal in MBR

MBR operates at quite high MLSS concentrations (4000–15,000 $\text{g}\cdot\text{m}^{-3}$), which lowers the oxygen transfer rate due to increased viscosity and increase aeration expense (Judd, 2010). Thus, an adjustment in the oxygen transfer coefficient is needed for MBR modeling studies (Germain et al., 2007; Insel et al., 2011). Furthermore, sludge mass is disproportionate through the reactor length as the MLSS concentration in the downstream zone (membrane tank) is higher as compared to up-stream zones of the MBR, and this is regulated through excessive inter-reactor recirculation to achieve uniform MLSS concentrations for better biological nitrogen and P-removal (Ramphao et al., 2005). Several studies found that the higher MLSS concentration had little to no effect on P-removal kinetics. Thus, parameters used for ASMs could be applied to MBR systems (Adam et al., 2003; Holakoo et al., 2005; Parco et al., 2007). It was further concluded that the complete solids

retention by the membrane might change the microbial population and thus impact the growth kinetics (Parco et al., 2007). Another study found that P-removal was positively affected by increasing the MLSS concentration to a certain level (7 g/L optimum), and beyond this, P-removal was negatively impacted possibly because of the higher fraction of the dead biomass due to higher SRT (Wang et al., 2015).

Besides, it has been further investigated that the SMPs inhibit the anaerobic uptake of PAOs in CASP (Ichihashi et al., 2006). It was also later observed and confirmed by Jiang et al. (2009) for MBR with two batch tests. The results were similar to the previously experienced in case of CASP. Based on these results, the authors recommended to design and to operate MBRs at lower SRTs to avoid the production and accumulation of the SMPs onto the membrane surface. This inference is based upon only two tests and it is therefore advised to conduct more studies to explore the relationship between the SMPs and P-removal performance of the membrane, while keeping in mind the role of SMP in membrane fouling, and the role they could play in P retention (Gao et al., 2004).

2.3.3. Microbial communities and P-removal in MBRs

Autotrophic nitrifying bacteria and heterotrophic bacteria (GAOs respiring nitrates) compete for their growth under the same operating conditions while it is commonly accepted that the level of BioP-removal is directly proportional to the number of PAOs present in the system. EBPR is a tailored process for BioP-removal based upon PAOs property of intracellular accumulation of phosphate in poly-phosphate (Poly-P) form under cyclic anaerobic-aerobic (mainly anaerobic). Accumulated phosphorus is then carried away with the extracted sludge (Lesjean et al., 2003) as presented in Fig. 3. Since PAOs can store excess phosphorus in the form of polyphosphate (poly-P) granules in their bacterial cells after their metabolic demand would be satisfied. Whereas, P-removal by the assimilation phenomenon (which takes place only during growth process) by non-PAO biomass is limited to 15–30% (Lesjean et al., 2009; Monclús et al., 2010; Ramasahayam et al., 2014). The COD serves as a substrate for the growth of PAOs, providing them volatile fatty acids (VFAs) for consumption and synthesis of the poly-hydroxybutyrate (PHB while PHA when considering the role of only specific PAO class known as “Candidatus Accumulibacter phosphatis”) by releasing phosphorus and degrading the glycogen under anaerobic conditions. In the second step, phosphorus is taken up by the PAOs at higher rates in the presence of the excess oxygen provided for the oxidation of the PHBs/PHA with an injection of the influent at the anaerobic zone (Mulkerrens et al., 2004; Zhang et al., 2018). In order to meet the required phosphorus discharge target through relying only upon the proliferation of PAOs is not practiced (PAO communities remain very low) and even hardly possible for the large-scale MBR facilities (de-Bashan and Bashan, 2004).

In addition to phosphate uptake under aerobic environment by the PAO, it is also consumed by the DPAO (Kishida et al., 2006) in the anoxic environment where they consume nitrates and enhances the overall P-removal efficiencies. In addition to the aerobic zone in the MBR process, the membrane is intensively aerated to avoid fouling issues. This aeration also improved the P-removal efficiency as no P-release occur during the effluent production, unlike CASP. Furthermore, complete retention of solids in MBR helps to reduce the TP concentration in the effluent (Monti et al., 2006).

Other carbon growing micro-organism like GAOs also consume VFAs without contributing to P removal. GAOs found in pilot and full scale EBPR system are reported to cause nutrient limited conditions due to higher consumption of substrate available for proper proliferation of PAO organisms (Oehmen et al., 2007; Silva et al., 2012). MBRs are generally operated at higher SRTs (> 20 days) and low F/M ratio, which creates starvation conditions for micro-organisms resulting in inactivation or even death of PAOs and thus reduces sludge activity (Han et al., 2015; Yilmaz et al., 2007). Minimizing the GAOs concentration in the EBPR

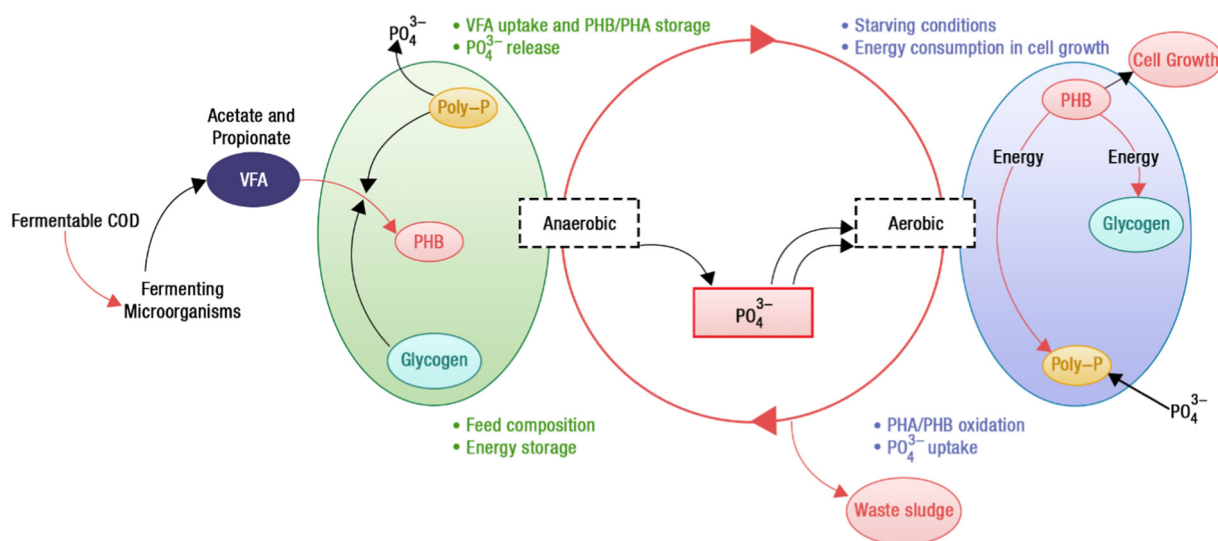


Fig. 3. PO_4^{3-} removal by the EBPR process under anaerobic/aerobic environment.

process in MBR may result in achieving higher P-removal efficiencies in the laboratory and pilot-scale systems.

Most of the studies at the laboratory and pilot scale systems have demonstrated that minimizing the GAOs concentration in the EBPR process in MBR resulted in achieving higher P-removal efficiencies. On the contrary, another study conducted at full-scale investigated the role of PAOs and GAOs for P-removal and concluded that GAOs do not appear to be a problem. Explanation of this could be due to the presence of competing DPAOs. It is, therefore, essential to rethink the PAO-GAO interaction for full-scale plants (Stokholm-Bjerregaard et al., 2017).

Furthermore, there is a conflict between the factors that govern nitrifying bacteria and PAOs. Nitrifiers (GAOs) have a slow growth rate and need a longer SRT (> 5 days) to grow, while the PAOs favor a shorter SRT ranging between 3 and 5 days (Onnis-Hayden et al., 2011). A longer SRT in MBRs means low net PAO biomass growth and thus limited phosphorus storage in new cell material and the system. PAOs need to be exposed to alternating anaerobic and aerobic conditions and thus are favored through the recycle streams, while the nitrifiers are maintained in the aerobic tank. Findings of the available studies have concluded that the type and amount of carbon source, pH, and temperature significantly affect the balance between PAOs and GAOs (Song et al., 2008).

Higher PAOs and DPAOs concentration help to achieve higher P-removal efficiency in MBR (Cho et al., 2005). It has been found that PAOs concentration in municipal wastewater varies from 0 to 1% of total COD (Sun et al., 2013), while in the sludge, the concentration of the PAO can be up to 7–10% of the total biomass (Mao et al., 2015; Silva et al., 2012). The PAOs can store up to 0.17 gP/gTSS, which is comparatively much higher than what can be typically stored in the sludge mass (0.015 gP/gTSS) (Henze et al., 2008). In a UCT-type, MBR operated with weak and strong wastewater to analyze the phosphorus uptake in an anoxic zone. DPAOs played a vital role in phosphorous removal, along with nitrogen. The removal rate in the anoxic zone was higher for high strength wastewater than weak wastewater and thus indicated that up to 40% phosphorus could be removed through P-uptake (Lee et al., 2009).

The roles of GAOs and PAOs have been studied for CASP full-scale plants, but studies for full-scale MBR with higher SRT are still missing. SRT and carbon sources seem to be determinant parameters in previous studies.

In the early interpretation of the EBPR, the *Acinetobacter* was considered as the only class of PAOs responsible for accumulating phosphorus in WWTPs (Barnard et al., 2017). By the late 1990s, understanding of the EBPR expanded, and it was found that the microbial class “Candidatus

Accumulibacter phosphatis” are primary organisms responsible for PAO characteristics (Liu et al., 2019). Several mathematical models; including, ASM2/ASM2d, TUDP, Barker and Dold’s, and UCTPHO⁺ are based upon the functional working of “*Candidatus Accumulibacter phosphatis*” for P-removal (Hauduc et al., 2013; Ni et al., 2010) and competition between the PAOs and GAOs is not considered. The study of Mielczarek et al. (2013) investigated 28 Danish EBPR based WWTPs and reported that 27% of the PAOs are “*Tetrasphaera*”. Similarly, Stokholm-Bjerregaard et al. (2017) studied 19 full-scale EBPR based WWTPs and found similar results to Mielczarek et al. (2013) with “*Tetrasphaera*” as the most abundant PAO accounting for 10.7% of the total active biomass. The most recent research conducted on 32 full-scale wastewater facilities located in 12 different countries found that “*Tetrasphaera*” is the most abundant (Nielsen et al., 2019) and confirmed the findings of the previous studies conducted on a full-scale. Apart from the *Accumulibacter*, *Acinetobacter*, and *Tetrasphaera*; other genera of PAOs are *Aeromonas*, *Enterobacter*, *Moraxella*, *Klebsiella*, and *Pseudomonas* (Mielczarek et al., 2013; Stokholm-Bjerregaard et al., 2017).

At lab-scale, mainly “*Candidatus Accumulibacter phosphatis*” were found, whereas at full-scale, *Tetrasphaera* were the most abundant microbial community. This could affect the mechanisms of P-removal at these different scales and their modeling and therefore require further research and improvements in the ASM models as far as the role of PAOs is concerned.

2.3.4. Sludge retention time and P-removal in MBRs

SRT may influence the sludge characteristics, such as viscosity, biomass concentration, microbial community’s composition, floc size, viscosity, and cell surface properties (Hocaoglu et al., 2011). The effect of SRT on P-removal in MBRs is still conflicting in the literature. It was initially believed that BioP-removal is difficult to achieve at higher SRTs as experienced in MBR; however Adam et al. (2003) first demonstrated that P-removal could be achieved in MBR operating at even higher SRTs with influent having a higher VFA/P ratio.

Some other studies have reported even superior (> 90%) BioP-removal at longer SRTs (25–75 days) in MBR systems with and without sequencing anoxic/anaerobic functioning (Ahn et al., 2003; Cho et al., 2005; Ersu, 2006). As such, there may be an optimum SRT needed for nutrient removal in an MBR. The evaluation of full-scale MBR facilities showed unexpected high BioP-removal operating at higher SRTs and technically not designed for EBPR. No explanation was provided for this peculiar behavior (Silva et al., 2009). Higher SRTs helped in lowering the rate of phosphate release during PHA storage (Adam et al., 2003;

Nopens et al., 2007; Rosenberger et al., 2000; Sun et al., 2019) and thus negatively affecting the P-removal due to higher growth of AOB (higher NO_2 production) which lead to compromise the functioning of heterotrophic non-PAOs and PAOs as explained by Mannina et al. (2020). Whereas Lesjean et al. (2009) found that higher EBPR in MBR can be achieved even at higher SRT. Based on these conflicting studies, it can be assumed that others parameters are more impacting than SRT, and it is difficult to ascertain the role of SRT in P-removal in MBRs.

2.3.5. Hydraulic retention time and P-removal in MBRs

There are limited studies focused on documenting the effect of anaerobic and anoxic HRTs on P-removal in MBRs. The study presented by Cho et al. (2005) evaluated the performance of a sequencing anoxic/anaerobic MBR through varying HRTs and flux. The authors found higher P-removal at shorter anaerobic HRTs (1 h) due to higher substrate loading. The same authors further found that, the membrane fouling increased with increasing flux linked with shorter HRT. Another study (Monclús et al., 2010) supported the same argument wherein insufficient anaerobic HRT (0.921–2.05 h) reduced the conversion of readily biodegradable substrates to stored PHA and negatively impacted the P-removal due to reduced activity of PAOs for phosphate accumulation.

In another study (Song et al., 2008), the effect of change in HRT on P-removal in MBR was evaluated. The authors found that shorter HRT between anoxic and anaerobic induced a higher F/M ratio and increased biomass yield activity, especially in denitrification rate, which then improved P-removal efficiency. Similarly, Brown et al. (2011) reported that an optimal anaerobic HRT (2 h) is needed for PAOs to actively uptake the phosphate and PHA and use it as an energy source. While, Sun et al. (2013) found better P-removal efficiency with an overall higher HRT (11.6 h) and with anaerobic HRT (3.3 h). Most recently, a lab-scale MBR treating domestic wastewater has been investigated at higher HRT (9.6 h) and resulted in higher P-removal (Sözüdoğru et al., 2021). While, in another recent study (Falahati-Marvast and Karimi-Jashni, 2020), it has been found that P-removal in A^2/O -MBR slightly improved with reduction of aerobic HRT from 12 h to 6 h, presumably due to an increased biomass concentration at constant F/M ratio and under controlled temperature. The aerobic HRT does not seem to impact the P-removal, although it must be maintained within a 6–12 h. While increasing anaerobic HRT (within range of 2–6 h) improved the P-removal to a certain limit where denitrification (by DPAOs) would be favored.

2.3.6. Effect of temperature on P-removal in MBRs

There are conflicting results from various studies documenting the effects of temperature on P-removal. A temperature ranging between 10 and 25 °C resulted in better P-removal, and inconsistencies have been found at high temperatures around 40 °C (Liau et al., 2015; Sayi-Ucar et al., 2015). Another study confirmed that beyond 90% P-removal could be achieved with 10 °C operating temperature (Wei et al., 2012). According to another study, PAOs are lower-range mesophiles or psychrophiles and are predominated only at 20 °C or lower temperature ranges. While on the other hand, GAOs are somewhat mid-range mesophilic organisms with an optimum temperature between 25 °C and 32.5 °C (Panswad et al., 2003).

2.3.7. Dissolved oxygen concentration and P-removal in MBR

To strike a balance between the required oxygen supply for nitrifiers in the aerated tank and creating an enabling environment for PAOs, a DO level of 2 mgL^{-1} is recommended (Hai et al., 2018). It has been found that too low (0.4–0.6 $\text{mgO}_2\text{L}^{-1}$) and too high (2.0–2.4 $\text{mgO}_2\text{L}^{-1}$) DO concentration has adverse effects on P-removal in MBR (Roberts, 2020). Similarly, Smith et al. (2014) found improved P-uptake in the aerobic reactor with DO concentration maintained at 2 mgL^{-1} . Another study (Fu et al., 2009) with A/O -MBR demonstrated that a concentration of 2.5 $\text{mgO}_2\text{L}^{-1}$ is enough to remove almost 90%

of the phosphorous. At concentration lower than 2.5 $\text{mgO}_2\text{L}^{-1}$, PHB stored in PAOs could not be decomposed effectively because of DO deficiency in the aerobic tank. Thus, not enough adenosine triphosphate (ATP) was produced, which led to the restraint of excessive phosphorus uptake of the continuous PHB accumulation in PAOs. On the other hand, when the DO level was higher (2–2.4 mgL^{-1}), a higher concentration of nitrate was sent back to an anaerobic tank creating a competitive environment for denitrifiers (GAOs and DPAOs) and PAOs, thus limiting the P-removal efficiency (Yuan et al., 2012). In contrast, Nopens et al. (2007) found that the optimal P-removal can be achieved at a concentration of 0.5 mgL^{-1} which also correspond to lower nitrates which are known inhibitory agents. It has also been investigated that too low DO concentration (0.3 mgL^{-1}) in the aerated MBR zone triggered the uncontrolled growth of filamentous micro-organism (Insel et al., 2014). Another study evaluated no considerable effect of DO concentration (1–4.1 $\text{mgO}_2\text{L}^{-1}$) on biological P-removal without ascertaining the role of PAOs and GAOs (Sayi-Ucar et al., 2015).

A DO concentration between 0.4 and 2.0 mgL^{-1} is found an optimum range for P-removal above 80%. At the same time, there is disagreement for higher and lower concentrations from this optimum value. However, the choice for maintaining the DO level depends upon several other factors such as feed characteristics, target removal performance for COD and nitrogen, and the cost of biological aeration. Further assessment of the role of DO concentration at full-scale MBR facilities may help understand its role in EBPR. A summary of several factors affecting the P-removal in MBR is given in Table 2. The summarized results reflect that further research is needed to explore and verify the conflicting studies, especially at the full-scale.

2.4. Mathematical modeling of MBRs

It is well understood that MBRs operate under different conditions than CASP and requires a comprehensive understanding of the processes and adjustments in models initially developed for CASP to successfully simulate the phosphorous removal process in MBRs. The need for adjustments in ASMs' applicability to MBR is mainly attributed to: i) different microbial composition leading to the calibration of stoichiometric and kinetic parameters ii) higher biomass concentration leading to reduced oxygen transfer and uptake, iii) production of EPSs (linked to flocs) and SMPs (dissolved) and their accumulation onto the membrane surface iv) additional aeration involved in membrane scouring and recirculation of oxygen saturated sludge from MBR to the aerated/unaerated zones and v) the role of membrane filtration on nutrient removal (Fleischer et al., 2005; Hai et al., 2018; Judd, 2010; Maere et al., 2011; Verrecht et al., 2008). The comparison of these parameters for CASP and MBR is proposed as tabulated format in Table 3.

In a detailed review article, Fenu et al. (2010) discussed the adaptation of the for ASM models to the MBR with and without modifications. The unmodified ASMs required adjustments of nitrification related parameters, e.g., dissolved oxygen half-saturation coefficient (K_{O_2}), which was assumed to be attributed to smaller floc size in MBR, which eases the O_2 transfer. For modified ASM, extension with EPS/SMPs is justified if the modeling objective is to study higher SRT, linking the biology with the filtration, and with the necessity of predicting soluble COD in bulk. Otherwise, it makes the calibration process cumbersome and challenging to calibrate EPS/SMP-related parameters. It was further proposed to undertake full-scale studies to rule out the difference in lab-scale to full-scale models' applications. P-removal was briefly touched, taking into account five relevant studies, and no specific conclusion was derived due to the dearth of studies and insufficient available data. Albeit, it was suggested to explore the biological P-removal kinetics through additional research. Another comprehensive review on MBR modeling was conducted by Naessens et al. (2012a, 2012b). However P-removal aspect is not discussed. Most recently, based on earlier review on integrated MBR modeling, IWA task group has proposed best practices to be adopted to model MBR taking into account key process

Table 2

Summary of the factors affecting the P-removal in MBRs.

Factor	Parameters	Impact on P-removal	Optimum range	Reference
Influent characteristics	PO ₄ loading	Higher BioP-removal at lower influent PO ₄ concentrations.	2–20 gTP·m ⁻³	Choi et al., 2011; Monclús et al., 2010
	COD/P ratio	Higher C/P ratio positively impacts the P-removal. While lower C/P ratio lead to unstable conditions required to achieve maximum P-removal. A lower C/P ratio may result in higher nutrients (P&N) removal in the absence of nitrates interference.	29–45	Al-Hashimia et al., 2013; du Toit et al., 2007; Kapagiannidis et al., 2012; Monclús et al., 2010; Wang, 2020 Falahati-Marvast and Karimi-Jashni, 2020
	BOD/P ratio	Higher BOD/P results in higher BioP-removal	12.33–45	Adam et al., 2003; Fleischer et al., 2005; Wang, 2020
	COD/N ratio	A low COD/N ratio negatively impacts Phosphorus removal.	5.3–7.3	Hu et al., 2014; Lee et al., 2015; Mannina et al., 2018
MLSS concentrations	Nitrite concentration	Higher concentrations of nitrite negatively affect PAOs under both aerobic and anoxic condition	< 1 g NO ₃ ·m ⁻³	Liu et al., 2011; Roberts, 2020; Saito et al., 2004; Sin et al., 2008
	MLSS concentration	Little or no effect on BioP-removal kinetics BioP-removal is positively impacted by increasing the MLSS concentration up to an optimum concentration of 7 g/m ³	<7 g TSS·m ⁻³	Adam et al., 2003; Holakoo et al., 2005; Parco et al., 2007 Wang et al., 2015
	EPS concentration	Higher EPS concentrations result in higher BioP-removal due to the fact that EPS act as a phosphorus reservoir (<i>approximately 5–10% of phosphorus in sludge is reserved in the EPS</i>)	20–130 gCOD·m ⁻³	Adoonsook et al., 2019; Cloete and Oosthuizen, 2001; Zhang et al., 2013
	SMP concentration	SMPs inhibit the luxury BioP-uptake by PAOs in the anaerobic reactor	24–86 gCOD·m ⁻³	Gao et al., 2004; Ichihashi et al., 2006; Jiang et al., 2009
Microbial community	PAOs & GAOs concentration	High PAO concentration resulted in higher BioP-removal in a MBR model based study.	7–10% of total biomass concentration	Jadhao and Dawande, 2012; Mao et al., 2015; Silva et al., 2012
	PAOs class and relative abundance	In full-scale WWTPs, PAOs class "Tetrasphaera" is found in abundance with relative fraction ranges between 10.7% and 27% of the total PAOs population.	10.7%–27% of the PAOs population	Mielczarek et al., 2013; Nielsen et al., 2019
Sludge retention time	SRT	Longer SRT and high MLSS/MLVSS concentration in MBR may induce a competitive advantage for GAOs over the PAOs, thus negatively impacting the BioP-removal performance. Longer SRTs positively impact the P-removal in MBRs. Especially when the system worked without sludge withdrawals	15–40 days	Ersu et al., 2010; Han et al., 2015; Hu et al., 2014; Lee et al., 2009; Nopens et al., 2007; Sun et al., 2019; Wang et al., 2015 Adam et al., 2002, 2003; Ahn et al., 2003; Ersu, 2006; Ersu et al., 2010; Mannina et al., 2020; Silva et al., 2012; H.-M. Zhang et al., 2006 Brown et al., 2011
	Hydraulic retention time	Anaerobic HRT Higher anaerobic HRT helps in higher BioP-removal Aerobic HRT Reduction in aerobic HRT (12 h to 6 h) slightly improved the BioP-removal performance due to increase biomass at the constant FM ratio. Total HRT Higher and stable BioP-removal efficiencies could be achieved at HRT lower than 10 h. Temperature Better BioP-removal at a temperature ranging between 10–25°C and inconsistencies at high temperatures around 40°C	2–2.9 h 3–6 h <10 h 10–25 °C	Cho et al., 2005; Monclús et al., 2010 Ahmed, 2012; Falahati-Marvast and Karimi-Jashni, 2020; Sun et al., 2019 Falahati-Marvast and Karimi-Jashni, 2020; Sözüdoğru et al., 2021; Sun et al., 2013 Liau et al., 2015; Sayi-Ucar et al., 2015
Dissolved oxygen concentrations	DO concentration	BioP-removal was found independent of the DO concentrations. Higher DO concentration of 2.5 mgL ⁻¹ resulted in higher BioP-removal. Optimum DO (0.4–2.5 mgL ⁻¹) is required for higher BioP-removal efficiency, and concentration above 2–2.4 resulted in decreasing the P-removal performance. Too low DO concentration (0.3 mgL ⁻¹) can help filamentous bacteria grow and counteract BioP-removal	0.4–2.5 gO ₂ ·m ⁻³	Sayi-Ucar et al., 2015 Fu et al., 2009 Downing et al., 2014; Hai et al., 2018; Nopens et al., 2007; Roberts, 2020; Smith et al., 2014; Yuan et al., 2012 Insel et al., 2014

indicators such as effluent quality index (EQI), membrane fouling, aeration, operating costs index (OCI), energy consumption, and mitigation of GHG emissions (Mannina et al., 2021).

MBR involves complex interactions which are taking into account through several combined modeling ways among i) biological processes, ii) membrane filtration – fouling phenomenon, and iii) hydrodynamics (Naessens et al., 2012a, 2012b; Ng and Kim, 2007). ASMs did not address the oxygen transfer/diffusion phenomenon, and aeration models were later developed and used as sub-models for MBR modeling studies (Delrue et al., 2010; Hocaoglu et al., 2011; Insel et al., 2011; Verrecht et al., 2008; Zarragoitia-González et al., 2008). Few integrated models have also been developed, taking into account the biological processes, filtration, fouling, and flow dynamics (Di Bella et al., 2008; Janus, 2014; Suh et al., 2013; Zarragoitia-González et al., 2008), but these still require validation at full-scale MBR plants. Their advantage for our purpose is to be available as a set of models where P-removal mechanisms may be integrated, with their impacts on other main variables. P-removal can be accomplished by biological reaction and chemical precipitation. Therefore different models can be classified with respect to

biological (metabolic, ASMs, and the combination thereof) and chemical P-removal mechanisms.

2.5. Modeling and simulation of BioP-removal in MBRs

Mathematically, the BioP-removal process can be described either by i) metabolic model, ii) activated sludge models-ASM or iii) coupling of metabolic and ASM models, e.g., TUDP (Baetens, 2001; Lanham et al., 2014). Both of these modeling approaches use a set of stoichiometric and kinetic equations to describe the transformation steps of the EBPR process (Oehmen et al., 2007). Metabolic models use biochemistry knowledge of the active metabolic pathways to explain the cells' biochemical transformations (Oehmen et al., 2007). Metabolic models have been used to investigate the much-debated GAO-PAO competition in EBPR and reveals that the role of GAOs is negatively impacting the P-removal. However, the yield coefficients for PAOs and GAOs in metabolic models are determined theoretically based upon the reaction stoichiometry for the assumed pathways. Indeed GAOs and PAOs are challenging to be obtained in pure culture. This lack of experimental data is therefore, the major limitation of these models. As compared to

Table 3
Considerations in applying ASMs to the MBRs for the EBPR process.

Parameter/factor	Conventional activated sludge	Membrane bioreactor
Microbial composition	CASPs are operated at lower SRTs ranging between 4 and 15 days (Hai et al., 2018; Judd, 2010)	SRT of MBRs ranges between the same to three-time of the CASP (Hai et al., 2018). MBR promotes slow-growing micro-organisms such as nitrifiers and micro-organisms that are usually washed out in a CASP system while the membrane retains 100%. Specific stoichiometric and kinetic parameters adjustments are required while applying ASM in order to model MBRs (Fenu et al., 2010; Naessens et al., 2012a). MBRs are operated at higher MLSS concentrations ranging between 4000 and 15,000 g·m ⁻³ (Hai et al., 2018) and, therefore, lower FM ratio. Higher MLSS in combination with higher SRT cause stress to the micro-organisms in an MBR, which requires more energy for cell maintenance and therefore leaves less energy for cell production. Higher MLSS may induce the competitive advantage of GAOs over PAOs. Accumulation of the EPS/SMPs onto the membrane surface may affect the fouling and the biological process. As discussed earlier, EPS also affects the P-removal. EPS/SMP concentration in MBR sludge may be different from the CASP sludge due to different microbial community and F/M ratio.
MLSS	CASP is operated at MLSS concentration ranging between 1500 and 3000 g·m ⁻³ (Hai et al., 2018)	MBRs are operated at higher MLSS concentrations ranging between 4000 and 15,000 g·m ⁻³ (Hai et al., 2018) and, therefore, lower FM ratio. Higher MLSS in combination with higher SRT cause stress to the micro-organisms in an MBR, which requires more energy for cell maintenance and therefore leaves less energy for cell production. Higher MLSS may induce the competitive advantage of GAOs over PAOs. Accumulation of the EPS/SMPs onto the membrane surface may affect the fouling and the biological process. As discussed earlier, EPS also affects the P-removal. EPS/SMP concentration in MBR sludge may be different from the CASP sludge due to different microbial community and F/M ratio.
EPS/SMP	EPS/SMP produces in CASP are washed away and/or removed through sludge. EPS/SMPs are not considered in unmodified ASMs	Accumulation of the EPS/SMPs onto the membrane surface may affect the fouling and the biological process. As discussed earlier, EPS also affects the P-removal. EPS/SMP concentration in MBR sludge may be different from the CASP sludge due to different microbial community and F/M ratio.
Fine bubble aeration	Aeration is used for carbon matter degradation and the nitrification conversion.	Aeration is used for carbon matter degradation and nitrification. However, due to higher MLSS concentration (unfavorable) and smaller floc size, (favorable) mass transfer limitations have been reported different in MBRs than in CAPs (Fenu et al., 2010).
Coarse bubble aeration	Coarse bubble aeration is not involved	In addition to fine bubble aeration, coarse bubble aeration aims at scrubbing the membrane surface to mitigate the membrane fouling. It creates turbulence and affects the floc size and mass transfer processes.
Separation	separation is carried out by the clarifiers (secondary) and the phenomenon is modeled through clarifier/settling models (Hai et al., 2018; Henze et al., 2008)	Separation is carried out by physical media (membrane). With this, MBR requires a sub-model to simulate filtration-fouling phenomenon and retention of the soluble/dissolved fraction of COD and nutrient (Di Bella et al., 2008; Mannina et al., 2018). Due to complete retention, the microbial composition in MBR may vary.

metabolic models, ASMs use a global “mechanistic approach” to characterize the energy, redox, and mass balances of cell processes within CASP/MBR to describe the EBPR process. These are focused on macroscopic phenomena and depend on biochemical transformation pathways of soluble and particulate compounds in sludge and metabolic activities (Baetens, 2001; Santos et al., 2020; Seviour et al., 2019). The role of GAOs is mainly neglected in the original ASMs. However, some modified ASMs consider the glycogen as a storage polymer in addition to PHA, taking into account the growth and activity of the GAOs (Baetens, 2001; Gernaey et al., 2004; Hauduc et al., 2013; Oehmen et al., 2007). The effect of temperature, pH, and carbon source on the competition between PAO and GAO populations has been discussed in modified ASM (Lopez-Vazquez et al., 2009). The yield coefficient is measured experimentally, making ASMs distinguished from metabolic models (Baetens, 2001).

During the last two decades, a couple of detailed reviews have been published focused on comparing the model structures, limitations, and differences of the processes such as hydrolysis, growth, and decay of organisms, including their limitations for P-removal. The first detailed review was conducted by (Baetens, 2001) considering all the published models, metabolic and ASM (original and modified) and combinations till 2000. Following this, (Gernaey et al., 2004) published a review on the white box models (ASM1, ASM2, ASM2/ASM2d, ASM3 TUDP, Barker & Dold’s model, and ASM3-BioP) and discussed that how the objective of the modeling exercise influences the model selection from the available range, data gathering, and model calibration. The authors further discussed the gray box (statistical), black-box (stochastic), and hybrid models and their potential applications in WWTPs, such as supervisory control system development. Following this, Hauduc et al. (2013) compared seven published models, including ASM1 (Henze et al., 1987), ASM2d (Henze et al., 1999), ASM3 (Gujer et al., 1999), ASM3-BioP (Rieger et al., 2001), ASM2d-TUD (Meijer, 2004), Barker & Dold’s model (Barker and Dold, 1997), and UCTPHO+ (Hu et al., 2007) using a structured approach and dealt with the difficulties in comparing models as highlighted by Baetens (2001). These models are compared based upon several standard processes among the range of models, including i) hydrolysis, ii) fermentation iii) growth and decay of OHOs iv) growth and decay of ANOs v) growth and decay of PAOs vi) storage of PHA and vii) storage of polyphosphate. Following the works of Gernaey et al. (2004) and of Hauduc et al. (2013), the mini-review of Zuthi et al. (2013) compared the five models (ASM2, ASM2d, TUDP,

UCTPHO+, and ASM3-BioP) that takes into account the P-removal for the CASP and MBR along with the advantages and disadvantages of these models. All these reviews had limited discussions related to the applications of these models to the full-scale WWTP and in particular to MBR and their corresponding adjustments and challenges faced during their adaptations to the MBRs.

P-removal from the wastewater can be mathematically described using several models derived from models of COD removal (Dold et al., 1981, van Haandel et al., 1981, Henze et al., 1987, Gujer et al., 1999), including i) ASM2 (Gujer et al., 1995) ii) ASM2d (Henze et al., 1999), iii) SIPHOR kinetic model (Johansson, 1996), iv) Dold’s mechanistic model (Barker and Dold, 1997), v) Wentzel’s model (Wentzel, 1989), vi) UCTPH model (Wentzel et al., 1992) vii) ASM2/ASM2d-TUDP model (Wentzel et al., 1988; Meijer, 2004), viii) New UCTPH (Hu et al., 2007), ix) EAWAG’s ASM3-BioP (Rieger et al., 2001), x) FCASM-1 (Sun and Song, 2009), and xi) modified ASM3-BioP-N2 (Ni et al., 2010). A new bio-kinetic model (modified Barker and Dold’s model) was developed considering the roles of GAOs and incorporating the new process concerning the GAOs (Varga et al., 2018) and validated at lab scale as well at full-scale CASP.

All these models can be studied under three groups i) models with considering the role of denitrifying PAOs, ii) models without considering the role of denitrifying PAOs, and iii) Models with the incorporation of the PAO and GAOs (Fig. 4).

2.5.1. Calibration of models

Metabolic models can be easily calibrated due to the limited number of parameters, and kinetic parameters are calibrated only when stoichiometric and kinetic reactions are changed to accommodate new process understanding (Lanham et al., 2014). Meanwhile, ASMs required intensive stoichiometric and kinetic parameters adjustments that compromise their predictive power and limit their practical applications for long-term EBPR process evaluations (Santos et al., 2020). Although it has been observed during the literature survey that most of the modeling studies involved calibration of the stoichiometric and kinetic parameters, albeit adjustment of the stoichiometric parameters is not considered as a good modeling practice (Rieger et al., 2012). The ASM’s calibration process is time-consuming and complex. Therefore, several protocols have been developed recently to cope with this complexity and perform a systematic calibration of the model parameters, including BioMATH, STOWA, HSG, WERF, and sensitivity analysis

based approach (Germaey and Sin, 2008; Mannina et al., 2011; Sin et al., 2005).

2.5.2. MBR simulation studies

This review is voluntarily limited to the ASM models (modified/un-modified) applied to the MBRs for P-removal. As per the Scopus data, there are 18 publications related to MBR modeling, and ASM2d is found to be widely used model (13 studies) while ASM3-BioP (1 study), TUDP (1 study), New General Model (3 studies) has rarely been used for MBR modeling. Table 4 summarizes selected papers related to MBR modeling. The details related to operational conditions and influent characteristics of the modeling studies are provided as supplementary material (Appendix C). The modeling and simulations case studies are discussed below.

A six-chambered pilot MBR plant was modeled using ASM2d (Fleischer et al., 2005). The authors concluded that the model was able to successfully predict the BNR performance of the MBR and was found capable to predict the phosphorus concentration below $0.1 \text{ mgP}_{\text{tot}} \text{ L}^{-1}$. Although, no information has been provided about the model structure, calibration, and validation procedures. Similarly, Daigger et al. (2010) successfully modeled the Traverse City full-scale MBR plant with a capacity of $32,000 \text{ m}^3\text{d}^{-1}$ and Broad Run pilot plant using ASM2d. The publication provides no details about the model structure calibration and validation process as of Fleischer et al. (2005). In the study of Monclús et al. (2010), a UCT-type pilot MBR is modeled using ASM2d with a special focus on biological P-removal. The model could simulate the P-removal performance without any change in the stoichiometric and kinetic parameters. Furthermore, the authors found that the decrease in the recirculation flow from the anoxic to anaerobic zone negatively impacted the P-removal.

A pilot plant receiving high strength wastewater from toilet flushing has been used for dynamic modeling using ASM2d (Verrecht et al., 2010). The model was first calibrated with steady-state, where it was required to have a correct representation of MLSS concentrations. The model parameters including $\mu_{\text{PAO_Max}}$ (2 d^{-1} vs default value of 1 d^{-1}), b_{PAO} (0.1 d^{-1} vs default value of 0.2 d^{-1}) and YPP_Stor_PAO ($0.2 \text{ gP}(\text{gCOD})^{-1}$ vs default value of $0.4 \text{ gP}(\text{gCOD})^{-1}$) were calibrated through heuristic approach without batch tests. However, the modeling of the removal of other nutrients could not be completely validated. While, with the dynamic simulation, PAO concentration increased without the adjustments to $\mu_{\text{PAO_Max}}$, b_{PAO} , and YPO and MLSS concentration were representative as the dynamic equilibrium was reached. The values of the parameters including m_{PAO} , b_{PAO} , and YPP_Stor_PAO , were then reverted to defaults.

The SMPs and ESP now has an established role in membrane fouling (Meng et al., 2009) and it has been proved that higher concentration of EPS deteriorates the P-removal performance (Ichihashi et al., 2006; Jiang et al., 2009). Keeping this in view, Jiang et al. (2008) extended ASM2d with SMP module (ASM2d-SMP) considering the soluble utilization associated products (SUAP) and soluble biomass related products to investigate the role of SMPs onto the membrane fouling. The introduction of SMP modules increased the model complexity and the additional parameters required calibration. Nevertheless, the ASM2d-SMP model successfully simulated the BNR behavior of a lab-scale MBR under steady-state conditions and was validated by the experimental data. The authors concluded that if the objective is to remove nutrients, an optimum SRT (17 days) agrees with published studies as discussed in Section 2.3.4. Following their previous research, Jiang et al. (2009) investigated ASMs' (unmodified) applicability and required adjustments to meet the MBR system biomass kinetics. Again, a lab-scale, MBR

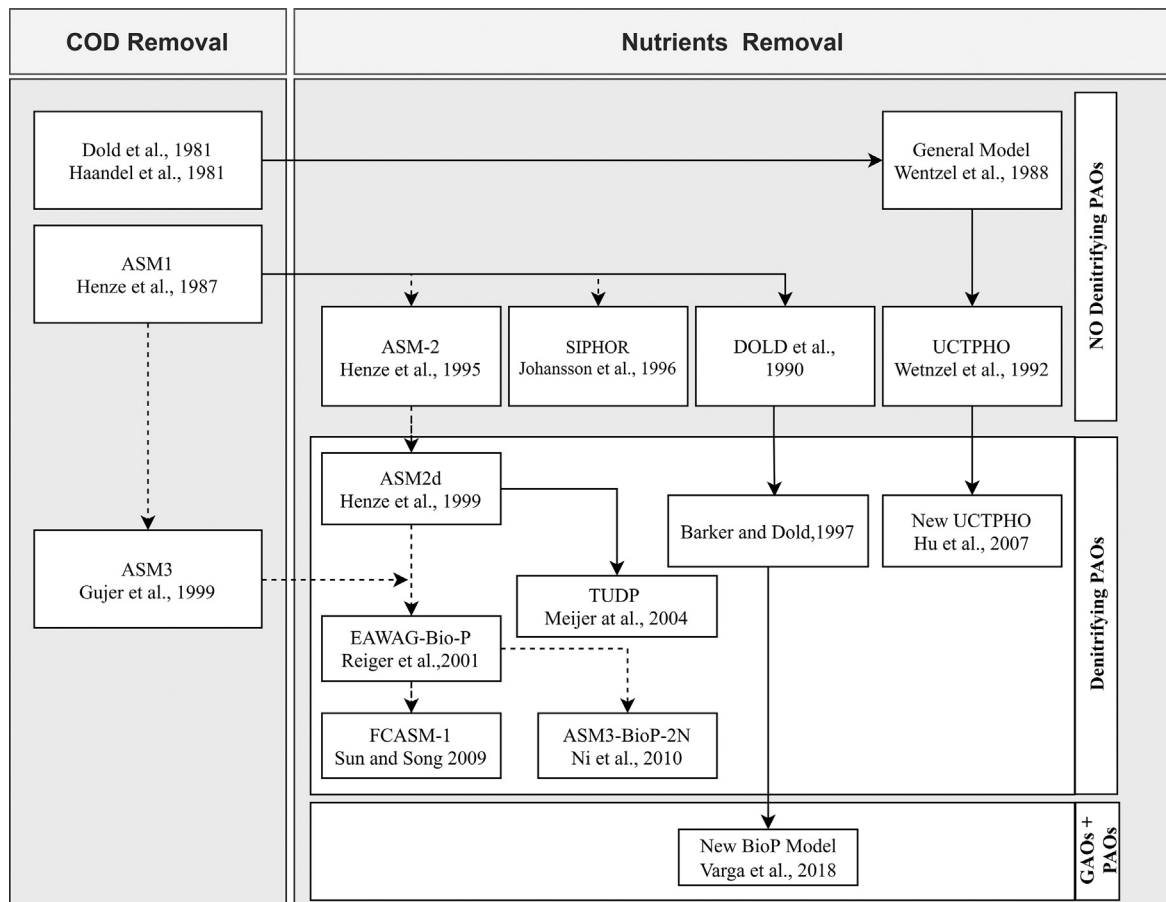


Fig. 4. Evolution and classification of the ASMs concerning GAOs and

Table 4
Summary of the key information and process conditions used in reviewed MBR modeling studies.

Reference	Model features														
	Model used	Wastewater	MBR scale	Configuration	precipitation	calibration protocol	Simulation environment	COD/TP influent ratios	COD/TN	Operating conditions	TP				
										HRT (h)	SRT (d)	MLSS (g·L ⁻¹)	DO ((g·m ⁻³))	Temp. (°C)	Predicted effluent traits (mgL ⁻¹)
Fleischer et al., 2005	ASM2d	R	P	Bardenpho	Al	-	-	24.2 ^a	3.8 ^b	8.4-8.8	13-19	4-9	8	-	0.1
Al-Astar, 2007	TUDP	R	P	UCT		STOWA	AQUASIM	88.8	10.6	7-10	12-20	7.4	3	16-24	0.15
Nopens et al., 2007	ASM2d	S	P	MUCT		heuristic		-	9.91	6.4	17.55	10	2	15	6.58*
Ersu et al., 2008	NGM	S	L	AO/AZO		heuristic	BioWin	45.3	12	14	25	8	2	22.3	1.37
Jiang et al., 2008	ASM2d-SMP	S	L	AO _{sequencing}		STOWA	-	36.2	7.29	17	7.2	10.9	0-2.5	15	2.2
Abegglen et al., 2008	ASM3-BioP	R	P	AO		heuristic	SIMBA	75	5.6	24-72	30-50	-	-	15	1.2
Jiang et al., 2009	ASM2d-SMP	S	P	AO _{sequencing}		STOWA	-	36.2	7.29	17	7.2	8.86	0-2.5	15	2.2
Monclús et al., 2010	ASM2d	R	P	UCT		heuristic	WEST	114	9.09	14-18	23	9.8	1.5	16.4	0.32
Verrecht et al., 2010	ASM2d	R	P	MLE	Al	heuristic	WEST	51.67 ^c	5.88	26	47	8	2	15-20	4.35
Daigger et al., 2010	ASM2d	R	F	VIP		-	-	58.22	5.95	8.4	19-23	1.5-9	-	16.1	0.33
Ersu et al., 2010	NGM	S	L	UCT		heuristic	BioWin	40.8	12.81	8	10-75	7.5	2	22-23	3.4
Liu et al., 2011	NGM	R	P	UCT	Al	WERF	BioWin	61.7	8.83	-	51	9-10	2	20	0.025
Gholikandi and Khosravi, 2012	ASM2d	S	L	AO _{sequencing}		STOWA	WEST	-	-	7	15-40	10-12.4	-	10-23	5.8*
Cosenza et al., 2013	ASM2d-SMP	R	P	UCT		GLUE	-	81.75	3.59	-	36	3.5-8	-	19-26	0.63
Cosenza et al., 2014	ASM2d-SMP	R	P	UCT		GLUE	-	81.75	3.59	-	36	3.5-8	-	19-26	0.63
Sarioglu et al., 2017	DND-GP	R	P	UCT		heuristic	AQUASIM	79.3	12	7.7	12	10-12	4	33.6	0.2
Mamma et al., 2018	ASM2d-SMP	R	P	UCT		Novel	-	-	10	-	-	-	-	-	0.4-3
Bis et al., 2019	ASM2d	R	F	UCT		heuristic	GPS-X	27.1 ^a	12.64	3.6	10.4	9.8	2	0.5	0.45

Notes: NGM: new general model, P: pilot scale, L: lab-scale, R: full-scale, F: full-scale, S: synthetic, Al: Aluminum coagulant, a-BOD/TP, b-BOD/TKN, c-COD/PO₄³⁻, * PO₄³⁻ could not be simulated.

system was modeled using ASM2d, and the model parameters were calibrated using the same approach as the previous study. By and large, the ASM2d model could characterize the EPBR removal performance of sMBRs after substantially modifying the rate constant for S_{VFA} uptake rate (q_{PAO,VFA_Stor}) and rate constant for storage of X_{PAO,PP} (q_{PAO,PO₄,PP}) and taking into account the differences discussed in Table 3 (Jiang et al., 2009).

In continuation to previous research, Gholikandi and Khosravi (2012) modeled a side stream MBR using ASM2d and expanded with an SMP module to improve the nitrification prediction capabilities of the MBR as suggested by Jiang et al. (2009). The default ASM2d parameters had to modify in order to increase the anaerobic VFA up-take and aerobic phosphorus up-take with the same protocol (Jiang et al., 2009). However, in the current ASM2d-SMP model, the development of UAP delayed the fermentation process and allowed the restoration of specific PAO-related parameters (n_{μPAO}, and q_{S_{VFA}_max}) to their default ASM2d values (Table 5).

It is established to some extent that higher SRT in MBRs change the biomass kinetics. Therefore, ASMs may require calibration of the kinetic parameters before their applications to the MBR. In order to investigate the impact of SRT on model calibration, Ersu et al. (2010) used BioWin's General Model (AS/AD) to mimic the nutrient removal behavior of a lab-scale MBR at SRTs ranging between 19 and 75 days. The model was calibrated with the measured data using the heuristic approach based on sensitivity analysis. Compared to the default values, the calibrated model based on adjusted kinetic parameters at SRT of 35 days accurately predicted effluent characteristics. A sensitivity analysis revealed that effluent phosphorus concentrations were impacted by i) heterotrophic anoxic yield, ii) anaerobic hydrolysis factors of heterotrophs, iii) heterotrophic maximum growth rate iv) hydrolysis rate v) oxic endogenous decay rate for heterotrophs, and vi) oxic endogenous decay rate of PAOs. The model simulated results for COD, TN, and TP matched the experimental data reasonably well at SRT of 35 days.

An integrated new general model (Liu et al., 2011) was implemented on a UCT type MBR system to characterize its P-removal performance with/without the addition of coagulant. This integrated version couples activated sludge anaerobic digestion models and is complemented with sub-models for pH, gas transfer, and chemical precipitation. The authors calibrated the model against the measured data using the WERF protocol (Melcer, 2004). The kinetic and stoichiometric parameters related to PAOs were set to default. The model accurately predicted PO₄³⁻ with/without the addition of coagulant. The authors observed BioP-removal though it was severely impacted by the nitrates recycled from the anoxic zone to the anaerobic zone (in addition to influent nitrates). Similar results were obtained in another study where an extension to standard ASM2d was proposed to accommodate the nitrates' inhibition (Sin et al., 2008). Authors further found that adding excessive coagulant induced nitrates much more than required and negatively impacted the BioP-removal due to the limited availability of readily available COD (starvation conditions) for PAOs. The model could predict the combined biological and chemical P-removal up to a concentration of 0.025 mgPL⁻¹.

In the study presented by Cosenza et al. (2013), a UCT type, pilot-scale MBR is characterized using an integrated and complex model with 17 state variables and 79 kinetic parameters using domestic wastewater. The authors had to calibrate the rate constant for storage of poly-P (q_{PAO}, PO_{4_PP}) at a much higher value than previous studies reported in the literature (Gholikandi and Khosravi, 2012; Jiang et al., 2009). This higher q_{PAO} and PO_{4_PP}, was demonstrated by the fact that PO₄³⁻ was assimilated not only in the aerobic zone but also in the anoxic zone. The model simulation accounted for the increasing storage rate during the K₂PO₄ addition and un-modeled phosphorus release due to potential anaerobic conditions occurring within the cake layer on the membrane surface, explaining the higher q_{PAO}_PO_{4_PP} value. In addition to q_{PAO}_PO_{4_PP}, several other kinetic and stoichiometric parameters were calibrated (Table 5) to improve the fit.

Table 5
Default and modified parameters related to P-modeling in reviewed studies (modified within parenthesis).

Reference	Model											Calibration range (ASM2d/ASM2d-SMP)
	Al-Atar, 2007	Nopens et al., 2007	Jiang et al., 2008	Abegglen et al., 2008	Ersu et al., 2008	Jiang et al., 2009	Verrecht et al., 2010	Gholikandi and Khosravi, 2012	Cosenza et al., 2013	Cosenza et al., 2014	Mannina et al., 2018	
Bio-kinetic model	ASM2d + TUDP	ASM2d	ASM2d SMP	ASM3-BioP	New general model	ASM2d SMP	ASM2d SMP	ASM2d SMP	ASM2d SMP	ASM2d SMP	ASM2d SMP	–
P-related processes	11	8	8	11	19	8	8	8	8	8	8	–
P-related kinetic parameters	27	18	18	21	11	18	18	18	18	18	18	–
P-related stoichiometric parameters	11	3	3	4	8	3	3	3	3	3	3	–
A. Calibration of stoichiometric parameters												
Y_{PAO}		0.625	0.625 (0.57)		0.639	0.625 (0.57)	0.625	0.625	0.625	0.625	0.625 (0.61)	0.57–0.61
$Y_{PP,Stor,PAO}$		0.4	0.4		0.52	0.4 (0.2)	0.4	0.4	0.4 (0.442)	0.4 (0.442)	0.4 (0.58)	0.20–0.58
$Y_{Stor,PP, Ox}$					0.95 (1.50)							
B. Calibration of kinetic parameters												
$q_{PAO,VFA,Stor}$		3 (1)	3 (1)		2	3 (1)	3	3 (5.5)	3 (3.699)	3 (3.699)	3 (4.36)	1–5.5
$q_{PAO,PO4,PP}$	0.1	1.5 (1.1)	1.5 (1.1)	1.5		1.5 (1.1)	1.5	1.5 (1.0)	1.5 (2.431)	1.5 (2.431)	1.5 (2.01)	1.1–2.43
$K_{S,PP,PAO}$		0.01	0.01	0.05		0.01	0.01	0.01	0.01	0.01	0.01 (0.008)	0.008–0.01
$K_{PP,PAO}$	0.01 (0.2)				0.01							0.01–0.2
$f_{PP,PAO,Max}$	0.35	0.34	0.34	0.20		0.34	0.34	0.34	0.34	0.34	0.34 (0.71)	0.34–0.71
$f_{Gly,PAO,Max}$	0.50											
$\mu_{PAO,Max}$		1	1	1	0.95	1	1	1	1 (0.716)	1 (0.716)	1	0.716–1.0
$\mu_{PAO,Max,lim}$		1	1		0.42	1	1 (2)	1	1	1	1	1.0–2.0
n_{iPAO}		0.60	0.60	0.60	0.40	0.60	0.60	0.60 (0.45)	0.60	0.60	0.60	0.45–0.60
m_{PAO}				0.20 (0.15)	0.04							
b_{PAO}		0.20				0.20	0.20 (0.1)	0.20	0.20 (0.235)	0.20 (0.235)	0.20	0.1–0.235
no.of kinetic processes impacted due to changes in kinetic parameters	3	6	6	2	0	6	6	6	6	6	6	

Notes: Complete set of stoichiometric and kinetic parameters (Table C.3) along with their default values can be accessed in Appendix C. In addition the kinetic rates equation/processes impacted are also highlighted (Table C.4).

However, It is generally not considered as a good modeling practice (Rieger et al., 2001). A novel calibration protocol developed by Mannina et al. (2011) based on global sensitivity analysis (GSA) helped in identifying the most influent parameters and reduce the calibration effort (65% reduction in number of candidate parameters chosen for calibration).

In the study of Mannina et al. (2018), a UCT-type pilot-scale MBR was modeled using an integrated approach employing ASM2d-SMP-GHG modules. The rate constant for storage of Poly-P (PAO, PO4_PP) was calibrated and the final value ($2.01 \text{ gX}_{PP} \text{ gX}_{PAO}^{-1} \text{ d}^{-1}$) was higher than the default ($1.5 \text{ gX}_{PP} \text{ gX}_{PAO}^{-1} \text{ d}^{-1}$) because, during aerobic and anoxic environments, an increase in PAO, PO4_PP contributed to an increase in the rate of polyphosphate storage and thus limited the possibility of the accumulating nitrification related intermediate compounds. Despite high values of the kinetic parameters i.e. PAO, PO4_PP, the model simulation fitted well with the experimental data. This model has further been used to propose ways to reduce the MBR's environmental footprint using the multi-criteria optimization and estimation of greenhouse gases (GHGs) emissions (Mannina et al., 2020).

DNN-GP model (Sarioglu et al., 2017) describes the BNR process of a pilot-scale UCT type MBR, receiving real municipal waste through the

coupling of two sub-models: a) GAOs-PAOs competition and b) two step N/DN process. The GAOs-PAOs competition is modeled by adjusting their endogenous decay and substrate storage rates. Relative sensitivity analysis was used to calibrate the model parameters using the heuristic approach. Initial parameter values were selected from the published literature, and then calibrated ones were used for dynamic simulations. The authors found that temperature-dependent substrate storage and endogenous decay rates were the critical determinants of GAOs-PAOs completion. As compared to PAOs, the GAOs had a lower decay rate while PAOs had much higher maximum substrate storage rates than GAOs. Besides, GAOs remained activated even at a higher temperature and substrate, although phosphorus uptake rate was considerably reduced. The model successfully simulated the EBPR process. The P-removal performance was found around 98% even with elevated influent VFA/COD ratio. The P-removal revealed to be sensitive to DO concentrations in the aerobic/MBR tanks which is in agreement with the findings of the study presented by Sayi-Ucar et al. (2015). In a recent theoretical study (Bis et al., 2019), the CASP and MBR modeling approaches were compared. Plants were configured in a virtual environment using the GSP-X simulation tool and were characterized using ASM2d, and for MBR, an additional filtration module in submerged

membrane configuration was added. The authors found a higher concentration of PAOs in MBR than CASP, marginally higher P-removal was observed in MBR. The authors, however did not provide any detail about the calibration and validation of the model used for simulations.

A lab-scale MBR (Ersu et al., 2008) with anaerobic, anoxic zones followed by oxic plate and frame membrane zone was considered for modeling with five different recirculation arrangements (Fig. B.2). The model was built in a BIOWIN environment using “New General Model” targeting carbon and nutrient removal. The model was calibrated using the experimental data. The model could predict well effluent TN, TP, and $\text{NO}_3^- \text{N}$ but over-predicted CODs and NH_4^+ . Based on sensitivity analysis, two kinetic parameters (μ_{PAOmax} , mPAO) were calibrated. The calibrated values against the default values can be seen in Table 5.

In another study (Al-Atar, 2007), a UCT-type MBR pilot plant was modeled in the AQUASIM environment. The calibrated model was able to predict the EBPR process reasonably. The simulation reasonably fitted well with the experimental data. However, the exact concentrations for anoxic nitrate and effluent PO_4^{3-} were not predicted well. The model was slightly calibrated by adjusting the rate of poly-phosphate formation, KPP_PAO, which was increased from 0.01 to 0.2 $\text{gP}(\text{gCOD})^{-1}$ to better predict the anoxic PO_4^{3-} concentrations results showed that the sludge distribution within the anaerobic and anoxic zones is critical for P-removal.

In the study conducted by Abegglen et al. (2008), a small-scale MBR receiving toilet wastewater is modeled using the ASM3-BioP model in the SIMBA simulation platform. The MBR modeled in this study is subject to extreme load fluctuation. The EAWAG BioP module was implemented, and the decay rate of PAO (mPAO) was adjusted only by changing its value from 0.2 to 0.15 d^{-1} . The implementation of the model allows to adjust the MBR return sludge ratio to 1.2 times the influent to achieve a stable EBPR with efficiency above 90%. Theoretically, PAOs concentration was found to be in the range of 300–1500 mgCOD L^{-1} for sludge recirculation ratio of 0.6–1.7 and there was no increase in the PAO concentration above 1.2.

As presented in Table 4, ASM2d (with and with EPS/SMP consideration) is a widely used model for simulation of BioP-removal in MBRs, for lab-scale as well as pilot systems, and very limited (5) studies have been published for full-scale systems ($10\text{--}100 \text{ m}^3\text{d}^{-1}$). No study has been published for super-large scale MBR ($\geq 100,000 \text{ m}^3\text{d}^{-1}$). Perceived competitive advantage of GAOs over PAOs under the longer SRTs (thus higher MLSS and MLVSS) is countered by modifying the design configurations of the MBRs to control the SRT and feed composition (e.g., step feed configuration). ASMs can successfully simulate the BioP-removal in MBRs when fundamental differences in MBR functioning (Table 3) compared to CASP are considered. However, due to complete biomass retention in MBRs and especially the PAOs due to their increased size, several PAOs related model parameters required adjustments (Table 4) through calibration approaches. The majority of the studies considered calibrating the kinetic parameters, while few studies even calibrated the stoichiometric parameters as well, which seems necessary to accommodate the effects due to different stoichiometric rates induced by the GAOs and PAOs competition for phosphorous uptake.

2.5.3. Wastewater simulation software

Table 4 shows that the modelers have used various special-purpose simulation software, which is far less straightforward and easy to use than the general-purpose simulation environments such as MATLAB/SIMULINK or spreadsheets. The most frequently used simulation software in simulation studies in academia and the industry are; BioWin, GPS-X, SIMBA, WEST (previously EFOR), SUMO, and EAWAG's AQUASIM. Simulators like ASIM, STOAT, lynx, and JASS are primarily used in academia for research and teaching purposes. The wastewater treatment industry is also using relatively new simulators like DESASS (design and simulation of activated sludge systems) and EPD (EnviroProDesigner) to design and optimize the WWTPs. A summary

of the simulators' features based on the information available in each product's description by its respective supplier is provided as supplementary material as Appendix D.

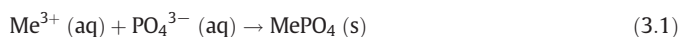
3. Chemical P-removal modeling and control

3.1. Phosphorus precipitation process

Chemical phosphorus removal, also called “removal by a salt addition,” can be applied in combination with BioP-removal as i) pre-precipitation, ii) simultaneous iii) post-precipitation and iv) sometimes side stream precipitation, which is usually adopted when P-recovery is intended (Van Haandel and Van Der Lubbe, 2007). Sometimes in one plant, metal salts are added in different points or locations along the treatment chain resulting in two or more “places” of precipitation. The metal salt is used to convert the dissolved inorganic phosphorus compounds in the wastewater into a low solubility metal phosphate, which can be removed in the process's subsequent stages. Among the various pathways; i) adsorption of phosphate onto hydrous ferric oxides (HFO) ii) co-precipitation of phosphate into the HFO structure iii) Precipitation of metal phosphate and iv) precipitation of mixed cation phosphates (i.e., calcium, magnesium, iron, or aluminum phosphates, or hydroxy phosphate) are the most commonly considered processes for removing phosphorus (Smith et al., 2008). In addition, these processes also improve the sludge properties via neutralizing the floc charge and improving the filterability by the changing the settling properties of the sludge (Lee et al., 2001; Asensi et al., 2019). Metal ions can bond with negatively charged groups within EPS and increase floc size, packing density (Asensi et al., 2019), and shearing resistance (Zhang et al., 2008).

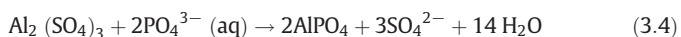
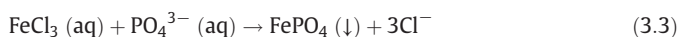
Chemical P-removal in MBR is relatively different than CASP due to i) limited or no soluble phosphorus in case of MBR effluent and metal carryover is comparatively less than CASP ii) operation of MBRs at higher SRT and thus higher MLSS which improved the flocculation and coagulation potential of phosphorus iii) addition of the coagulant at the downstream of the process in case of MBR, and iv) interaction of metal ions with EPS and membrane surface to cause/reduce fouling (a controversial in the literature). Chemical P-removal in the following sections is discussed in details keeping in view all these aspects of MBR functioning. Furthermore, a brief discussion on the fundamentals of the chemical precipitation is dedicated keeping in view the metal and hypothetical phosphate complexes assumed in chemical precipitation models.

Chemical P-removal depends upon effective coagulation and flocculation processes. Multivalent metals salts, e.g., ferric chloride (FeCl_3), ferric sulfate ($\text{Fe}_2(\text{SO}_4)_3$), ferrous sulfate (FeSO_4), ferrous chloride (FeCl_2), alum chloride (AlCl_3), and lime, are generally used as a coagulant (Van Haandel and Van Der Lubbe, 2007) in wastewater treatment to improve P-removal and the removal of higher molecular weight organics such as EPS and SMPs (Sun et al., 2019; Wang et al., 2014). The coagulation is then followed by flocculation process, which helps to form neutralized larger-sized flocs due to collision and electrostatic attraction of the sludge particles under slow mixing (Takács et al., 2005; Zhang et al., 2015). Afterwards, the larger-sized flocs formation (Asensi et al., 2019), along with phosphorus entrapped into, is removed via separation process or sludge extraction (Tchobanoglous et al., 2002). Ideally, 1 mol of the trivalent metal salt is required to remove 1 mol of phosphorus (Me/P of 1:1) as described by the theoretical stoichiometric expression (Eq. (3.1)).

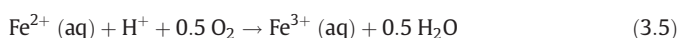


For example, when FeCl_3 is used as a metal salt, the weight ratio of Fe^{3+} to phosphorus is 1.8. Similarly, 1 mol of aluminum is required to

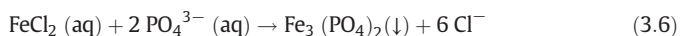
remove one mole of phosphorus. The weight ratio for Al/P is thus 0.87.



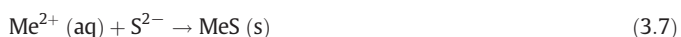
The use of ferrous sulfate (divalent metal) in full-scale MBRs is also preferred over ferric salts due to low cost (Wu et al., 2015) and comparable efficiency to ferric salts (Wang et al., 2014). When FeSO_4 is used as metal salts, it is first oxidized to trivalent metal ions (Me^{3+}) from divalent (Me^{2+}) as described by Eq. (3.5) or precipitate as vivianite ($\text{Fe}_3(\text{PO}_4)_2 \cdot 8\text{H}_2\text{O}$). Prediction of the chemical precipitation in MBRs is difficult because of oxidation of ferrous to ferric combined with several interactions between iron, phosphorus, and other ligands (Wu et al., 2015). Furthermore, among several factors that may impact the oxidation process including; i) DO concentration ii) the catalytic response by the microbes or influent constituents like sulphur and iii) inhibition by the water matrices such as carbonate and pH (Thistleton et al., 2001). It has been investigated that in-situ production of Fe^{3+} by oxidation (Eq. (3.5)) from the Fe^{2+} is a more efficient phosphate precipitant than the external addition of Fe^{3+} (Thistleton et al., 2002).



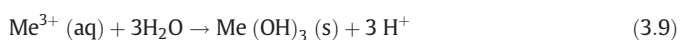
For example, when ferrous chloride is used, the weight ratio of Fe^{2+} /P is 2.7 (three moles of ferrous required to remove two moles of phosphorus) as described by the stoichiometric reaction (Eq. (3.6)).



The trace amount of sulfide coming from the sewer system due to longer retention times is also present in the influent, which may react with the divalent/trivalent metal ions and form metal sulfides (and thus remove the rotten eggs like odor) as described by the stoichiometric reactions (see Eqs. (3.7) & (3.8)). The sulfide is usually removed by the chemical precipitation process as demonstrated in the study (Gutierrez et al., 2010) wherein sulfide concentration were reduced from 7.6 mgSL^{-1} to 0.1 mgSL^{-1} .



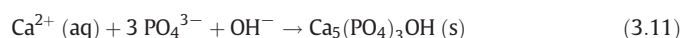
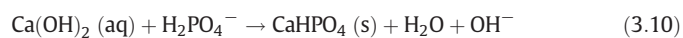
For practical applications, metal salts (inorganic coagulants) are normally dosed with higher Me/P ratios (molar ratio ranges between 1 and 4) than the theoretical stoichiometric ratio (i.e. Me/P: 1). It is presumably due to the higher pH of the sludge than the theoretical (between 5 and 6) required for precipitation of metal phosphate complexes (De Haas et al., 2000; Van Haandel and Van Der Lubbe, 2007). Secondly, a higher dose is also required because the metal ions react with water to produce hardly soluble metal hydrated complexes, e.g., $\text{Me} (\text{H}_2\text{O})_6^{3+}$ as described by Eq. (3.9). Thirdly, this excess dose is most likely explained by the competition between OH^- and PO_4^{3-} and the need of excess Fe^{3+} to destabilize FePO_4 and other colloids, and this can be examined through estimating the equilibrium log constants of the reactions (Thistleton et al., 2002). Furthermore, P-removal efficiency is affected by the influent characteristics, the discharge limit, and operating conditions (Tchobanoglous et al., 2002). The factors affecting the P-removal are discussed in Section 3.2.



In addition to the precipitation as insoluble metal phosphate (MePO_4), adsorption of positively charged hydrated metal complexes (insoluble metal hydroxide such as unbound HFO with high sorption

or amorphous ferric oxyhydroxide (AFO) helps to remove phosphorus from wastewater. If the PO_4^{3-} concentration is lower in the influent and higher metal salt is dosed owing to the competition between the formation of metal phosphate and metal hydroxides, additional metal ions will react with water (Eq. (3.9)) or OH^- present in the wastewater. As a result, an increased solids production in MBRs or increased concentrations of metal salts in CASP effluents are noticed (De Haas et al., 2000; Gnirss et al., 2003; Thistleton et al., 2002; Zhang et al., 2015).

The precipitation of phosphate as calcium hydroxyapatite ($\text{Ca}_5(\text{PO}_4)_3\text{OH}$) is the primary removal process when lime $\text{Ca}(\text{OH})_2$ is used for simultaneous P-removal (see Eqs. (3.10)–(3.12)). Since lime reacts with bicarbonate alkalinity present in wastewater to form CaCO_3 and raising the pH above 8. The concentration of lime required for P-removal is determined by the wastewater's alkalinity. The chemical reactions involved in P-removal through lime are:



Lime is not used in simultaneous P-removal applications as the pH must be raised to a value ≥ 10 for calcium to react and precipitate as phosphate and therefore not compatible for biological treatment. It increases the complexities involved in handling the process and storage of the lime in addition to complexities associated with pH handling (Tchobanoglous et al., 2002). The usage of different salts provides different results and associated problems. Therefore, a careful selection of the right kind of metal salts or coagulants is essential. Ferric salts are widely used and are more advantageous due to their low costs, least handling complexities and are slightly more effective than aluminum salts (Mishima and Nakajima, 2009).

3.2. Factors affecting chemical precipitation for phosphorus

Performance of chemical P-removal is linked to several factors, including i) influent chemistry and composition ii) operational conditions such as DO, zeta potential and pH control iii) type of coagulant, coagulant/P molar ratio and dosing location and v) mixing conditions of the coagulant with the sludge (Kim and Chung, 2014; Zhang et al., 2015). These factors can influence the types of Fe and P species present in the influent/sludge and the degree and magnitude of transition between these species (Fleischer et al., 2005; Wang and Waite, 2010). For example, when an inorganic ferric (Fe^{3+}) salt is first added to the mixed liquor, it can form AFO precipitates, which can then adsorb phosphate, organic compounds like SMP, and other dissolved constituents and may influence the membrane fouling. The precise pH conditions and local concentrations of sorbing species decide whether this process involves successive or simultaneous precipitation/adsorption phenomena between iron and phosphate. The factors influencing the chemical precipitation are briefly discussed in the following paragraphs and a summary is given in Table 6.

3.2.1. Influent chemistry and composition

The majority of early studies on chemical P-removal used simple water matrices composition (i.e., hydrogen (H), metal (Me), oxygen (DO), hydroxides, carbonates and phosphorus). Understanding the influence of more complex water chemistry is expected to enhance precipitation models' ability to predict chemical P-removal in wastewater. Chemical P-removal is affected by influent constituents, such as alkalinity, soluble and total COD, and TSS concentrations. The extent of their influence on P-removal is discussed in the following sections regardless of the technology (CASP or MBR).

Table 6

Summary of the factors influencing the chemical precipitation in MBR.

Factor	Description of the influence	References
Alkalinity	Lower influent alkalinity reduced the P-removal due to sudden changes in the pH after addition of the coagulant. The alkalinity of wastewater between 98 and 120 mg CaCO ₃ L ⁻¹ is sufficient to maintain the sludge pH close to circumneutral level (6–6.5) during the FeCl ₃ dosing and flocculation and achieving ≥98% of TP-removal.	Banu et al., 2009; Kim and Chung, 2014; Li et al., 2018
Dissolved oxygen	Higher DO (1.5–2.5 g·m ⁻³) concentration helps in better P-precipitation, mainly when ferrous (iron) is used as a coagulant.	Kim and Chung, 2014; Ren et al., 2019; Wu et al., 2019; Zhang et al., 2015
Basicity/pH	Change in pH of the sludge in MBR did not affect the P-removal though significantly higher fouling was observed at a pH range of 4–5. The optimal pH for chemical precipitation in MBR was found in the range of 6.5–7.	Li et al., 2017; Meng et al., 2009
Type of coagulant	The P-removal performance of each of the coagulants is above 85%, depending upon several other factors discussed in Section 2.3. The coagulant should be selected based on its price, regulatory acceptance and toxicity risk for the receiving bodies (in case of carryover).	Alibardi et al., 2021; Holba et al., 2012; Li et al., 2017; Ren et al., 2019; Song et al., 2008; Sun et al., 2019; Wang et al., 2014; Wu et al., 2015; Yang et al., 2011; Zhang et al., 2015
Molar ratio	Based on several studies presented Table 8 the optimum molar ratio varies between 1.5 and 4. Higher molar ratios can lead to breach of regulatory iron concentration due to carryover. Carryover in MBR is limited to soluble fraction (due to 100% solid retention) of phosphorus which do participate in floc formation. This aspect requires further study.	Yang et al., 2011; Zhang et al., 2015
Dosing location and numbers of dosing points	The coagulant's addition in the aerobic zone of the MBR provided the best results or P-removal. Two points (upstream and downstream before the filtration process/secondary clarifiers) coagulant dosing is common in full-scale plants.	Mbamba et al., 2019; Wu et al., 2015
Mixing	Single point dosing (12–14 mgL ⁻¹) can also achieved 95 ± 3% of the TP removal. Mixing had little to no effect on P-removal in MBRs. Rapid mixing improved the P-removal in both MBR and CASP. It risks to damage the microorganisms when provided with greater shear with values greater than 425 s ⁻¹ and therefore shear induced by coarse bubble aeration is sufficient for mixing of coagulant in MBR.	Alibardi et al., 2021 Kim and Chung, 2014 Autin et al., 2016; Li et al., 2019

3.2.1.1. Alkalinity. An optimum alkalinity of the influent is essential for the adequate precipitation of phosphorus. In low alkalinity ($\leq 50 \text{ mgL}^{-1}$) influent, the addition of coagulant induced sudden pH changes, creating a challenging environment and resulting in lower P-removal (Banu et al., 2009). In an MBR study (Zhang et al., 2015), it was observed that the addition of metal salt in excess resulted in consumption of the alkalinity at unprecedented rates and thus decreased the pH of the sludge (≤ 4) and slowed the hydrolysis rate of the metal. Therefore, the nitrification was substantially impaired at these low pH values. Nevertheless, nitrogen reduction can be restored by adjusting the pH to neutral levels via control system. Two other studies (Clark et al., 2000; Philips et al., 2003) investigated the iron salts' toxicity for the microbial community (AOB and Nitrobacter) responsible for nitrification process. The authors found that the metal's toxicity negatively impacted the nitrification rate contrary to the findings of previous study presented by Sun et al. (2019).

The study presented by Szabó et al. (2008) explored the influence of the P-precipitation in wastewater in the presence of excessive alkalinity (0–600 mgCaCO₃L⁻¹). The authors found that the higher alkalinity resulted in significantly higher residual soluble P (PO₄³⁻), but this phenomenon remained unexplained. A hypothesis was that the metal hydroxide (MeOH) formation occurs more rapidly in higher alkalinity waters because the hydrogen ion (H⁺) trapping potential is greater, resulting in a kinetic advantage for fast MeOH precipitation and a less likelihood of metal phosphate (MeP) and MeOH co-precipitation. The same authors also tried to explain this phenomenon by assumed competition between bicarbonate ions (HCO₃⁻) and monohydrogen phosphate (HPO₄²⁻) for active sites. This phenomenon, however, necessitates further study and confirmation. While two other studies reported contradictory findings, wherein changes in alkalinity (i.e., from 0 to 400 mgCaCO₃L⁻¹) had a negligible effect on P-precipitation (Kang et al., 2003; Newcombe et al., 2008). A relatively recent model-based study (Hauduc et al., 2015), with influent alkalinity of 125 mgCaCO₃L⁻¹ (controlled pH at 6.5 and higher mixing) resulted in more than 95% P-removal. Phosphorus precipitation with metal salts consumes alkalinity and demands supplemental addition of CaCO₃ (or NaHCO₃) to avoid dramatic pH drop to acidic range and is not suitable for the microbiology. It is therefore essential to control the pH of the wastewater treatment process (especially with alum and iron salts) and therefore must be studied along with the alkalinity.

In the absence of a sufficient number of studies and contradiction in the published literature, it is hard to establish the role of alkalinity in P-removal. Therefore, more research is needed to determine the impact of alkalinity on chemical P-removal, especially for the MBR, which is almost non-existent within the last two decades.

3.2.1.2. Initial phosphorus load. The P-removal efficiency is determined by the raw wastewater's soluble phosphorus concentration. As discussed previously, higher the initial concentration of phosphorus, lower is the Me/P ratio. The relative efficiency of the P-removal increased with the increased concentration of initial soluble phosphorus since low concentration of PO₄³⁻ are more difficult to remove than higher ones. Although, with increasing coagulant dosing P concentration decreases down to certain level (0.01 gPm⁻³), higher coagulant concentration did not help to reduce any further the residual P concentration (Smith et al., 2008; Szabó et al., 2008).

3.2.1.3. Presence/absence of colloidal organic matter. Several studies have reported the influence of organic matter on P-removal (Aleta et al., 2018; Mao et al., 2012; Szabó et al., 2008). The presence of citrate (50, 100, and 200 μM) decreased P-adsorption onto the fresh HFO (Mao et al., 2012). According to Szabó et al. (2008), increasing the influent COD and TSS concentrations resulted in lower chemical P-removal. The authors assumed that the corresponding decrease in the P-removal rate may be due to the competition to fill the binding sites available onto the metal hydroxide surface between phosphate ions and carboxylic-phenolic groups present in the organic matter.

3.2.2. Influence of operational conditions

3.2.2.1. Dissolved oxygen concentrations. Optimum DO concentration become essential with the use of ferrous salts required for Fe²⁺ oxidation to ferric (Fe³⁺) and subsequent precipitation of vivianite (Fe₃(PO₄)₂·8H₂O). These processes are limited by the oxygenation rate and sludge pH (Wu et al., 2019) specially in the anoxic chamber where DO < 0.1 mgL⁻¹ and the oxidation is slow (Zhang et al., 2015).

3.2.2.2. Basicity/pH. Phosphate and metal reactions commonly used to remove phosphate have been reported to be pH-dependent. The effect of pH on P-removal has been studied extensively (Caravelli et al., 2012, 2010; Kim and Chung, 2014; Mao et al., 2012; Smith et al.,

2008; Szabó et al., 2008). According to Smith et al. (2008), effective P-removal occurred between pH range of 5.5–7.0 for CASP. Due to other chemical reactions in the wastewater, the optimum pH range is situation-dependent. To fully remove phosphorus, the exact coagulant dose must also be calculated as function of pH and this may vary from case to case depending upon factors discussed in Section 2.3 as well as other factors discussed in the current section. On the other hand, some other studies (Caravelli et al., 2012; Szabó et al., 2008) found low precipitation of MeOH at lower pH (< 4) and formation of soluble P-complexes and phosphate precipitates with Mg^{2+} and Ca^{2+} ions at higher rates at pH greater than 10 (Szabó et al., 2008). The study of Zhang et al. (2015) investigated the effect of pH on P-removal and membrane fouling. Although the pH of the mixed liquor in MBR was within circumneutral region (6–7) and therefore the P-removal performance was not compromised, it did have an impact on the nature of membrane fouling, which was possibly due to the relative rates of formation of Fe^{3+} -SMP complexes and amorphous ferric oxyhydroxides. AFO formation was especially favored at neutral pH value (6–7), and it resulted in successful EPS removal from solution, but at the expense of filtration results, either through the formation of a Fe^{3+} -polysaccharide gel layer or pore blocking. Higher pH values (8–9) have been reported to increase the precipitation of $CaCO_3$ and results in inorganic fouling of the membrane (Meng et al., 2009). In another MBR study (Li et al., 2017) where the pH after adding the coagulant was controlled between 6.5 and 7, it was possible to achieve the P-removal above 80% with ferric and aluminum salts.

There is no general agreement on the optimum pH range for P-removal based on the reviewed literature. However, it can be inferred that successful removal occurs in the circumneutral pH range typical of WWTP.

3.2.2.3. Biomass concentration. Caravelli et al. (2012) investigated the impact of biomass concentration on chemical P-removal. For pH values below 5 and 6, biomass presence had little to no effect on P-removal. For higher pH values (7–8), biomass population significantly increased P-removal, presumably due to sweep flocculation favoring phosphate anions' retention under the alkaline conditions. Further research is needed to explore if biomass concentrations reduce P-removal due to competition mechanisms (for MeP and MeOH production) and smaller floc sizes which is the case for MBRs.

3.2.3. Type of coagulant, molar ratio, and dosing location

3.2.3.1. Type of coagulant. As discussed in Section 3.1, multivalent salts (such as ferric chloride ($FeCl_3$), ferric sulfate ($Fe_2(SO_4)_3$), ferrous sulfate ($FeSO_4$), ferrous chloride ($FeCl_2$) and alum chloride ($AlCl_3$)), lime, and polymers are used to co-precipitate phosphorus in wastewater treatment. A careful selection of the coagulant is essential to achieve the maximum P-removal efficiency in MBR while keeping the fouling low. There is a dearth of full-scale MBR studies focusing on performance assessment of different coagulants and fouling development. Ferric chloride is a widely used coagulant for P-removal and odor control via sulfide precipitation (Tchobanoglous et al., 2002). In a lab-scale MBR study, it was found that the ferric salt performs better (97%) as compared to ferrous (95%), which is comparatively cheaper than ferric with the same Fe/P molar ratio (i.e. 2) (Wu et al., 2015). Meanwhile the study presented by Song et al. (2008) evaluated the aluminum and ferric salts and found the same P-removal (98%) for both but with a 15% higher aluminum concentration (Me/P ratio < 1.5). Several MBR studies listed in Table 7 summarizes the type of coagulant used and their respective P-removal efficiency. Irrespective of the type of coagulant, above 85% P-removal could be achieved. However, this performance must not be studied in isolation from the cost of sludge production, toxicity induction, and inorganic fouling, as reported in few studies.

3.2.3.2. Molar ratio. In practice, metal dosages are usually determined based on the bench, and full-scale tests as current chemical precipitation models do not consider competing reactions (Tchobanoglous et al., 2002). MBR requires higher doses of metal salts than CASP due to higher SRT, as reported by Conidi and Parker (2015). The Fe/P molar ratio of 2–4 is optimum to achieve P-removal above 85%. P-removal achieved with different coagulant/P molar ratio in several MBR studies is given in Table 7. However, most of these studies have considered coagulant addition (low molar ratio) as fouling abatement and P-removal achieved as their secondary objective. The effect of coagulant addition on the fouling development/fouling removal, in addition to P-removal, is discussed in Section 3.3.

3.2.3.3. Dosing location. The right place for adding the precipitant to the influent or the effluent channel of the activated sludge/MBR tanks must be tested in each case, particularly regarding the essential intensive and rapid mixing with sewage flow to the treatment lane. The coagulant's addition in the aerobic zone of the A/O MBR was found to be the most effective, with P-removal efficiency over 85% (Zhang et al., 2015). A pilot-scale CASP study (Mbamba et al., 2019) analyzed the dosing of $FeSO_4$ at three different locations: aerobic, anoxic, and RAS/DEOX tanks. Feeding at the aerobic tank had a slightly higher effect on the chemical P-removal; consequently, this location is preferable due to high DO and good mixing conditions that could maximize HFO particulates and iron phosphate formation.

3.2.4. Mixing conditions

Apart from the coagulant type and dosage, mixing speed is the most critical aspect from the engineering point, given the slow P-removal kinetics. Understanding the role of mixing in P-removal kinetics is essential as almost all equilibrium-based precipitation models are based upon the assumption of ideal mixing and instant reactions (De Haas et al., 2000; Smith et al., 2008; Takács et al., 2011). Several studies have reported the importance of providing sufficient mixing for efficient P-removal, and the literature review revealed diverse results.

(Szabó et al., 2008) carried out studies with pre-polymerized metal salts rather than freshly formed salts to mimic conditions of inadequate mixing under which metal hydroxides form in the absence of phosphorus. Pre-polymerized salt removal was found to be less effective. Thus, a greater likelihood of interaction between P molecules and HFO flocs is provided by adequate mixing due to increased co-precipitation and adsorption. Mixing with a G value of 425 s^{-1} achieved almost 90% P-removal in 10–20 min, while coagulant addition during low mixing rate (20 rpm, $G = 6\text{ s}^{-1}$) hampered P-removal (Smith et al., 2008). G value of 425 s^{-1} in a wastewater treatment facility is normally undesirable since the strong shear can break up flocs. The earlier studies with higher mixing rates had little to zero influence on P-removal performance (Thistleton et al., 2002; Kim and Chung, 2014). A recent MBR study (Li et al., 2019) documented the impact of mixing on P-removal. $FeCl_3$ was dosed into the MBR with rapid mixing (200 rpm) for around 5 min. It was possible to achieve over 95% P-removal with rapid mixing.

Keeping view the literature discussed, it is hard to establish the role of mixing in P-removal effectiveness. However, it can be inferred that the mixing speed of 20–200 rpm is an optimum depending upon the time span of mixing which vary between with an inverse relationship with the rpm.

3.3. Chemical P-removal and membrane fouling

The precipitant is usually added downstream of the CASP, while in the case of MBR systems, it is added in the mixed liquor and before the filtration process or sometimes directly into the membrane tank (Gómez et al., 2013; Song et al., 2008). Therefore, the logistics and control of chemical addition and the dynamics and reliability of chemical P-removal are all more complicated in the MBR process than CASP (Zhang et al., 2015). It is now well established that the colloidal and SMPs found

Table 7
Chemical P-removal in MBR, SMP/EPS production and fouling development and/or mitigation.

Sr. no.	Reference	Wastewater	Scale	Configuration	Influent wastewater characteristics (mgL ⁻¹)					Coagulant information			Total P-removal %	Effluent P mgL ⁻¹	EPS/SMP/macro molecules Concentration	Fouling
					COD	TN	NH ₄	NO ₃ -N	PO ₄ ³⁻	TP	Coagulant	Molar Ratio				
1	Gnirss et al., 2003	RW	Lab	A/O	998	70	41	0.4	10.5	GFH	2.12	40	99.0%	0.1	NC	NE
		RW	Pilot	A/O	740	61	43	~	9.1	GFH	2.45	40	98.9%	0.1	NC	NE
2	Adam et al., 2003	RW	Lab	UCT	998	69.7	41.3	0.42	10.5	FeCl ₃	1.3	25	99.5%	0.05	NC	NC
3	Song et al., 2008	SW	Lab		45	35.4			11.1	AlCl ₃	1.51	30	98.0%	0.05	NC	↓
		SW	Lab		27	30.8			13.1	FeCl ₃	1.28	30	98.0%		NC	↓
4	Mishima and Nakajima, 2009	SW	Lab	A/O	560					FeCl ₃	1.0	2260	92.5%		↓	↓
		SW	Lab	A/O	25					FeCl ₃	2.0	4520	99.7%		↓	↓
5	Gómez et al., 2013	RW	Pilot	A/O	250		49	1.4	4.6	FeCl ₃	2.0	12.4	84.7%	0.7	↓	NC
6	Yang et al., 2011	RW	Pilot	A/O	196	20.5	18.5	1.2	2.9	PFC	2.4	12.5	91.0%	0.26	↓	↑
7	Holba et al., 2012	RW	Pilot	MLE	724		40.8		4.2	FeSO ₄	0.5	3.5	84.7%		↓	NC
		RW	Full	MLE	1140		105		8.7	FeSO ₄	0.2	3.5	85%		↓	NC
		RW	Full	MLE	655		73.4		14.5	FeSO ₄	0.1	3.5			↓	NC
8	Wang et al., 2014	RW	Pilot	UCT	400	50	25		9	FeCl ₃	2		>96%	0.05	↓	↑
		RW	Pilot							FeSO ₄	2		>96%	0.05	↓	↑
9	Zhang et al., 2015	SW	Lab	UCT	400	60	25		10	FeCl ₃	4		99.8%	0.02	↓	↑
										Fe ₂ (SO ₄) ₃	2		99.6%	0.03	↓	↑
10	Wu et al., 2015	SW	Pilot	A/O	400	50			8	AlCl ₃	2		95.2%	0.38	NC	NC
			Pilot							FeCl ₃	2		96.7%	0.26	NC	NC
11	Li et al., 2017	RW	Lab		50	17	15		2	Ferric	5.6	20	80.3%	0.29	↓	↓
12	Lee et al., 2017	SW	Lab	UCT	293	24.3	20.7	0.4	4.25	AlCl ₃	17.2	30	80.3%	0.33	↓	↓
13	Li et al., 2018	RW	Lab		360	28.5			6.35	PAC	3.4		85.5%	0.62	↑	↑
14	Wu et al., 2019	RW	Pilot	UCT					3.05	Fe ₂ (SO ₄) ₃	1.8	20	95.6%	0.28	↓	↓
15	Sun et al., 2019	RW	Lab	UCT	268.1	40.4	38.6		4.97	AlCl ₃	2.3	10	85.4%	0.73	↓	↓
16	Ren et al., 2019	SW	Lab	A/O	425	45	27.5		10	Fe ₂ (SO ₄) ₃	1		96.9%	0.31	NC	↓
			Full		579	26.9			9.18	Fe ₂ (SO ₄) ₃	3.75	14	96.2%	0.38	NC	↓
17	Alibardi et al., 2021	RW	Full	A/O									95%	0.46	NC	NC

SM-synthetic wastewater; RM-real wastewater; NC- not confirmed; NE- not confirmed; PAC- Poly-aluminum chloride, GFH- granular ferric.

in mixed liquor are mainly responsible for membrane fouling (Gao et al., 2013). These products have a similar size to membrane pores and often appear to form impermeable gels on the membrane surface (Meng et al., 2017). The addition of precipitant (if not dosed optimally) into the MBR is assumed to be influential for organic, inorganic, and biofouling in addition to the P-removal (Hai et al., 2018; Meng et al., 2017).

As extensively discussed in the MBR fouling literature, inorganic fouling in MBRs is caused by biological and/or chemical precipitation of the metal cations (e.g., Fe^{3+} and Al^{3+}) and anions of ionizable biopolymers (e.g., as PO_4^{3-} and SO_4^{2-}) onto the membrane (Iorhemen et al., 2017; Wang et al., 2008). Several studies investigated the addition of a small concentration of coagulant into the MBR with the intention to control membrane fouling. It reduced the extent of the TMP evolution, which is assumed to be attributed to the large-sized floc formation (formed due to neutralization) and reduced organics concentration in the supernatant (reduction of organic fouling), limiting the pore blockage (Fan et al., 2007; Fleischer et al., 2005; Koseoglu et al., 2008; Wu et al., 2006; Zhang et al., 2008). In addition, it has been perceived that an optimum coagulant addition decreases the compressibility of the sludge flocs, changes the particle size distribution (PSD), reduces the concentration of EPS (and thus the reduction in the biofouling), increases the porosity of the membrane, and increases the MLSS filterability (Gómez et al., 2013; Song et al., 2008; Sun et al., 2019). Finally, it is assumed to pose an unknown risk to the membrane life when operated over more extended periods (Gnirss et al., 2003) and requires further research.

As described by Song et al. (2008), the hydraulic resistance of the cake is influenced by the coagulant addition, and a noticeable reduction was observed with the addition of coagulant above 200 mgL^{-1} . Usually, higher concentrations (*Fe/P in the range of 2–4*) are used for P-removal, and therefore results of these studies might be irrelevant to understand the trade-off between the P-removal and membrane fouling. If the objective is complete P-removal, the Fe/P ratio is adjusted in the range of 1.5 and 4 (Sun et al., 2019; Takács et al., 2011; Zhang et al., 2015). Thus, an in-depth analysis of various studies focused on P-removal and membrane fouling is essential to derive the conclusions.

In addition to P-removal, coagulant (alum) addition enhanced membrane filtration performance and reduced the membrane fouling due to incorporation of the colloidal solids into the flocs (later removed with sludge) rather than sticking onto the membrane surface (Fleischer et al., 2005). Effect of ferric chloride and alum addition was investigated on P-removal, and on membrane fouling in a lab-scale study (Song et al., 2008). The authors found that the alum addition (13.3 mgAlum/mgP) resulted in 98% P-removal and reduced the specific cake resistance due to increased sludge particle size (from $45 \mu\text{m}$ to $57 \mu\text{m}$). It was further found that the nitrification process was not impacted by the addition of the FeCl_3 , while the phosphorus was removed and the membrane fouling effectively controlled. However, it reduced the pH to almost half (≈ 3.4) of its initial value as the concentration increased from 0 to 500 mgL^{-1} .

In another lab-scale study, the addition of FeCl_3 ($\text{Fe/P ratio} = 2$) successfully removed 99.7% of the phosphorus from the synthetic wastewater and decreased the fraction of SMPs (having protein and carbohydrates sizes falling between 0.1 and $0.4 \mu\text{m}$) by 50% and thus reduced the membrane fouling propensity due to increased floc size (Mishima and Nakajima, 2009). Another pilot-scale study (Yang et al., 2011) employing polymeric ferric chloride (PFC) revealed an increase in membrane fouling. The authors conducted cake layer analysis using a three-dimensional excitation-emission matrix (EEM) and gel filtration chromatography (GFC). The analysis indicated that higher molecular weight organics ($> 1000 \text{ kDa}$) were the main contributors of the membrane biofouling. Fourier transform infrared spectrum (FTIR) analysis further confirmed that these higher molecular weight organics were mostly protein and polysaccharides. The same authors also observed the formation of a dense and nonporous gel layer onto the membrane

surface with clusters of bacteria and bio-polymers with help of a scanning electron microscope (SEM). Besides, X-ray elemental diffractometric analysis elucidated that ferric metal was the most prominent foulants responsible for inorganic fouling of the membrane.

The study of Holba et al. (2012) investigated the effect of lower coagulant concentration on P-removal and SMP production at pilot and full-scale. A fixed coagulant dose of $3.5 \text{ mgFeSO}_4\text{L}^{-1}$ was used for all three systems (one pilot and two full-scales). A better P-removal efficiency (84.7%) with a 68% reduction of SMPs was observed in pilot-scale plant. However, large load fluctuations and constant coagulant dosing in the full-scale plant caused in unsatisfactory P-removal. In another pilot-scale MBR study (Wang et al., 2014), similar effects have been documented where the coagulant dosing required optimization to balance the enhanced P-removal and coagulant-driven membrane fouling. The addition of metal salt (i.e. FeCl_3) resulted in 89%–97% P-removal efficiency and produced more sludge (Chae et al., 2015). In a lab-scale investigation (Sun et al., 2019), the addition of metal salt (20 mgL^{-1} of $\text{Al}_2(\text{SO}_4)_3$) resulted in achieving over 90% P-removal and reduced TMP (from 11.3 kPa d^{-1} to 0.57 kPa d^{-1}) due to a decline in EPS concentrations and accumulation of particles on the membrane surface and increase in the particle size. The study of Asensi et al. (2019) revealed that the addition of FeCl_3 (in neutralized condition) linearly increased the floc size and resulted in more compact floc structure. This increases the settling properties of the sludge to an extent where the coagulant concentration reaches to an optimum level and beyond that floc is not able to retain the coagulant and the settling properties are then negatively impacted. This suggests that while modeling the membrane fouling using phenomenological models, changes in the settling properties of the sludge due to addition of coagulant should not be ignored. In another pilot-scale study (Gómez et al., 2013), the addition of $12.4 \text{ mgFeCl}_3\text{L}^{-1}$ improved the P-removal efficiency from 54.7% to 84.7%. The authors observed reduction in SMP concentration with the addition of the coagulant. The X-ray elemental diffractometric analysis of the cake layer found a significant increase in the Fe^{3+} concentration and a substantial contribution to inorganic fouling. The same authors further noticed that, at lower coagulant concentrations, it was not easy to ascertain the trade-off between the fouling reduced by the SMP removal and Fe^{3+} deposition and resulting inorganic fouling. The higher coagulant doses resulted in significant pH decrease and precipitated TP present in the sludge, resulting in a lack of available phosphorus for PAO to grow on and participate in the EBPR process. This becomes pertinent for MBR systems, which are operated at higher biomass concentrations as compared to CASP.

As phosphorus is essential for microbial communities other than PAOs, participating in nitrification and denitrification processes (Daigger et al., 2010). Similarly, Zhang et al. (2015) found that the precipitant dosing lowered the membrane fouling when with Fe/P molar ratios < 1 . However, P-removal was compromised and severe fouling (due to Fe-rich gel layer) was observed with higher molar ratios due to the formation of Fe-SMP complexes and amorphous ferric hydroxides ($\text{Fe/P} > 2$). The severity of the membrane fouling was observed when coagulant was dosed in the anoxic zone compared to dosing in the aerobic zone (much closer to membrane). The authors further found that molar ratio of 2 for Fe^{3+}/P successfully removed 99% phosphorus without impeding the nitrification/denitrification. However, it impacted the nitrification process significantly with a higher dose (molar ratio of 4) due to increased consumption of alkalinity associated with a decrease of pH value to almost 4 and thus required the pH adjustment. Minimizing overall iron dosage is also important, not only for cost reasons but also to avoid unwanted side effects like membrane fouling. Furthermore, the same authors found that lowering the phosphorus concentrations to 0.1 – 0.2 mgPL^{-1} using higher coagulant doses did not inhibit nitrification. Instead, the harmful effects of the chemical precipitants (Fe^{2+} and Fe^{3+}) were blamed for suppressing nitrification and denitrification due to toxicity induced by the metals as reported by Philips et al. (2003).

Some studies used flotation (DAF) and biologically aerated filters (BAF) as post/pre-treatment to membrane filtration to improve the P-removal performance and as well as to counter the inorganic fouling caused by the metal deposition onto the membrane surface (Lee et al., 2016; Li et al., 2017). Similarly, Ren et al. (2019) reported the change in the microbial composition due to addition of ferrous in a ceramic MBR with a molar ratio of 2. The authors found that the relative abundance of the aerobic denitrifying bacterial community "Zoogolea" increased consistently. While the population of other bacterial community including "Dechloromonas, Hyphomicrobium and Thaueria (anoxic denitrifying bacteria), Nitrospira (NOB) and Candidatus Accumulibacter (PAO) which is responsible for BioP-removal reduced sharply due to toxic effects of iron dose on bacterial physiology.

A trade-off between precipitant dose, fouling, and P-removal is reported differently for different studies, and hence further research is needed to explore the relationship between the type (cake formation, pore blocking, etc.) and quantum of the fouling induced (Loderer et al., 2015; Zhang et al., 2015). Majority of the MBR studies summarized in Table 7 are either lab-scale or pilot scale units while experiences of the P-removal and fouling abatement/fouling increase is limited and therefore more research is needed in this direction.

3.4. Modeling chemical precipitation of phosphorous

Several chemical precipitation models were developed for P-removal in wastewater systems, and very few of them have been adopted for precipitation modeling in CASP and MBRs. The precipitation model commonly used in combination with ASM models has been discussed briefly regarding its conceptualization and processes involved, application in MBR modeling, and the P-removal limitations. Mainly ASM2d is being used for modeling chemical P-removal in MBRs. Chemical precipitation models employ three main approaches i) chemical equilibrium approach, ii) kinetic model approach, iii) combined chemical equilibrium-kinetic approach and geo-chemical complexation (De Haas et al., 2000; Smith et al., 2008). These precipitation models are briefly reviewed for quick comparison in terms of processes considered, limitations, and their usefulness for coupling with the biological and filtration model in the MBR case. Although this review's scope is limited to the last two decades, it is still worth encompassing the modeling knowledge briefly beyond two decades.

3.4.1. Equilibrium-based models

Chemical precipitation modeling gained importance in the late 1970s, and Ferguson and King (1977) introduced the first equilibrium-based model using empirically derived solubility products. The model employed co-precipitation as the sole mechanism for removing PO_4^{3-} . It did not consider the removal of TP or even removal of precipitated orthophosphate. The molar ratio for Al/P in the precipitate is deemed to be constant at all pH values. The model was sufficient to describe the effects of alum dosage qualitatively and pH observed in several experimental studies. However, the model is based on a solubility equilibrium of a fictitious precipitate, i.e., $\text{Al}_{1.4}\text{PO}_4(\text{OH})_{1.2}$, and solubility products are empirically derived. Furthermore, the model did not consider pathways other than co-precipitation for the P-removal. It also did not include the effect of the competition of ions such as calcium, magnesium, iron, or hydroxy phosphate.

The previous model's limitation led to the development of an improved P-removal equilibrium model with ferric salt employing simultaneous precipitation and adsorption mechanisms (Luedecke et al., 1988). The model consists of i) mass balance equations (based on equilibrium) describing the acid-base reactions for dissociation (by hydrolysis) of PO_4^{3-} species and ferric ion-pair complexation with phosphate or OH^- ii) mass balance equations for phosphate and ferric iii) equations defining the split between the type of precipitate formed ($\text{Fe}_3\text{PO}_4(\text{OH})_{3-3}$ and FeOOH) at given pH and iv) adsorption equilibrium. The model has four parameters (Stoichiometric coefficient, solubility

product, equilibrium constants ferric phosphate, and adsorption coefficient) with unknown values, estimated from the jar test under aerobic conditions and pH control. The model is again based on fictitious precipitates ($\text{Fe}_3\text{PO}_4(\text{OH})_{3-3}$ and FeOOH), and the solubility products are thus empirically derived. The adsorption coefficient is modified to each experimental condition over a wide range, inferring that the hypothetical adsorption mechanism is over-simplified and does not fully describe the actual adsorption phenomenon (De Haas et al., 2000; Hauduc et al., 2015).

The Water Environment Federation (WEF) suggested a chemical equilibrium-based precipitation model that took into account the amount of PO_4^{3-} removed as well as the pH sensitivity and is based on hypothetical ferric phosphate precipitate, i.e., $\text{Fe}_{1.6}\text{H}_2\text{PO}_4(\text{OH})_{3.8}$, which has been verified to be non-existent (Smith et al., 2008). The model is provided for both ferric and aluminum salts. Stoichiometric reactions are used to measure iron/alum dose close to a residual of $1.6 \text{ mgPO}_4\text{L}^{-1}$, at which point the equilibrium is reached, and the competition reaction starts. Additional metal salt is needed to achieve effluent P-removal below 1 mgPL^{-1} since stoichiometric estimations are no longer reliable. In addition to metal phosphate precipitate, metal hydroxide is formed. The model predicts a minimum of $35 \text{ }\mu\text{gPO}_4\text{L}^{-1}$ at an optimum pH of 6.95 (Takács et al., 2005). The model was then recalibrated by Takács et al. (2005) by combining it with equilibrium-based pH calculations, enabling the solution's ionic composition and termed as 'enhanced WEF'. The model was later implemented in BioWin to mimic the P-removal in full-scale facilities as a sub-module to the bio-kinetic model. The current version of the WEF model available in BioWin has improved the limitation caused by the fixed Fe/P molar ratio. The model is sufficient to estimate the pH, iron dose, and sludge production. Although it does not take into account the P-removal pathways other than co-precipitation.

3.4.2. Models based on empirical kinetics

The bio-kinetic model (ASM2d) was extended with two chemical processes (precipitation and re-dissolution) to simulate two hypothetical compounds, namely metal hydroxide (X_{MeOH}) and metal phosphate (X_{MeP}). The model assumed that these two processes are opposite of each other and are in equilibrium at steady state conditions (Gujer et al., 1995). The precipitation and re-dissolution are modeled as simple first-order reactions using process kinetic rate (K_{pre} and K_{red}). The authors used the example of ferric ion dosing in an activated sludge environment. MeOH is assumed to be $\text{Fe}(\text{OH})_3$ in this situation, and MeP is considered FePO_4 . The model does not account for the loss of hydroxide ions (a reduction in alkalinity) from the bulk process during the formation of $\text{Fe}(\text{OH})_3$, but pH is assumed to be near neutral. The model does not provide any guidance for the estimation of the kinetic rates. Neither gives any indication for the model applicability with alum salts or other coagulants. It is mostly used in WEST software as standalone as well as with the bio-kinetic model (ASMs).

3.4.3. Models based on combined chemical equilibrium-kinetic approaches

Based on the steady-state equilibrium model of Luedecke et al. (1988), a dynamic model employing processes including i) co-precipitation, ii) dissociation, iii) hydrolysis, iv) adsorption, and v) biological nutrient requirements; was proposed by Briggs (1996). The model's basis is almost the same as inherent one with further addition of rate expressions for the precipitation/dissolution and adsorption/desorption. The Elovich equation (Manchado et al., 1989) is used to model P-adsorption, with switching functions for the residual phosphorus and full-adsorption potential. Besides, it considers the release of the adsorbed phosphorus during the dissociation process. The model further considered the biological components affecting the chemical phosphorus removal. Since the model is based on the same assumptions as of Luedecke et al. (1988), it inherited the same limitations. Furthermore, the model was found unable to simulate the effluent's solids concentrations and thus its impacts on effluent total P concentrations.

Table 8
Comparative assessment of the chemical P-removal models.

Model reference	Model characteristics							
	Ferguson and King, 1977	Luedecke et al.,1989	Briggs et al.,1996	WEF Model,1998	IWA-ASM2d Model, 1999	Smith et al., 2008	Hauduc et al., 2015	Giwa and Hasan, 2015
Modeling approach								
Kinetic					✓			
Chemical equilibrium	✓	✓		✓		✓	✓	
Combined kinetic-equilibrium			✓					
Geo-chemical reaction						✓	✓	
Electro-coagulation								✓
System behavior (change in concentration over time)								
Steady state	✓	✓		✓		✓	✓	
Dynamic			✓	✓		✓	✓	✓
Removal mechanisms considered								
Hydrolysis		✓	✓					
Dissociation		✓	✓					✓
Precipitation		✓			✓		✓	
Co-precipitation	✓		✓	✓		✓	✓	
Adsorption		✓	✓			✓		
De-sorption			✓					
Surface complexation/aging							✓	
Biological nutrient requirements			✓					
Dissolution			✓					
Coagulants considered								
Aluminum	✓		✓	✓				✓
Ferric		✓	✓	✓	✓		✓	
Type of model with respects to calculations								
Empirical	✓	✓	✓					
Mechanistic					✓	✓	✓	✓
pH dependence	✓	✓	✓					
Alkalinity dependence						✓		
Influence of aging/surface consolidation						✓		
Ability to predict low P-concentration			✓	✓	✓	✓	✓	✓
Limited	✓	✓	✓					
Excellent				✓	✓		✓	
P-species considered								
Ortho-phosphate	✓		✓	✓	✓		✓	
Total phosphorus		✓						
Influent P-fractionation			✓					

3.4.4. Models based surface complexation and chemical equilibrium

Current equilibrium models are based on dissociation and solubility principles and cannot account for variable precipitate stoichiometry and time dynamics. The model proposed by Smith et al. (2008) employs geochemical principles in combination with chemical equilibrium. This model takes into account the interactions between amorphous HFO and P (i.e. adsorption and co-precipitation). The model uses an active site factor (ASF) to describe the availability of reactive oxygen atoms or "surface sites" for P complexation before, after, and during precipitation. The value of the ASF was found to be a function of parameters i.e. dosing, mixing, pH and aging conditions already discussed in detail in Section 3.2. The surface complexation model (SCM) has many benefits over previously used equilibrium and kinetics based models, including the fact that it is based on known chemical elements, as opposed to equilibrium or kinetic models which are based on fictitious precipitates. It also incorporates surface geochemical interactions, which were neglected in previous equilibrium and kinetic models. The SCM is best suitable to predicting behavior in systems with low effluent P limits. The model is based on simple hydrogen-iron-P system and completely ignores the variable water chemistry. Furthermore this model is not capable to describe the kinetic behavior of P-removal i.e. initial fast removal followed by the slow removal and the influence of HFO aging as observed by Szabó et al. (2008).

A recent study proposed a mechanistic chemical P-removal modeling method that explains HFO precipitation and flocculation, as well as P-adsorption onto HFO particulates and co-precipitation (Hauduc et al., 2015). All process reactions are described using kinetic rate

expressions. The aging aspect of the model described by Hauduc et al. (2015) is one of the main modifications to the model by Smith et al. (2008), and therefore the model can provide more details than the previous version. There are still some limitations to address: i) HFO aging was calibrated to experimental data with a maximum age of 30 min and does not account for solid ages typical of wastewater treatment and ii) The effects of pH, TSS, and COD were not investigated.

Table 8 presents a comparative summary of the briefly presented models. The literature review has revealed that models now have improved the ability to predict the effluent P-removal for low concentrations. However, these are still unable to reliably describe the removal kinetics and impacts of the metal complex aging with higher SRTs. Not even a single study has been reported for their application and validation with the system operating at higher SRTs such as MBR. The recently developed model (Hauduc et al., 2015) yet requires validation at full-scale. Furthermore, the models have not been tested for their ability to predict chemical P-removal behavior in complex waters, where organics and other organisms could obstruct removal mechanisms. Improving the models' ability to explain these processes would result in more stable effluent P-levels and chemical sludge output at lower dose rates, resulting in cost savings in chemical and sludge treatment.

3.5. Application of chemical precipitation models to MBRs

There is a shortage of studies reporting the chemical precipitation model's application to MBRs operated at higher SRTs, and floc size is already smaller and sticky due to the higher concentration of EPS/SMPs.

Two of the reported MBR studies used ASM2d's kinetic precipitation model (Daigger et al., 2010; Fleischer et al., 2005) and did not provide any details about the kinetic and stoichiometric parameters and their adjustments. Meanwhile (Liu et al., 2011) used an enhanced WEF model available in BioWin simulation software with its default kinetic and stoichiometric parameters.

4. Control systems for P-removal

The conventional controllers (e.g., P, PI, PID) and advanced controllers (e.g., predictive, robust adaptive, sliding, and multi-model) have successfully been adopted in wastewater for process improvement and optimization. Artificial intelligence-based model controls such as artificial neural networks (ANN), deep learning neural networks (DNN), fuzzy techniques, and hybrid controls (neuro-fuzzy) are adapted in wastewater (Garikiparthi et al., 2016). However, their success is not satisfactory for complex phenomena such as P-removal (Sin and Al, 2021). Logarithm-based phosphorus control systems are deployed and are automated using SCADA to reduce consumption of the precipitants (to reduce cost) and maintain effluent compliance. The control systems used for precipitant addition are based on: i) continuous fixed dosing, ii) scheduled dosing, iii) influent proportional flow dosing-feed ratio controller, iv) phosphorus load proportional, and v) feedback control or PI controller (Garikiparthi et al., 2016; Kern, 2016). The chemical precipitation control system can be implemented with CASP and the MBRs or with any other technology requiring additional P-removal to meet the discharge limits (Shiek et al., 2020). The majority of the published studies in the precipitant dosing control domain are related to CASPs, while few studies have been found related to the MBR (Alibardi et al., 2021; Mbamba et al., 2019).

The most straightforward dosage controls have historically been based on flow rate and turbidity measurements. Still, a primary feedback controller was used to achieve outstanding control efficiency and substantial chemical savings with an online in situ phosphorus sensor in place (Ingildsen, 2002). The sensor was mounted in the flocculation chamber located near the effluent discharge point. The sensor-based online phosphorus measurements enabled (Devisscher et al., 2002) to monitor the chemical dose required to remove the phosphorous as per the set-point. The study of Craig et al. (2014) evaluated the application of a feedback-based "off the shelf" control system for precipitant dosing on full-scale extended aeration-based CASP. The system monitored the PO_4^{3-} concentration at the secondary clarifier's exit and controlled the ferric feeding upstream. The controller used an advanced algorithm capable of calculating the P-load and the required ferric dose, taking into account the stoichiometric relationship. This automated chemical P-removal system resulted in a 56% reduction in ferric consumption and fully recovered the investment in just 6.8 months. Another study (Garikiparthi et al., 2016) evaluated the application of four different controllers, i.e., feedback, feedforward and feed-ratio, and fixed-rate dosing, for a CASP to control the effluent phosphorus concentration by regulating the coagulant dosing and saving the chemical costs. The authors found that the feedback controller improved the P-removal by 52.54% and performed better than others. However, the same authors also found that the phosphorous removal (EQI) and the P-removal (OCI) cost are conflicting and, therefore, suggested further research of the plant-wide dosing control. In a most recent full-scale MBR study (Alibardi et al., 2021), application of feed-forward control based on PO_4^{3-} load proportional, reduced the iron consumption from 14 mgL^{-1} to 12 mgL^{-1} . The schematics of various control systems discussed in this section are given in Appendix E.

5. Knowledge gaps and future research directions

This review is dedicated to summarize the current 'state of the art' in progress toward understanding of the P-removal in MBR, and application of existing bio-kinetic and chemical precipitation models. Keeping

in view the findings from large number of scientific publications considered, identified research gaps and speculations about the future research are presented in this section.

The review of the global regulations (Appendix A), revealed that the majority of the countries have based their discharge regulations considering TP concentrations while eutrophication is mainly caused by bio-available fraction of the phosphorus (Preisner et al., 2020) which mainly included PO_4^{3-} . Therefore, it is recommended to revisit the phosphorus discharge limits and should be ideally based on PO_4^{3-} concentrations. However, literature review revealed that there is no standardized and globally accepted approach for classifying the phosphorus fractions and this presents an urgent need to address this issue along with the standardized notation of each fraction and corresponding testing method (Rosario et al., 2021). Despite overall effectiveness of different wastewater treatment technologies for TPremoval is well known, there is a dearth of studies documenting the effectiveness of various technologies for removal of specific fractions of phosphorus. Most of the full scale application use EBPR and chemical precipitation in conjunction to maximize the P-removal and therefore sufficient data is not available to quantitatively describe how much of the P-removal is due to EBPR or chemical precipitation. Further research is required to assess the degree of BioP-removal that can be achieved in full scale MBRs that requires chemical addition to reliably meet the discharge limits.

EBPR is well established and yet poorly understood process for P-removal from wastewater (Bunce et al., 2018). Full-scale facilities are still being challenged by the process instability of EBPR process due to lack of understanding of the role of microbial communities, especially PAOs and their selection in anaerobic zone in comparison to GAOs. Over the last five decades, primary PAO responsible for EBPR has been changing from *Acinetobacter* to *Accumulibacter* and most recently *Tetrasphaera* have been found in large number of full scale CASP facilities (Hauduc et al., 2015; Mielczarek et al., 2013; Stokholm-Bjerregaard et al., 2017). This is probably because of the fact that PAOs responsible for EBPR have not been identified in pure culture neither GAOs responsible for deterioration of EBPR. Theoretically, anaerobic zone is compelling requirements for proliferation of PAOs and their selection for P-removal, while some full-scale studies without having an anaerobic zone have demonstrated 50–68% removal efficiencies with operating conditions not suitable to EBPR process (Gabarrón et al., 2014; Itokawa et al., 2014). Future research should be dedicated to enhance the EBPR process stability in MBRs by improving the MBR design configurations and optimizing operational conditions without compromising its capabilities to remove carbon and nitrogen. Majority of the full-scale MBR facilities are anoxic-aerobic or MLE configurations while UCT and MUCT are widely in research at bench and pilot scales. To the best of authors' knowledge, no full-scale study is dedicated to assess the BioP-removal in relation to the microbial composition involved in each MBR configuration and unit cost of biological and chemical precipitation should be investigated to present a fair comparison.

The bio-kinetic models appeared to be successful in simulating the P-removal in MBRs, provided fundamental differences in CASP and MBR functioning (Table 3) are considered in the modeling approach. It has been found that ASMs when applied to MBR require substantial modifications of the stoichiometric and kinetic parameters (Table 5) to adjust the lack of understanding in details and dynamics of the process as a generalized EBPR model. Efforts dedicated to develop systematic calibration procedures have resulted in decreasing the required calibration efforts. However, the calibration processes lacks identification of bio-kinetics involved in parametric adjustments and results in diverse range of parameters values. Moreover, the existing model consider only one PAO community i.e. *Accumulibacter* while literature suggested a number of PAOs communities are responsible for EBPR and therefore bio-kinetic models require improvements to accommodate the role of other microbial communities as well. A combination of metabolic and kinetic models can be helpful in apprehension of the complex

biological interactions in relation with their population dynamics and thus improving the EBPR modeling (Al-Atar, 2007; Hauduc et al., 2015). Some experimental studies have investigated that the EPS/SMP play a positive role in BioP-removal (Adoonsook et al., 2019; Ichihashi et al., 2006; Jiang et al., 2009; Zhang et al., 2013) while the current ASMs do not consider this phenomenon. It is therefore proposed to undertake full-scale studies to rule out the difference in lab-scale to full-scale models' applications.

Chemically enhanced P-removal (CEP) is a well-established yet again poorly understood because of formation of unknown metal complexes and multiple pathways involved. P-removal in MBR is different than CASP due to specificities involved in its functioning and risk of fouling development (Mbamba et al., 2019; Sun et al., 2019). Moreover, the effect of MLSS concentration, floc size (which is smaller in MBRs) on CEP performance should be investigated considering the competing mechanism for MeP and MeOH production. Diverse results have been reported for fouling development/abatement and P-removal trends in various bench-pilot scale studies, while experiences from the full scale facilities are yet required to be explored. A dedicated research is therefore needed to explore the effect of coagulant addition on membrane fouling in addition to P-removal along with clear description of type and quantum of the fouling induced. Precipitation models have now good prediction capabilities yet unable to reliably describe the removal kinetics and impacts of metal complex aging at higher SRTs which is the case in the MBRs. Further research is need to explore the applicability of these precipitation models at full-scale MBRs operating at higher SRTs and with smaller floc size, generally undesirable for effective coagulation and operating under higher shear induced by the coarse bubble aerations.

The control of coagulant and carbon dosing, recirculation rates and DO concentration are key factors in achieving maximum P-removal in MBRs. There is a shortage of studies documenting the role of the automatic controls for chemical dosing in MBRs and techno-economic assessment of the proposed control systems. Future research should be dedicated to apply the various instrumentation and control systems for advanced automated controls for enhanced and cost effective P-removal.

6. Conclusion

MBRs are successful in P-removal below regulatory limits by employing combined EBPR and chemical precipitation mechanisms. P-removal in MBR is a complex and poorly understood phenomenon which is sensitive to design configurations (MLE, A/O, UCT, A²/O, VIP and Bardenpho), influent characteristics and several operating parameters related to EBPR (Microbial composition, MLSS, SRT, HRT, temperature and DO) and chemical precipitation (influent chemistry and composition, DO, pH, MLSS, Me/P molar ratio, dosing location and number of dosing points and mixing conditions). It is hard to establish the role of EBPR and chemical precipitation methods when used in conjunction. The bench and pilot scale UCT and MUCT configurations provide excellent control over nitrates interference in the anaerobic zone resulting in higher BioP-removal efficiencies (up to 90%). However, MLE and Bardenpho are widely used configurations at full-scale facilities. MBR can provide enhanced Bio-P removal, if:

- The influent COD/TP, BOD/TP, COD/TN ratio are provided in the ranges of 2.9–45, 12.33–45, and 5.3–7.3 respectively. Furthermore, influent nitrate are kept lower than $1 \text{ g} \cdot \text{m}^{-3}$ to keep the anaerobic zone efficiency intact with PAOs proliferation.
- The MLSS concentration is controlled under $7 \text{ g} \cdot \text{m}^{-3}$ with SRT in the range of 15–40 days. Anaerobic and aerobic HRT are controlled in the range of 2–2.9 h and 3–6 h respectively, while total HRT is kept around 10 h.
- 7–10% of the sludge biomass are PAOs.
- The DO concentration of $0.4\text{--}2.5 \text{ g} \cdot \text{m}^{-3}$ is maintained in the aerobic zone

Similarly, chemically precipitation in MBR can provide excellent results, if:

- Alkalinity of the influent wastewater is within $98\text{--}123 \text{ CaCO}_3 \text{ g} \cdot \text{m}^{-3}$
- The pH of the sludge is controlled near circumneutral level (6.5–7)
- Relatively higher DO concentration ($1.5\text{--}2.5 \text{ g} \cdot \text{m}^{-3}$) are maintained in the aerobic zone
- Metal to Phosphorus molar ratio is adjusted in the range 1.5–4 with an optimum ratio of 2.5.
- Two point dosing is practiced, first dosing at the inlet or at 3/4th of the anoxic reactor and 2nd dosing in the aerobic zone.
- Rapid mixing is provided with G values ranging between 100 and 200 s^{-1}

Several bio-kinetic, precipitation, filtration as well as integrated models are available to simulate the MBR process including P-removal. The addition of the precipitant affects the relative abundance of microbial community sludge composition and fouling phenomenon. Therefore, integrated models should have provision to adjust the stoichiometric changes induced due to change in the microbial composition and competition among them as well as the reduction in EPS concentration. Filtration model must be adjusted to take into account the influence of floc size reduction, role of coagulant (metal ions e.g. Fe^{2+}) in inorganic fouling and reduction in the stickiness between the membrane and sludge particles due to reduced deposition of EPS onto membrane. Majority of the modeling studies are focused on pilot-scale units and lab-scale units and there is dire shortage of the full-scale MBR systems. It is therefore suggested to undertake full-scale integrated modeling studies in future taking into account the bio-chemical P-removal and fouling development as well as the influence of the precipitant addition on microbial communities.

IWA standard nomenclature for model parameters

$q_{XCB_SB,hyd}$	maximum specific hydrolysis rate
$n_{q_{hyd},An}$	correction factor for hydrolysis
$K_{NHx,OHO}$	half saturation coefficient for S_{NHx}
b_{OHO}	decay rate of X_{OHO}
q_{PAO,VFA_Stor}	rate constant for X_{PAO} storage
$q_{PAO,PO4_PP}$	rate constant for storage of X_{PAO_PP}
$K_{PP,PAO}$	half saturation coefficient for X_{PAO_PP}
$\mu_{PAO,Max}$	maximum growth rate of X_{PAO}
μ_{PAO,Max_lim}	maximum growth rate of X_{PAO} (when P is limiting)
$\eta_{\mu_{PAO}}$	reduction factor for anoxic growth rate of X_{PAO}
m_{PAO}/b_{PAO}	decay rate of X_{PAO}
$\mu_{ANO,Max}$	maximum growth rate of X_{ANO}
b_{ANO}	decay rate of X_{ANO}
$K_{O2,ANO}$	half saturation coefficient for S_{O2}
$K_{NHx,ANO}$	half saturation coefficient for S_{NHx}
$K_{O2,hyd}$	inhibition coefficient for S_{NHx}
$q_{SF_VFA,Max}$	Maximum specific fermentation growth rate
$\mu_{OHO,Max}$	Max. growth rate of X_{OHO}
$\eta_{\mu_{OHO,Ax}}$	reduction factor for anoxic growth rate
Y_{OHO}	Yield for X_{OHO} growth
$Y_{OHO,Ax}$	Yield for X_{OHO} growth (anoxic)
$Y_{Stor_OHO,Ax}$	Yield for X_{OHO} growth per X_{OHO_stor} (anoxic)
$f_{XU_Bio,lys}$	fraction of X_{UJ} generated in X_{OHO} decay
Y_{PAO}	Yield for X_{PAO} growth
$Y_{PP_Stor,PAO}$	Yield for $X_{PAO,PP}$ requirement
i_{N_XU}	N content of X_{UJ}
i_{N_XCB}	N content of X_{CB}
i_{P_XU}	P content X_{UJ}
i_{P_XBio}	P content of biomass
i_{P_XCB}	P content of X_{CB}

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Credit authorship contribution statement

Kashif NADEEM, Data collection, Data analysis and interpretation, Drafting the article, Final approval of the version to be published.

Marion ALLIET Conception or design of the work, Data analysis and interpretation, Drafting the article, Final approval of the version to be published.

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Claire ALBASI Conception or design of the work, Data analysis and interpretation, Critical revision of the article, Final approval of the version to be published.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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