Modelling anaerobic, aerobic and partial nitritation-anammox granular sludge reactors - A review

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ABSTRACT

Wastewater treatment processes with granular sludge are compact and are becoming increasingly popular. Interest has been accompanied by the development of mathematical models. This contribution simultaneously reviews available models in the scientific literature for anaerobic, aerobic and partial nitritation-anammox granular sludge reactors because they comprise common phenomena (e.g. liquid, gas and granule transport) and thus pose similar challenges. Many of the publications were found to have no clearly defined goal. The importance of a goal is stressed because it determines the appropriate model complexity and helps other potential users to find a suitable model in the vast amount of literature. Secondly, a wide variety was found in the model features. This review explains the chosen modelling assumptions based on the different reactor types and goals wherever possible, but some assumptions appeared to be habitual within fields of research, without clear reason. We therefore suggest further research to more clearly define the range of operational conditions and goals for which certain simplifying assumptions can be made, e.g. when intragranule solute transport can be lumped in apparent kinetics and when biofilm models are needed, which explicitly calculate substrate concentration gradients inside granules. Furthermore, research is needed to better mechanistically understand detachment, removal of influent particulate matter and changes in the mixing behaviour inside anaerobic systems, before these phenomena can be adequately incorporated in models. Finally, it is suggested to perform full-scale model validation studies for aerobic and anammox reactors. A spreadsheet in the supplementary information provides an overview of the features in the 167 reviewed models.

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1. Introduction

Granular sludge technology underpins key emerging and established technologies for wastewater treatment across industrial and municipal sectors. Pollutant removal in these reactors relies on microorganisms that grow in approximately spherical aggregates (biofilms) that can freely move within the reactor. Compared to flocs, granules have a higher biomass density and a more regular shape; they are mechanically stronger and they can become larger (Liu and Tay, 2004). These characteristics can lead to microscale substrate concentration gradients inside the aggregates, but even more importantly, they lead to high settling velocities (Winkler et al., 2018). Fast settling is the common benefit of all types of granular sludge. It facilitates solid-liquid separation and therefore allows high reactor biomass concentrations (Nicolella et al., 2000). In addition, granular sludge reactors provide a high biofilm specific surface area (Morgenroth, 2008) and the geometry and free movement of granules limits external boundary layer resistances, promoting mass transfer of substrate towards theorganisms. The combination of the high biomass concentration and fast mass transfer allows high removal rates, ultimately enabling compact installations (Heijnen et al., 1993).

Anaerobic granular sludge has been applied in upflow sludge blanket reactors since the 1970s (Lettингa et al., 1980) and later in variations of this technology, to remove organic pollution from wastewater through biological conversion into biogas. The potential to recover energy in compact installations with a low sludge production made anaerobic granular sludge technologies very popular (van Lier, 2008), especially for industrial wastewaters. Aerobic treatment processes using small spherical carriers for biofilm growth were developed in the 1970s for removal of organics and ammonium (Jeris et al., 1977), later combined with nitrate and nitrite removal under anoxic conditions, via denitrification (Nutt et al., 1984). Self-sustained granules for aerobic treatment only became feasible much later, with the development of a sequencing batch reactor technology, which also enables biological phosphorus removal (de Kreuk and van Loosdrecht, 2004). This has evolved into a mature technology with distinct benefits compared to conventional activated sludge systems. It requires 25–75% less space due high biomass concentrations and the absence of settling tanks and it has 20–50% lower energy demands due to the lack of recycle and sludge return pumps and mixers (Prонk et al., 2017). Moreover, the waste sludge offers potential for recovery of valuable biopolymers (Lin et al., 2015). The discovery of the anammox reaction (Mulder et al., 1995) stimulated the development of a third important type of granular sludge technology. One-stage partial nitritation-anammox is now frequently used to treat nitrogen-rich wastewaters (Lackner et al., 2014). This also offers potential for the recovery of phosphorus, because it can accumulate inside the granules (Johansson et al., 2017). Granular sludge is also promising for emerging biological treatment processes, such as phototrophic processes (Abouhend et al., 2016) and sulfide-based organics removal (Hao et al., 2013) and denitrification (Yang et al., 2016), but these are not yet available at commercial scale.

Modelling is a widely acknowledged tool for fundamental understanding, design and optimization of wastewater treatment processes (Van Loosdrecht et al., 2008). Reviews on wastewater treatment models have generally focussed on either anaerobic (Batstone et al., 2015; Liotta et al., 2015; Sadino-Riquelme et al., 2018; Tomei et al., 2009) or aerobic processes (Hauduc et al., 2013; Karpinska and Bridgeman, 2016; Liotta et al., 2014), because these require different redox conditions. Except for Nicolella et al. (2000), Liu and Tay (2004) and Milferstedt et al. (2017a), reviews focussing on granular sludge have discussed anaerobic (Chong et al., 2012; Saravanan and Sreekrishnan, 2006; Schmidt and Ahring, 1996) and aerobic processes (Bengtsson et al., 2018; Ni and Yu, 2010a; Winkler et al., 2018) separately as well. However, from a physical and chemical point of view, anaerobic, aerobic and partial nitritation-anammox granular sludge reactors share much in common, such as hydrolysis of particulate substrates, interactions between gas bubbles and the water phase, mass transfer of substrates from the bulk liquid to the granule surface and acid-base reactions. Also stable granule formation probably relies on the same interplay between mass transfer of solutes, conversion rates and detachment forces (van Loosdrecht et al., 2002). Therefore, modelling these different reactors poses largely the same challenges.

This review discusses models of anaerobic, aerobic and partial nitritation-anammox granular sludge reactors together to assess commonalities and discrepancies in approaches. First, the scope of the review is defined. Next, the different modelling goals of the studies are discussed. The two main sections describe differences in assumptions about transport phenomena and transformations and explain them based on the different reactor types and modelling goals wherever possible. As such, suggestions for further modelling studies are extracted and habitual assumptions and gaps in the knowledge are identified. Afterwards, the trends, advantages and disadvantages of model complexity are discussed. Next, the availability of calibration and validation studies for different reactor scales is discussed and finally, future research needs are summarized.
2. Scope and key phenomena

2.1. Scope of the review

This review focuses on mechanistic models for reactors with mixed microbial granules published in the English scientific literature. The review was limited to anaerobic, aerobic and partial nitritation-anammox (often referred to as simply 'anammox' further on) granular sludge reactors that are commercially available, as these are commonly applied and numerous modelling studies exist. Eight different reactor types with or without small carriers were identified: upflow sludge blanket, expanded granular sludge bed, internal circulation, baffled, fluidized bed, air-lift, sequencing batch and simple aerated reactors (Fig. 1). Mechanistic models are here understood as models that are based on mass balances with transport and reaction terms. Models that only describe batch tests were not considered, because the focus is on models for operational reactors as a whole, meaning they can predict the effluent concentration of at least one substrate (i.e. pollutant). In case more than one model for a single reactor type was described in a publication, only the model that the authors labelled as the most accurate or reference was analyzed.

With these selection criteria, this review covers 164 publications including 167 models (Table 1 provides an overview and the spreadsheet in supplementary information shows the model features). Omitting yearly fluctuations, there has been an increase in publication rate from the first publication in 1981 until 1997, followed by a brief decline and then another increase until it stagnated around 2006 (Fig. 2A). The first publications modelling aerobic systems described fluidized bed and air-lift reactors, but most models for aerobic systems were developed for sequencing batch reactors (Fig. 2B) because the focus shifted almost completely since 2001, preceding the commercialization of this technology (de Kreuk et al., 2007a). Anaerobic granular sludge models appeared a
few years after the invention of the technology (Lett inga et al., 1980). Models for upflow sludge blanket reactors are by far the most abundant in literature, followed by anaerobic fluidized bed reactors (Fig. 2B), but interest in the latter type has sharply declined in the last decade, as application of the technology also decreased (van Lier et al., 2016). In 2007, the first model for one-reactor partial nitritation-anammox granular sludge was published, quickly following the full-scale implementation of the technology (Wett, 2007).

### Table 1
Publications selected for analysis by process and reactor type.

<table>
<thead>
<tr>
<th>Anaerobic - Upflow sludge blanket</th>
<th>Anaerobic - Expanded granular sludge bed</th>
<th>Aerobic - Sequencing batch</th>
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<tbody>
<tr>
<td>Batstone et al. (2004)</td>
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<td>Chou and Huang (2005)</td>
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<td>Işık and Sponza (2005)</td>
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<td>Elmitwalli et al. (2006)</td>
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<td>Huang et al. (2006)</td>
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<td>Kalyuzhnyi et al. (2006)</td>
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<td>Soroa et al. (2006)</td>
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<td>Pontes and Pinto (2006)</td>
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<td>Angulo et al. (2007)</td>
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<td>Mu et al. (2007)</td>
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<td>Vlissides et al. (2007a)</td>
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<td>Vlissides et al. (2007b)</td>
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<td>Bhunia and Changrekar (2008)</td>
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<td>Mu et al. (2008a), Tartakovsky et al. (2008)</td>
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<td>Mu et al. (2008b)</td>
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<td>Narayananan and Narayan (2008)</td>
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<td>Sponza and Ulukoy (2008)</td>
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<td>Fuentes et al. (2009a)</td>
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<td>Lopez and Borzacconi (2009)</td>
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<td>Dereli et al. (2010)</td>
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<td>Diamantis and Aivasidis (2010)</td>
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<td>Rodriguez and Moreno (2010a)</td>
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<td>Zhao et al. (2010)</td>
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<td>Turkdogan-Aydinol et al. (2011)</td>
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<td>Coskun et al. (2012)</td>
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<td>Thamsiriroj et al. (2012)</td>
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<td>Willquist et al. (2012)</td>
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<td>Yerlimetzoy (2012)</td>
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<td>Yu et al. (2012)</td>
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<td>Chen et al. (2015)</td>
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<td>Haugen et al. (2015)</td>
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<td>Lohani et al. (2016)</td>
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<td>Rodriguez-Gomez and Renman (2016)</td>
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<td>Sun et al. (2016)</td>
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<td>Pokorna-Krayzelova et al. (2017)</td>
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### Key phenomena in granular sludge reactors

A number of key phenomena (based on Wanner et al. (2006)) that occur in granular sludge reactors can be included in models (Fig. 3).

- **Transformations.** Both biological conversions (e.g. nitrification) and purely physico-chemical reactions occur, such as precipitation and acid-base reactions. These are the main drivers for...
the removal of pollutants (e.g. ammonia, organics or phosphate) and production of sludge (biomass and precipitates).

- **Liquid phase transport.** Liquid flows through a reactor with a certain pattern, influencing the transport of solutes and colloidal matter through advection. The hydraulics determine whether the liquid phase is well-mixed or shows spatial gradients.

- **Granule transport.** Granules move as a result of gravity, drag and contact forces (e.g. collision with a separator). The balance of these forces regulates the retention of granules inside the system and the distribution of biomass along the reactor height. There is a mutual interaction between the movement of granules (solid phase) and liquid.

- **Intragranule transport.** Solute move through the granule matrix primarily via diffusion, but also advection can take place via pores. Together with the biological conversions and physico-chemical reactions, this determines the concentration gradients inside granules. Also matrix-embedded particles and microorganisms move, which influences the microbial population distribution inside granules.

- **Liquid-granule mass transfer.** Solute are exchanged across the external mass transfer boundary layer surrounding granules and are subsequently potentially adsorbed onto the biofilm matrix. These processes can influence the removal rate of pollutants from the wastewater. Also, exchange of particulate components through detachment and attachment occurs. This can affect the microbial population distribution inside granules and the granule size.

- **Granule transformations and size distribution.** Granules can grow, shrink and break up. Together with granule transport (e.g. wash-out of small granules), this determines the size distribution, which ranges from small flocs to millimetre thick granules (Pereboom, 1994; Pronk et al., 2015; Vlaeminck et al., 2010).

- **Gas phase transport.** The gas phase (air or biogas) mostly moves upwards due to buoyant forces, but can also flow downwards, e.g. in the downcomer of air-lift reactors. The gas transport mutually interacts with the liquid phase transport. The different gas-phase constituents, such as oxygen (O₂), carbon dioxide (CO₂), methane (CH₄), hydrogen sulfide (H₂S), nitrous oxide (N₂O), nitric oxide (NO) and nitrogen gas (N₂), can be well-mixed or show concentration gradients.

- **Liquid-gas mass transfer.** Components such as oxygen and methane experience a resistance when exchanged between the gas and the liquid phase due to stagnant layers on the inside and outside of a bubble. This resistance determines the mass transfer rate and therefore affects the distribution of compounds between the liquid and gas-phase.
• Heat transport and production. Heat enters and leaves via the liquid, through work done by mechanical equipment, and through heat exchange with the environment, while biological conversions and physico-chemical reactions can produce heat. These phenomena determine the temperature (distribution) in the reactor.

A spreadsheet in the supplementary information provides a complete overview of the assumptions that were made for these key phenomena in all the analyzed models. It also indicates the applications that were proposed or demonstrated, the scale of the largest modelled reactor, whether calibration and/or validation was performed and which software was used for modelling and simulation. This spreadsheet is available as a tool for practitioners and scientists to find an appropriate model to tackle their specific problem.

3. Modelling goal

Given the vast number of published models and the variation among them, it becomes ever more important to specifically state the model purpose in every publication. Still, one third of the analyzed models had no specific goal — or at least it was not clearly defined. The publications may state that the model is capable of characterisation, description, understanding, prediction, simulation, design, optimization or control of a system, but this is very general. It does not differentiate the model in comparison with other available models. For example, if a model is said to be suitable for design, it is not clear whether this means that it can determine the required reactor volume, the optimal height to width ratio or the required capacity of aerators. Therefore we did not consider such general applications or goals. This does not mean that the models are not useful, only that the applications are not clearly communicated.

Generally speaking, three different types of applications of granular sludge reactor models could be distinguished (Table 2). The first type is to gain fundamental insight in the relationship between micro- and mesoscale phenomena (e.g. intragranule transport) and macroscale reactor operation (e.g. influent characteristics) and performance (e.g. effluent quality). The second type is the assessment of alternative operational strategies or reactor/plant designs to have a better overall reactor performance. For this, micro- or mesoscale phenomena were sometimes included, but only the prediction of the macroscale reactor behaviour was of interest. Finally, the third application is to extract more information from measurements, e.g. about the microbial activity. Of course, this pragmatic classification of goals is not absolute, e.g. models that are primarily used for fundamental insight are often used also for design and optimization and some models that were purely used for design/optimization questions in a publication also have potential to gain more fundamental insight. Still, it gives a structure to the vast amount of publications.

4. Transport and mass transfer phenomena

This section discusses how transport of substrates and organisms inside the liquid, granule and gas phase or transport of these phases themselves has been included in models to calculate different kinds of spatial heterogeneity in a reactor. Fig. 4 illustrates the multi-scale nature of this spatial heterogeneity, which will be further elaborated in the subsections.

4.1. Liquid phase transport

Liquid phase transport patterns depend on the reactor geometry (Hu et al., 2017), inlet flow (Wang et al., 2009), inlet distribution (Asif et al., 1992) and gas production or injection (Buffiere et al., 1998). For anaerobic granular sludge reactors, the absence of aeration-induced mixing explains why spatial concentration gradients of soluble compounds are often considered (Fig. 5). This was done assuming an ideal plug flow (Bonnet et al., 1997; Wang Shi and Zhou, 1994), combinations of tanks (Bolle et al., 1986; Tanaka et al., 1981), the advection-dispersion equation (Kalyuzhnyi et al., 2006; Seifi and Fazaelipoor, 2012) or computational fluid dynamics (CFD) (Yang et al., 2015). The latter is the only technique that can predict the mixing behaviour instead of assuming it. Only some authors verified the hydraulic behaviour using tracer tests or measurements of concentration profiles in the specific modelled reactor (Huang et al., 2003; Lin and Yang, 1995). No studies experimentally verified complete mixing in baffled, fluidized bed or internal circulation reactors, even though it has been assumed. It seems contradictory that an ideal plug flow or non-ideal flow has been assumed slightly more often for large scale than for lab-scale anaerobic sludge blanket reactors (zoom on Fig. 5) because experiments have shown that mixing improves with the scale (Batstone et al., 2005). In expanded granular sludge bed reactors, both thorough mixing (Chou et al., 2008; Fuentes et al., 2011) and (semi-) plug flow behaviour has been observed (Yang et al., 2015; Zheng et al., 2012) on both large- and small scale.

Because the liquid phase transport can differ even between two anaerobic reactors of the same type, tracer tests or measurements of gradients are preferred whenever possible. If the reactor is still to be built, e.g. if a model is used for design, tracer tests from reactors with a similar scale, operation and construction can give an approximation, but also preliminary CFD calculations (with a simplified conversion model) could help find an appropriate assumption. For baffled reactors in particular, the experimental evidence and physical compartmentation encourages to always

Table 2

Different types of applications that can be distinguished for published granular sludge reactor models.

<table>
<thead>
<tr>
<th>Application</th>
<th>Description</th>
<th>Specific examples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Insight</td>
<td>Understand the relationship between small scale phenomena and large scale operation or performance</td>
<td>- Microbial competition for substrate (Kalyuzhnyi and Fedorovich, 1997)</td>
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<td>- Role of internal storage compounds (Beun et al., 2001)</td>
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<td>- Microbial interactions inside granules (Corbala-Robles et al., 2016)</td>
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<td>- Degree of mixing (Wu and Hickey, 1997)</td>
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<td>- Intragranule and external mass-transfer resistances (Huang et al., 2011)</td>
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<td></td>
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<td>- Bicarbonate addition (Batstone and Keller, 2003)</td>
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<td>- On-line temperature control (Angulo et al., 2007)</td>
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<td>- Selective retention of granules (Wett et al., 2010)</td>
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<td>- Required aeration capacity and recirculation flow (Tanaka et al., 1981)</td>
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<td></td>
<td>- Minimal required reactor volume (Toshani et al., 2016)</td>
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<td></td>
<td></td>
<td>- Monitoring microbial populations/activity (López and Borzacconi, 2009)</td>
</tr>
<tr>
<td>Design/optimization</td>
<td>Assess the effect of alternative operational strategies or design on the reactor/plant performance</td>
<td></td>
</tr>
<tr>
<td>Monitoring</td>
<td>Extract the value of unmeasured variables from measurements</td>
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</table>
assume a non-ideal flow (Bachmann et al., 1985; Li et al., 2016b). Finally, (semi-)plug flow behaviour is essential for specific applications, e.g. to assess the effect of an internal recirculation flow (Mu et al., 2008a) or short-circuiting (Bolle et al., 1986).

For aerobic and partial nitritation-anammox reactors, the choice of a flow pattern is often more straightforward because active air injection induces mixing. Only for fluidized bed reactors, tracer tests and measurements of concentration gradients have confirmed that complete mixing cannot always be assumed (Seifi and Fazaelipoor, 2012; Stevens et al., 1989), which is reflected in Fig. 5. For one-stage partial nitritation-anammox reactors, the assumption of a completely mixed liquid phase has not yet received experimental verification, even though it seems a reasonable assumption due to the active aeration. For aerobic sequencing batch reactors, it is known that semi-plug flow behaviour exists during the un aerated feeding phase, but this has not been considered in whole-reactor models yet (Weissbrodt et al., 2017).

4.2. Granules

4.2.1. Granule transport

No heterogeneous vertical distribution of biomass is considered in aerobic and anammox-based granular sludge reactors (except sometimes during a settling phase). This is justified by the air-induced mixing of the sludge bed. In contrast, a vertical biomass concentration profile is considered in 31% of the anaerobic granular
sludge models because it is also often experimentally observed (Lettin
ga et al., 1980; Wu and Huang, 1996). Sometimes the profile is predefined in the model based on measurements (Feldman et al., 2017) or an educated guess (Sam-soon et al., 1991), but often the profile is predicted more mechanistically through mass transport as a result of upward drag forces and gravity acting upon the sludge bed (Bolle et al., 1986; Bonnet et al., 1997). Only when plug flow characteristics are considered, do the simulated effluent concentrations become sensitive to the assumption of a vertical biomass distribution. An uneven distribution of biomass has no effect on the reactor performance if the liquid phase is completely mixed.

To avoid that the simulated biomass concentration becomes unrealistically high, models with biomass growth need to include washout from the reactor as well, apart from decay. Models for anaerobic reactors often (49%) consider imperfect retention of washout from the reactor as well, apart from decay. Models for aerobic and partial nitritation-anammox include biomass growth (using apparent kinetics). Biofilm models calculate the fixed solids separation efficiency (Bolle et al., 1986).

\[
\frac{dX_{\text{reactor},i}}{dt} = \frac{Q_iX_{\text{in},i}}{V_{\text{reactor}}} - \frac{X_{\text{reactor},i}}{\text{SRT}} + \sum_i \mathbf{r}_i
\]

(1)

where \(X_{\text{reactor},i}\) is the concentration of a microbial group \(i\) or other particulate variable in the reactor, \(X_{\text{in},i}\) is its concentration in the influent (g.m\(^{-3}\)), \(Q_i\) the influent flow rate (m\(^{-3}\).d\(^{-1}\)), \(V_{\text{reactor}}\) the reactor volume (m\(^3\)), SRT the solids retention time (d), and \(r_i\) the conversion rates of the group, e.g. of decay and growth (g.m\(^{-3}\).d\(^{-1}\)). Sometimes the degree of retention is incorporated more mechanistically via the settling behaviour of granules and advective upflow (Kalyuznyhi et al., 2006; Saravanan and Sreekrishnan, 2008), as described above for granule transport inside the reactor.

Only a few models for aerobic and partial nitritation-anammox reactors estimate the washout of granules via a fixed separation efficiency (Wett et al., 2010) or mechanistic settling behaviour (Kagawa et al., 2015; Su et al., 2013). In fact, most aerobic (87%) and anammox (80%) models that consider biomass growth, assume that granules are perfectly retained (Beun et al., 2001; Stevens et al., 1989; Tang et al., 1987; Volcke et al., 2010). This means that no granules are lost via the effluent, but loss of biomass through detachment followed by wash-out of suspended cells or by intentional sludge removal can still be considered. The two approaches can also be combined by defining a perfectly retained granule fraction and imperfectly suspended biomass fraction (Corbala-Robles et al., 2016; Hubaux et al., 2015; Pons et al., 2008).

It is not immediately clear why imperfect granule retention is so common for anaerobic systems, while perfect retention is often used for aerobic and anammox systems (Fig. 6). It could be partly habitual within each of these scientific domains. Possibly, authors interested in modelling anaerobic systems were inspired by previous models of such systems and those about to model aerobic and anammox systems based their assumptions on existing models of these systems respectively. As such, a habit can arise. A few models did not consider any retention of biomass compared to the liquid phase (i.e., \(\text{HRT} = \text{SRT}\)) (Ghobadian et al., 2014a, b; Setiadi et al., 1996, Shi et al., 2016). This last approach misses one of the key characteristics of granular sludge reactors, namely that granules settle fast and are therefore easily retained, reach high concentrations and as such allow high volumetric conversion rates.

4.2.2. Intragainule transport

Models of granular sludge reactors can be divided into two main categories: those that consider the intragainule transport (biofilm models) and those that treat biomass as a suspension in the liquid phase (using apparent kinetics). Biofilm models calculate the macroscale conversion rate by solving the mass-balance of substrates inside granules. The intragainule substrate concentration profile and related flux are derived considering simultaneous conversion and diffusion (Wanner et al., 2006), e.g. Eq. (2) assumes Monod kinetics and 1D radial transport.

\[
\text{Biofilm model:} \quad \frac{\partial C_i(z, t)}{\partial t} = \frac{D_i}{z^2} \frac{\partial^2 C_i(z, t)}{\partial z^2} + \frac{2}{z} \frac{\partial C_i(z, t)}{\partial z}
\]

\[
+ r_{i,\text{max}} \frac{X_i(z, t)}{C_i(z, t) + K}
\]

(2)

where \(C_i\) is the local substrate concentration (g.m\(^{-3}\)) at a certain distance from the granule core \(z\) (m) at time \(t\) (d), \(D_i\) the substrate diffusivity, \(r_{i,\text{max}}\) the maximum specific substrate uptake rate (g.m\(^{-3}\).d\(^{-1}\)), \(X_i\) the local biomass concentration (g.m\(^{-3}\)) and \(K\) the intrinsic half-saturation coefficient (g.m\(^{-3}\)). Appropriate boundary conditions are applied, normally spherical symmetry and a bulk concentration. Biomass concentrations inside the granules and granule geometry (size) may be likewise included in biofilm models as state variables (see below).

Mass transport between the bulk liquid and the microorganisms inside the biofilm decreases macroscale conversion rates compared to suspended systems with the same amount of biomass. This effect becomes more pronounced with a thicker biofilm, faster local conversion rates (through a higher local biomass concentration and/or specific uptake rate) or lower substrate diffusivity (Wang Shi and Zhou, 1994). This may be considered fundamentally, through biofilm modelling, as outlined above. However, when granular sludge is treated as a suspension, intragranule transport is not explicitly simulated, so this effect on the macroscale rates should be represented through modified kinetic parameters. ‘Apparent’ kinetics are thus obtained, for example by increasing half-saturation coefficients (Eq. (3)) to incorporate the diffusional resistance (Beccari et al., 1992; Manser et al., 2005; Pérez et al., 2005).

\[
\text{Apparent kinetics } r_{i,\text{apparent}}(t) = r_{i,\text{max}} \frac{C_i(t)X(t)}{C_i(t) + K_{\text{app}}}
\]

(3)

where \(r_{i,\text{apparent}}\) is the macroscale substrate uptake rate (g.m\(^{-3}\).d\(^{-1}\)) at time \(t\) (d), \(C_i\) the bulk substrate concentration (g.m\(^{-3}\)) and \(K_{\text{app}}\) the apparent half-saturation coefficient (g.m\(^{-3}\)). Changes in the granule size, microbial population distribution and competition between different microbial groups for the same substrates, e.g.
competition for oxygen between nitrifiers and heterotrophs, can alter the apparent kinetics, which may require (re)calibration (Baeten et al., 2018).

Only a minority explicitly modelled intragranule transport of substrates (using a biofilm model) for anaerobic granular sludge (21% of the analyzed models), even though intragranule transport has been shown to significantly influence mass transfer rates for at least sludge blanket (Wu and Hickey, 1997), expanded granular sludge bed (Chou et al., 2008) and fluidized bed reactors (Seok and Komisar, 2003b). This means that the use of apparent kinetics is widespread for anaerobic granular sludge (77% of the reviewed models). In contrast, more than three quarters of the aerobic and anammox granular sludge models explicitly describe intragranule solute transport.

It is not immediately clear why biofilm models are more common for aerobic and anammox systems, while apparent kinetics are mostly used for anaerobic ones (Fig. 6). ADM1 might be more complex than most aerobic or anammox conversion models, but it is still reasonably technically straightforward to apply it in a biofilm model with dedicated software like Aquasim (Reichert, 1994). The difference in popularity can also not be solely explained by an increased availability of tools or computing power, since many models for anaerobic systems with apparent kinetics were published in the same period as biofilm models for aerobic systems (Fig. S1). A possible explanation is that publications on aerobic (Beun et al., 2001; de Kreuk et al., 2007a; Kagawa et al., 2015; Winkler et al., 2015b) and anaerobic systems (Castro-Barros et al., 2018; Corbala-Robles et al., 2016; Mozumder et al., 2014; Volcke et al., 2010) often looked into intragranule substrate profiles and ecological interactions between different microorganisms (insight), while those dealing with anaerobic systems generally focussed more on overall reactor design and optimization (Batstone and Keller, 2003; Kleerebezem, 2003; Lohani et al., 2016; Mu et al., 2007; Yetilmezsoy, 2012) or were used for monitoring biological activity (Lopez and Borzacconi, 2011; Perez et al., 2001).

Not only solutes, but also matrix-embedded particles and microorganisms can move throughout a granule, which influences the microbial population distribution along the granule depth. Therefore, this distribution is dependent on the reactor operating conditions, even for the same type of reactor (Batstone et al., 2004). Such dynamics can be described mechanistically with an intragranule mass balance for biomass as state variables, including re-action and transport terms, analogous to Eq. (3). This time, diffusive transport is often neglected, but advective transport is required because a net growth of microorganisms causes biofilm geometry to move, i.e., deeper layers push outer layers outwards. The advective velocity of particles and microorganisms towards the granule surface is often calculated in 1D with Eq. (4), as this is used in Aquasim (Wanner and Reichert, 1996).

\[
\text{Advective velocity } u_{\text{advection}}(t, z) = \frac{1}{A(z)} \int_0^z \frac{1}{1 - e_1} \sum_{i=1}^{n} r_x(t, z) A(z) dz
\]

(4)

were \(u_{\text{advection}}\) is the advective velocity of the biofilm incl. microorganisms (m.d\(^{-1}\)) at a certain distance from the granule core \(z\) (m) at time \(t\) (d), \(A(z)\) the area of a sphere with radius \(z\) (m), \(e_1\) the porosity of the biofilm (\(\cdot\)), \(r_x\) the local production rate of particulate component (microbial group) (g.m\(^{-1}\).d\(^{-1}\)) and \(p_{x_i}\) the density of that component (g.m\(^{-3}\)). Individual-based models have also been used to consider these dynamics in granules (Kagawa et al., 2015; Xavier et al., 2007), which considers microorganisms as discrete entities instead of a continuum. This approach is especially relevant to study the 2D (or 3D) heterogeneity of granules (Fig. 4C), but comes at a high computational cost (de Kreuk et al., 2007a).

A dynamic microbial population distribution has been predicted mechanistically in all the biofilm models for anammox reactors, but some of the biofilm models for aerobic and anaerobic systems used a predefined distribution, either heterogeneous (Huang et al., 2006; Saravanan and Sreekrishnan, 2008; Tang et al., 1987; Wu and Hickey, 1997) or homogeneous (Chou et al., 2011a; Huang et al., 2011). Huang et al. (2006) claimed that 1D biomass spatial heterogeneity is necessary to effectively simulate the performance of an upflow sludge blanket reactor. However, the fact that 28 of the analyzed publications used a homogeneous distribution (Bachmann et al., 1985; Rodriguez-Gomez et al., 2014) or even neglected all intragranule transport processes indicates that these effects are often lumped in apparent kinetic parameters. Moreover, it has been shown that dense micro-colonies of nitrifiers, which can only be described in 2D or 3D, can influence the macroscale kinetics (Picioreanu et al., 2016). This shows that even kinetic parameters that are calibrated for 1D biofilm models might cluster some processes when dense micro-colonies are present.

4.2.3. Liquid-granule transfer

Adsorption of solutes on the biofilm matrix is hardly ever considered (Kennedy et al., 2001; Sam-soon et al., 1991; Tsuneda et al., 2002), even though organic compounds can adsorb onto anaerobic granules (Ning et al., 1997) and ammonium can adsorb on anammox (Li et al., 2016c) and aerobic granules. The latter is probably caused by the attraction between the negatively charged granule matrix and positively charged ammonium ions (Bassin et al., 2011). These adsorption processes could play a role in the removal of these pollutants. The compounds are transferred from the liquid phase to the solid phase, decreasing their discharge with the effluent and enabling removal with the waste sludge. At the same time, adsorption may reduce the availability for biological conversions. Desorption can occur when bulk liquid concentrations decrease again (Bassin et al., 2011).

Solutes need to pass through an external boundary layer prior to reaching the granule surface. Its negative effect on the overall conversion rates becomes stronger when the local uptake rate inside the biofilm or boundary layer thickness increases and when the limiting substrate concentration or diffusivity decreases (Picioreanu, 2015). Wu and Hickey (1997), Chou et al. (2008) and Wu and Huang (1995) estimated that the external resistance was not rate-limiting in the upflow sludge blanket, expanded granular sludge bed and anaerobic fluidized bed reactors they respectively studied. Stevens et al. (1989) also found that it was not rate-limiting in an aerobic fluidized bed reactor, whereas Tang et al. (1987) judged it to be important for an airlift reactor. Vangsgaard et al. (2012) found that it can sometimes be influential for anammox-based systems. So even though the extra turbulence created by aeration in anammox-based and aerobic systems can decrease the boundary layer thickness, other influencing factors can make its effect significant, especially a rapid oxygen uptake rate and relatively low bulk oxygen concentrations. Understandably, an external mass transfer resistance is considered most often in aerobic granular sludge models (31%). It is advisable for future modelling to make a rough estimation of the effect for aerobic and anammox-based reactors under the specific operating conditions, as in Stevens et al. (1989), if quantitative predictions are aimed at. Alternatively, the effect can be clustered in apparent kinetic parameters if no significant changes in the turbulence or other influencing factors are expected.

Also solids and microorganisms undergo liquid-granule transfer through attachment onto the granule surface and detachment from the surface. Detachment has a strong impact on biofilm thickness in the long-term (Wanner et al., 2006). Consequently, steady-state
simulations with biofilm models that included intragranule transport of microorganisms always considered detachment to avoid predicting unrealistically large granule sizes, except for Kagawa et al. (2015), who focused on the start-up of a reactor. The detachment rate is often defined with an equation to obtain a specific predefined steady-state granule size, for example Eq. (5) was used by Volcke et al. (2010).

\[
\text{Detachment velocity: } u_{\text{detachment}} = \left( \frac{\delta}{\delta_{\text{steady-state}}} \right) u_{\text{advection}} (\delta) \tag{5}
\]

where \( u_{\text{detachment}} \) represents the detachment velocity \( (\text{m.d}^{-1}) \), \( \delta \) the simulated granule radius, \( \delta_{\text{steady-state}} \) the predefined steady-state radius which is either measured or based on experience with similar systems \( (\text{m}) \) and \( u_{\text{advection}} \) is the advective velocity with which the granule expands due to biomass growth \( (\text{Eq. (4)}) \). This approach allows a dynamic microbial population distribution while keeping the granule size below a realistic limit, but it cannot actually predict the granule size. Also the convergence rate to the steady-state size relies on the arbitrary exponent 10. A realistic convergence rate can only be obtained after calibration of such empirical parameters \( (Ni et al., 2010) \). Finally, only a few publications considered attachment. Batstone et al. (2004) showed that modelling attachment was necessary to explain the observed acidogenic layer in anaerobic granules treating brewery wastewater.

4.2.4. Granule transformations and size distribution

The majority of granular sludge models (82%) assume a single granule size with a predefined steady-state value, or implicitly assume a constant granule size by using constant apparent kinetics. Sixteen models predict the size dynamically without a predefined steady-state value, but this has never been done for partial nitritation-anammox based reactors. Some predicted the granule size without using detachment, e.g. Rodriguez-Gomez et al. (2014), meaning that the granule size is only limited by decay. The accuracy of the predicted size is doubtful for long-term simulations in these cases. Since detachment is driven by liquid shear and granule shear strength, its rate is a function of the operating conditions. For example, Odriozola et al. (2016), Su et al. (2013) and Fuentes et al. (2008a) applied an empirical equation linking the detachment rate to the biogas production rate and liquid upflow velocity or aeration rate. The generalizability of such equations is uncertain, as they contain an empirical parameter that might require calibration for different reactor operating conditions, which again makes it non-predictive. Detachment rates can in principle be determined mechanistically from biofilm strength and liquid shear rate \( (Horn and Lackner, 2014; van Loosdrecht et al., 2002) \). It might also be possible to estimate the granule shear strength based on influent wastewater properties \( (Batstone and Keller, 2001) \). Yet, integrating these aspects to fundamentally dynamically predict the granule size has not yet been done, even though it is technically possible. Note that a dynamic granule size only affects the predicted macroscale conversion rates directly if intragranule solute transport is included. Otherwise, only indirect effects are considered, such as the better settleability of bigger granules and accompanying higher biomass concentration, e.g. in Kalyuzhnyi et al. (2006) and Fuentes et al. (2009a).

Some models consider the size distribution of granules inside a reactor. The simplest approach is to distinguish two classes, granules and suspended sludge \( (Fuentes et al., 2008c; Hubaux et al., 2015; Pons et al., 2008) \), but more size classes have also been used \( (Feldman et al., 2017; Huang et al., 2003; Su and Yu, 2006a; Wu and Huang, 1996) \). Volcke et al. (2012) showed that the use of a single, average granule size is sufficient to predict the effluent nitrogen concentration and specification from a partial nitritation-anammox reactor. Odriozola et al. (2016) also found that the predicted methane production and effluent soluble substrate concentrations were similar with and without considering the granule size distribution in an anaerobic system. This means that a size distribution is only necessary for fundamental understanding, e.g. to get insight in the solute exchange between different size classes.

To get insight in the formation of a granular sludge bed, a size distribution as well as the dynamics of each size class need to be considered. This has been done in three publications \( (Odriozola et al., 2016; Seok and Komisar, 2003a; Su et al., 2013) \). Su et al. (2013) provides the most comprehensive approach, using a population balance model for an aerobic granular sludge reactor. Apart from the mass balances of particulate and soluble compounds, such models use balances on the number of particles in each size class, by estimating wash-out, growth, break-up into smaller pieces etc. As such, the selective retention of larger granules can be simulated, which is seen as one of the factors promoting granulation of sludge \( (Beun, 1999) \). Because this approach still relies on calibrated relationships for the detachment rate, as discussed above, and a calibrated granule breakage probability, it is difficult to quantitatively predict the granule size distribution before a reactor is operational, e.g. for design purposes. This might even be impossible in a quantitative, deterministic way, given the complex mutual interaction between the shear stress and granule shear strength and stochastic processes, like the influent composition, temperature, microbial species (and related kinetics) and breakage events.

4.3. Gas phase transport and liquid-gas transfer

More than half of the selected models (56%) assume an instant equilibrium between the gas and liquid phase concentrations, or do not consider transfer of gases at all. For example, the liquid-gas transfer resistance of methane in anaerobic systems is often neglected, probably because it is the final product of the conversions and will therefore not directly influence the predicted conversion rates. Nonetheless, if the biogas composition, flow or induced mixing is of interest, liquid-gas transfer is important \( (Pauss et al., 1990) \). Gas entrapment onto or inside granules can also cause sludge to rise in anaerobic \( (Bolle et al., 1986) \) and anammox-based reactors \( (Van Hulle et al., 2010) \). This effect has never been included explicitly, but it has been incorporated through calibrated correction parameters to predict the vertical biomass concentration distribution in a reactor as well as wash-out \( (Bolle et al., 1986; Kalyuzhnyi et al., 2006) \). When modelling aerobic and anammox-based reactors, the influence of the liquid-phase oxygen concentration has often been studied directly, so the gas-phase is actually not of interest. Therefore resistances in the liquid-gas boundary layer for oxygen are neglected by choosing an unrealistically high transfer rate \( (Beun et al., 2001; Volcke et al., 2010) \). Yet, a realistic oxygen transfer resistance would be required to quantify the necessary airflow rate to obtain a certain liquid-phase oxygen concentration \( (Garcia-Ochoa and Gomez, 2009) \). The mass transfer dynamics are also crucial to compare different online control strategies for aeration, e.g. under variations in load, a well-known application of activated sludge models \( (Amand and Carlsson, 2012; Belchior et al., 2012) \). The delay between an increase in the aeration rate and the resulting increase in the dissolved oxygen concentration due to the mass transfer resistance can be important along with other dynamics, like the sensor response times \( (Alex et al., 2008) \).

When a liquid-gas mass transfer resistance is used for gases, an assumption about the transport phenomena within the gas-phase
needs to be made. Mostly, a completely mixed gas-phase is used, but some publications consider that vertical concentration gradients can exist within the gas-phase. For now, the only clear benefit of this model feature is that concentration gradients inside the gas-phase can be estimated (WiseCarver and Fan, 1989), but there might be some indirect effects on the predicted effluent quality, aeration requirements or biogas quality which have not yet been investigated explicitly.

5. Transformations

5.1. Biological conversions

Biological conversions occurring in anaerobic, aerobic and partial nitritation-anammox granular sludge reactors show overlap because some processes are not specific for certain redox conditions (e.g. hydrolysis) and because anaerobic and anoxic conditions can exist in aerobic and anammox-based reactors due to spatial and temporal heterogeneity (Fig. 7). Most models include biological conversions of substrate as a primary function. These can include different types of organics, ammonium, nitrate and nitrite, phosphorus and sulfate, under anaerobic, anoxic and aerobic conditions. Models of anaerobic granular sludge reactors all include the conversion of organics to methane because this is the primary goal. A varying degree of detail has been used regarding the actual pathways. Conversions of sulfate are generally not included (Fedorovich et al., 2003; Pokorna-Krayzelova et al., 2017; Sun et al., 2016), but are important for sulfate containing wastewaters, since sulfate is readily reduced to sulfide, causing odour, corrosion and safety issues. The sulfide concentration in biogas increases almost linearly with the influent sulfate to organics ratio, at low sulfate concentrations (Batstone, 2006; Pokorna-Krayzelova et al., 2017). High sulfate concentrations (≥0.125 g S.g COD⁻¹) also significantly reduce methane production due to inhibition of conversions by sulfide (Vavilin et al., 1995) and because sulfate reducing bacteria compete with methanogens for organic substrates.

For aerobic systems, most models consider organics and ammonium conversions, sometimes combined with denitrification of nitrite/nitrate, depending on the wastewater composition and presence of anoxic conditions. Only four out of 22 models for aerobic sequencing batch reactors include biological phosphorus removal, because they were often developed for lab-scale reactors with short feeding phases. However, in full-scale systems (Nerda³), slow anaerobic feeding is applied to promote phosphate accumulating organisms and achieve more stable granules (de Kreuk and van Loosdrecht, 2004; Pronk et al., 2015). This constitutes a substantial discrepancy between available models and practice. Furthermore, the models that did consider phosphorus conversions in a reactor with slow anaerobic feeding required significant alterations of the kinetic parameters of phosphate accumulating organisms in order to match experimental results (de Kreuk et al., 2007b; Winkler et al., 2015b; Xavier et al., 2007), which might be due to an inappropriate description of their metabolism (Barnard et al., 2017).

In anammox-based systems, ammonium and nitrate/nitrite conversions via nitritation and anammox are always considered, but conversions of organics are sometimes neglected because these are generally not the main target. Nevertheless, about 300–1400 mg COD⁻¹ is removed in full-scale reactors (Lackner et al., 2014) and these conversions influence the total nitrogen removal (Mozumbic et al., 2014), so it is always recommended to include denitrification and aerobic oxidation of organics.

Particulate organics were considered as separate state variable(s) in thirty models for anaerobic granular sludge reactors, but this was rarely done for aerobic or anammox-based systems. This lack of particulates conversions in the latter two probably originates from the more complex hydrolysis kinetics that are usually assumed for aerobic (Henze et al., 2000) compared to anaerobic sludge (Batstone et al., 2002). Activated sludge models (ASMs, Henze et al. (2000)) use a hydrolysis rate dependent on the concentration of heterotrophic organisms (Eq. (6)).

\[
\text{ASM : } F_{\text{particulate}} = k_{\text{particulate}} X_{\text{particulate}} X_{\text{heterotrophs}}
\]

where \( F_{\text{particulate}} \) is the removal rate of particulates (g.m⁻³.d⁻¹), \( k \) is a rate coefficient (d⁻¹), \( M_i \) are Monod expressions (–), \( K_i \) is a half-saturation coefficient (–), \( X_{\text{particulate}} \) the concentration of particulate organics (g.m⁻³) and \( X_{\text{heterotrophs}} \) the concentration of heterotrophs (g.m⁻³). Such kinetics are not easily compatible with biofilm models, which are often used for aerobic and anammox granular sludge reactors. The reason is that biofilm models assume that (most of the) heterotrophic organisms reside inside the granules. On the other hand, particulates enter via the bulk liquid and thus they do not come into contact with heterotrophs. As such, the predicted rate (Eq. (6)) is zero in the granules because \( X_{\text{particulate}} \) is zero and the rate is small (or zero) in the bulk liquid because \( X_{\text{heterotrophs}} \) is small (or zero). Removal would thus only be predicted if liquid-granule transfer, attachment and/or intragranule transport of particulates are defined to bring this substrate in contact with heterotrophs, but the rates of these transport processes are rarely analyzed and the exact mechanisms are poorly understood (Boltz et al., 2010; Pronk et al., 2015). It is unclear how some biofilm models predicted removal of influent particulates without defining any transfer of particulates to aerobic granules (Ni et al., 2009, Ni et al., 2008, Su and Yu, 2006a, b), Lübken et al. (2005) and Pons et al. (2008) could apply ASM-type kinetics because they assumed a suspension of biomass (no biofilm modelling), meaning that both the particulate organics and heterotrophs reside in the bulk liquid. Simpler kinetics could be used in future biofilm models for aerobic and anammox granules (Eq. (7)), as often applied for anaerobic granular sludge systems (Batstone et al. (2004) and Mu et al. (2008a)). This more empirical approach predicts conversion, irrespective of the contact between the substrate and heterotrophs.

\[
\text{ADM } r_{\text{particulate}} = k_X r_{\text{particulate}}
\]

Biological conversions of substrates are generally linked to biomass growth. Biological wastewater treatment is an autocatalytic process in the sense that biomass is the catalyst for the degradation of substrates and at the same time more biomass is created during these conversions. Most models consider this explicitly by using one or more microbial groups, e.g. heterotrophs and nitrifiers, as state variables. The total amount or microbial composition of the biomass is then calculated via mass balances which include a growth term. Nevertheless, about one third of the models for anaerobic and aerobic reactors do not include microbial growth. They assume a fixed amount of biomass for every microbial group, or simply use a fixed maximal conversion rate. When these models are used to design the required reactor volume, the expected biomass concentration or maximal conversion rate must be assumed. For an already operational reactor, these parameters can be measured directly and used as parameters. Yet, without growth, changes in the biomass or conversion capacity cannot be predicted. Therefore, these models are less suitable to simulate long-term changes in operating conditions or to design reactors for a significantly different wastewater composition, concentration or loading rate.
5.2. Physico-chemical reactions

The reactor pH is affected by acid/base consumption and production during biological conversions and can in turn influence biological conversion rates. The pH can be calculated by solving implicit algebraic acid-base equations (Costello et al., 1991a), by solving differential equations with reaction rates (Batstone et al., 2004) or through reformulation to an explicit expression for the hydrogen ion concentration ($pH = -\log [H^+]$) in simple cases (Alvarez et al., 1992). The former two can be complicated when intragranule transfer (Batstone et al., 2004) or a non-ideal liquid phase (Kalyuzhnyi and Fedorovich, 1997) is considered because the equations need to be solved in each grid point. pH prediction is particularly important in anaerobic systems fed with poorly buffered industrial wastewater, which is often the case. These are prone to failure due to acid type overload. Also in other cases, pH can be critical to determine the biogas quality in terms of carbon dioxide and hydrogen sulfide concentrations and when ammonia inhibition is likely to take place. Accordingly, pH calculations are included in a fairly high proportion (35%) of the anaerobic granular sludge models, despite the technical challenges.

For aerobic and anammox granular sludge reactors, pH calculations are only included in models for one aerobic (Stevens et al., 1989) and two anammox (Jones et al., 2007; Wett et al., 2010) reactors, which means that a constant pH is assumed in most cases. In aerobic treatment, problems with nitrification are expected for low alkalinity or nitrogen-rich wastewaters (Sötemann et al., 2005) and about half of the full-scale anammox reactors have been reported to have experienced negative effects due to pH fluctuations (Lackner et al., 2014). The large difference in popularity of pH calculations between anaerobic and aerobic/anammox reactors might not be purely based on commonly encountered issues. It seems that the broad application of pH prediction for anaerobic digestion (non-granular sludge) in literature, including ADM1 (Batstone et al., 2002), has been adopted for granular sludge systems. On the other hand, ASMs do not include extensive pH predictions and this might have influenced aerobic and anammox granular sludge reactor modelling. Only alkalinity is used as a state variable to identify when pH inhibitions can occur in ASM2 and ASM3 (Henze et al., 2000).

Precipitation reactions occur when the solubility of mineral components in the bulk liquid or granules is exceeded. These are complex to model, as acid-base reactions are generally a prerequisite to determine supersaturation and the chemistry involved is not always completely understood (Wiltfert et al., 2015). Precipitation reactions can occur for all three wastewater treatment processes. Common precipitants are calcium and magnesium phosphates and carbonates, including struvite MgNH₄PO₄·6H₂O.
For example, in anammox (Johansson et al., 2017) and aerobic systems (Lin et al., 2012), phosphorus precipitates have been encountered (Huang et al., 2015). This can both contribute to phosphorus removal and open up possibilities for recovery of this nutrient, but it is also associated with reduced reactor performance (Li et al., 2011). Phosphorus precipitation can also be actively induced by addition of metal salts inside aerobic granular sludge reactors to supplement biological phosphorus removal (Prönk et al., 2015). Excessive formation of calcium precipitates in calcium-rich industrial wastewaters can cause cementation of anaerobic granular sludge beds (Batstone and Keller, 2003). Iron sulfide precipitation on the other hand, can decrease the sulfide concentration and thus improve the biogas quality (Wei et al., 2018). Despite the possible influence of precipitation reactions on the process performance, they are only included in eight granular sludge models (Batstone and Keller, 2003; Feldman et al., 2017). Feldman et al. (2017) provided the most rigorous approach with the largest amount of possible precipitation reactions that can occur in the bulk liquid and even inside granules. Their work showed that intragranule precipitation may reduce conversion rates due to physical displacement of active biomass from the granules. Similar phenomena might occur when phosphorus precipitates inside aerobic or anammox granular sludge, but this has not been studied yet.

6. Model complexity

The literature survey (spreadsheet in supplementary information) shows that there is not one generally accepted way to model granular sludge reactors. To get an overall idea of the complexity of the models, every model feature was assigned a certain score, qualitatively representing its complexity, and these were summed up to a complexity index of the model as a whole (defined by Table 3). The complexity of models appears to diverge over time (Fig. 8). More complex models become available in literature (Feldman et al., 2017; Fuentes et al., 2009c; Kagawa et al., 2015), but simple models are still being used. The benefits of simple models are the more straightforward interpretation, easier calibration and lower computational demand. However, complex models describe more processes and thus provide more detailed predictions, such as intragranule substrate gradients. It is also sometimes believed that more complex models lead to more accurate predictions of the overall reactor performance over a broader range of operational conditions. Yet, as Wanner and Gujer (1986) state: “A model should be as simple as possible, and only as complex as needed”. This means that the burden of proof for a higher predictive accuracy always lies with those that develop more complex models. In other words, a more complex model should only be used if a simpler model failed for the intended application, e.g. a completely mixed reactor has failed to predict the effect of an internal recirculation (Mu et al., 2008a) and a biofilm model without attachment has sometimes failed to predict the microbial population distribution qualitatively (Batstone et al., 2004). Moreover, it is difficult to validate some sub-models, like intragranule transport, for realistic conditions. For example, micro-sensors (e.g. hydrogen gas, pH or oxygen) can only be used for granules after harvesting them from the reactor and positioning them in specialized laboratory equipment. The gradients also differ for different sites on a granule and between different granules (van Loosdrecht et al., 1995; Winkler et al., 2011). This makes it difficult in practice to know whether the quantitative accuracy of these microscale models is high enough to improve the quantitative predictions of the macroscale

### Table 3

Definition of the complexity index. For every phenomenon (column 1), certain model features (column 2) were assigned a complexity score that was added to the value of the complexity index.

<table>
<thead>
<tr>
<th>Phenomenon</th>
<th>Model feature</th>
<th>Complexity score</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transformations</td>
<td>Per acid base reaction considered to predict the pH, biological conversion or precipitation reaction, biomass growth</td>
<td>+0.1</td>
</tr>
<tr>
<td>Liquid phase transport</td>
<td>Non-ideal flow using a combination of tanks, non-ideal flow using advection-dispersion or ideal plug flow, non-ideal flow using CFD</td>
<td>+0.5, +1, +2</td>
</tr>
<tr>
<td>Gas phase transport</td>
<td>Non-ideal flow or ideal plug flow</td>
<td>+1</td>
</tr>
<tr>
<td>Granule transport</td>
<td>Fixed vertical heterogeneous biomass distribution, vertical biomass distribution mechanistically predicted, fixed imperfect granule retention</td>
<td>+0.5, +1</td>
</tr>
<tr>
<td>Liquid-granule transfer</td>
<td>External mass-transfer resistance or adsorption of solutes, attachment, detachment</td>
<td>+0.5, +0.5</td>
</tr>
<tr>
<td>Liquid-gas transfer</td>
<td>Per component with a liquid-gas transfer resistance</td>
<td>+0.1</td>
</tr>
<tr>
<td>Intragranule transport</td>
<td>Per dimension considered for solute transport, fixed heterogeneous microbial population distribution, microbial population distribution mechanistically predicted</td>
<td>+1, +0.5</td>
</tr>
<tr>
<td>Granule transformations and size distribution</td>
<td>Two size classes (suspended and granular biomass), more than two size classes, dynamic granule size without fixed steady-state value</td>
<td>+0.5, +1</td>
</tr>
<tr>
<td>Heat transport</td>
<td>Temperature mechanistically predicted</td>
<td>+1</td>
</tr>
</tbody>
</table>
reactor performance.

A mechanistic model could be very strictly defined as one that takes into account every known phenomenon, which would lead to a very high complexity. Our survey found that none of the models do this. Thus, a more pragmatic definition was used, namely a model based on mass balances with transport and reaction terms. One should acknowledge that every single wastewater treatment model has parameters that cluster several processes, because these were not described explicitly. For example, none of the selected models include the many different species within every microbial group (Vannecke and Volcke, 2015). As a second, but definitely not conclusive example, the 3D architecture of granules is far more complex than ever considered: 3D micro-colonies exist inside granules (Picoreanu et al., 2016), granules are not perfect spheres (de Kreuk and van Loosdrecht, 2004) and Herrling et al. (2017) stated that even the diffusivity inside granules is spatially heterogeneous due to variations in the density of the matrix. The inclusion of these aspects could lead to more fundamental understanding, e.g. about 3D substrate gradients and microbial competition, but it is not per se necessary for optimization or design purposes. In practice, the estimation of parameters via experiments is always necessary for quantitative simulations to compensate for neglected phenomena. Problems arise when simulation results are interpreted quantitatively when using parameter values for a complex model that were calibrated for a simpler model or the other way around, or when a simulation scenario differs strongly from the conditions during calibration or validation. To aid further developments, it is thus essential to always clearly state which processes are neglected. Unfortunately, we were unable to identify the assumptions for at least one of the analyzed model features in 32 of the 167 analyzed models.

7. Model calibration and validation

Most publications included calibration of the model (75%), but less included validation with an independent data-set (43%). There is especially a lack of published validation results for large-scale anammox and aerobic granular sludge reactors. Only one study shows limited validation results for a full-scale anammox-based reactor (Corbala-Robles et al., 2016) and only one for a pilot-scale aerobic system (Stevens et al., 1989). Of course, calibration and/or validation is not necessary for all modelling goals. For example, most partial nitritation-anammox granular sludge reactor models did not aim at quantitative predictions but at qualitatively understanding the relationship between micro- and mesoscale phenomena. Also the effect of alternative operational strategies on the reactor performance was explored in general, but not for one specific reactor with a specific influent and effluent requirements. Aerobic granular sludge models on the other hand, were mostly used for simulations of lab-scale systems. This indicates that aerobic and anammox granular sludge models are not (yet) used in the way that activated sludge models are used for flocculent sludge systems, e.g. to find a cost-effective approach to improve an existing full-scale plant through changes in the operation or configuration or to design a plant for the treatment of a specific wastewater stream (Brjjanovic et al., 2015). It could also be that models are already used for these purposes, for example using commercial simulators, but that the results are simply not published in scientific literature.

8. Future directions

8.1. Finding the appropriate degree of complexity

It is clear that the appropriate complexity of a model depends on the modelling goal. For fundamental insight, the required model complexity follows logically from the research question. For example, if the goal is to assess the influence of the oxygen set-point or wastewater type on the microbial population distribution, obviously the dynamics of the microbial population should be included by modelling intragranule transport of particulates (Batstone et al., 2004; de Kreuk et al., 2007b). However, if the focus is on optimizing the operation or design for a better overall reactor performance, the search for optimal complexity has to find the optimal model complexity for design and optimization of the overall reactor performance.

![Fig. 9. Two approaches that can be used to find the optimal model complexity for design and optimization of the overall reactor performance.](image)
influent needs to be elucidated. Different particle sizes probably react differently. For example, small colloids could enter the granule matrix, while larger particles probably cannot, but they might still attach to the surface. Also the exact role of protozoa in the degradation of particles in aerobic systems is unknown (Pronk et al., 2015). Secondly, the liquid phase transport in anaerobic systems appears to be unpredictable because of the high sensitivity towards changes in the reactor scale and operation. CFD simulations including interactions between the gas and liquid phase of differently sized reactors could help to better understand this dependency and develop rules of thumb for suitable assumptions in simpler reactor models. Also the hydraulic behaviours of full-scale anammox-based reactors and aerobic reactors during the un aerated feeding phase may warrant further characterisation. Finally, the mechanistic dependency of detachment and breakage rates on operating conditions needs further study to better understand the development of a granular sludge bed and to simulate changes in the granule size (distribution).

8.3. Model applications

First of all, more publications with validation results on full-scale aerobic and anaerobic granular sludge reactors could increase confidence in the quantitative simulation results and thus extend the applications beyond general, qualitative optimization projects. Secondly, heat balance modelling has further potential since it has only been applied once (Haugen et al., 2015). It could be used to find the optimal balance between heat recovery (which lowers the temperature) or heating (which increases the temperature) and biological conversion rates (which increase at higher temperatures). Thirdly, given that only a few aerobic granular sludge models considered phosphorus conversions, there is still room to better understand and optimize the phosphorus removal that occurs in full-scale systems, e.g. to determine the optimal aeration control strategy for phosphorus removal. Furthermore, problems with pH fluctuations in anammox systems could be diagnosed and possibly solved with models. Further research can also combine available precipitation models (Mbamba et al., 2015; Solon et al., 2017) with granular sludge models, like Feldman et al. (2017) did for an internal circulation reactor. This could help to understand precipitation dynamics in these systems, tackle problems caused by excessive precipitation and optimize recovery of resources like phosphorus. Recent additions of aerobic granular sludge reactor models in commercial simulation software, such as BioWin and SIMBA#, might stimulate a wider application of models for granular sludge.

9. Conclusions

This contribution reviewed granular sludge reactor models for anaerobic, aerobic and partial nitrification-anammox processes.

- A clearly defined modelling goal is not always provided, but it is necessary to find an appropriate model complexity and to differentiate between the many other available models.
- The immense variation in assumptions about the key phenomena in a granular sludge reactor can partly be explained by the different reactor types and goals.
- To eliminate habitual assumptions, further research should more clearly define the range of operational conditions and goals for which certain approaches can be used. In particular the applicability of biofilm models versus the use of apparent kinetics needs further study.
- More mechanistic understanding is needed on the dependency of the detachment rate on the operational conditions, the fate of particulate organics and the transition between plug flow and mixed conditions with increasing anaerobic reactor scale.
- More full-scale calibration and validation studies would help in extending the applications of aerobic and anammox granular sludge models beyond qualitative studies and quantitative predictions on lab-scale, e.g. to diagnose and prevent pH problems and optimize biological phosphorus removal.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2018.11.026.

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