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Model-based evaluation of an integrated high-rate activated sludge and mainstream anammox system

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Abstract

Wastewater treatment plants of the future aim at energy autarky. This could potentially be realized through a high-rate activated sludge (HRAS) process for COD redirection followed by a mainstream partial nitritation-anammox (PNA) process for nitrogen removal. The potential of this promising process option was evaluated in this study, through modelling and simulation. The impact of operating conditions on the unit processes was investigated first. Operation of a HRAS stage often implied a trade-off between maximizing the COD capture for energy recovery and minimizing residual COD in the effluent fed to the subsequent autotrophic PNA process. Moderate DO (0.3-0.5 g O₂.m⁻³) and SRT (0.3-0.5 d) were suggested to balance these needs, whereas maximizing settling efficiency in the subsequent settler was always desirable. Regarding the mainstream PNA process, the optimal DO setpoint corresponding with maximum nitrogen removal decreased with increasing biomass concentrations. Anammox remained the dominating nitrogen removal process during long-term dynamic simulations with fluctuating HRAS stage effluent (1.3-4.3 g COD.g⁻¹ N, 10-20°C), indicating the resilience and long-term stability of the PNA process at mainstream conditions. Plant-wide evaluations revealed that the combined HRAS-PNA system could achieve a comparable effluent quality as the CAS system, complying with EU regulations, while allowing 56% more influent COD to be redirected to sludge for energy recovery and over 60% savings in aeration energy. This illustrates the potential of being energy-neutral of the integrated HRAS-PNA system.

Keywords: mainstream anammox; partial nitritation; HRAS; energy-neutral; energy recovery; plant-wide modelling; dynamic simulation;
1. Introduction

Conventional municipal wastewater treatment plants (WWTPs) are typically designed to obtain a high removal efficiency of organic matter (expressed as chemical oxygen demand - COD) and nitrogen. This does not only require a large amount of aeration energy, but also leads to the loss of the energy present in COD as metabolic heat [1,2]. New configurations have been proposed to shift from the current energy-inefficient WWTPs towards energy-neutral ones [1,3,4], based on the AB process already established in the 1970s [5]. The first stage of such energy-neutral (or even energy-positive) WWTPs utilizes a high-rate activated sludge (HRAS) process, in which COD is concentrated and redirected for subsequent energy recovery through anaerobic digestion. This stage is followed by a partial nitritation-anammox (PNA) process for nitrogen removal (Fig. 1).

The HRAS process generally uses a higher food-to-microorganism (F/M) ratio, a shorter sludge retention time (SRT < 2d), a shorter hydraulic retention time (HRT ~ 0.5 h) and a lower dissolved oxygen concentration (DO < 1 g O₂.m⁻³), compared to conventional activated sludge (CAS) processes [6]. These differences result in a less energy-intensive (lower aeration energy) and more compact system. They also affect the COD removal behavior. For instance, due to the low SRT in a HRAS process, fast-growing heterotrophic bacteria are selected, which are only able to use the most readily biodegradable organics. As a result, part of the soluble COD which is treated as easily biodegradable in long-SRT (> 3d) CAS system should be considered as inert in the short-SRT HRAS system [7]. Therefore, the impact of operating conditions on the COD conversions in the HRAS process needs to be further specifically explored.

The PNA process consists of two steps. During the partial nitritation step, about half of the ammonium in the wastewater is oxidized to nitrite by ammonium oxidizing bacteria (AOB), while further oxidation to nitrate by nitrite-oxidizing bacteria (NOB) is prevented. In the
subsequent anammox conversion, the remaining ammonium and the produced nitrite are
combined to form dinitrogen gas (N$_2$), in the absence of oxygen and using CO$_2$ as a carbon
source [8]. Compared to the conventional nitrogen removal processes based on nitrification-
denitrification over nitrate, the PNA process (1) consumes up to 63% less aeration energy; (2)
does not require external COD for denitrification, which omits the need for chemical addition
and at the same time maximizes the possible alternative use of the COD present in municipal
wastewater for energy production through anaerobic digestion; (3) produces less (costly) sludge
to be disposed of and (4) emits less CO$_2$ due to the autotrophic nature of the process and lower
energy requirements [1,9]. The two steps of the PNA process can be realized in two separate
reactors or in a single reactor. In 2014, there were already more than 100 full-scale PNA reactors
operational. They were successfully applied to treat various types of warm wastewater with
high ammonium concentrations, such as reject water originating from anaerobic digestion (i.e.,
sidestream treatment) [10]. More than 80% of them were single reactor systems, as will be
considered in this study. Moreover, granular sludge systems were put forward because of its
high volumetric conversion rate and biomass retention [3].

The current challenge is in applying the innovative PNA process for the treatment of
municipal wastewater in the main line of the WWTP (i.e. mainstream anammox) [3]. The major
issues in this respect are the low temperature and low ammonium concentration of municipal
wastewater, combined with the presence of COD which may stimulate the growth of
heterotrophs, outcompeting the slowly growing anammox bacteria [11,12]. The feasibility of
PNA under mainstream conditions has been demonstrated in several laboratory and pilot-scale
studies [13–15]. Nevertheless, these studies also point out that the long-term stability of PNA
process (i.e. maintaining high process rate and low effluent nitrogen concentration) under
varying temperature (especially low temperature) and loading rates (e.g. COD and ammonium)
in full-scale WWTPs should be evaluated further [12,16].
Sufficient COD removal in the HRAS stage is crucial for avoiding the proliferation of heterotrophs and thus for a successful PNA stage. However, a higher COD removal in the HRAS stage does not necessarily mean a higher energy recovery [6,17]. While the HRAS process has already been successfully applied at full-scale in the past to maximize energy recovery from the influent COD, as the first step in a conventional AB process [18], the additional challenge for it nowadays is to simultaneously realize a sufficiently high COD removal efficiency in order to meet the influent requirements (i.e. low COD/N) of the downstream PNA stage. Therefore, to comprehensively assess the combined HRAS-PNA system, a plant-wide perspective is needed, as applied in the past for the evaluation of sidestream anammox processes [19].

Model-based investigations are useful for fast and rigorous assessment of the performance of WWTPs, in particular to analyse the interrelations among unit processes. As such, they can also be utilized for feasibility studies of leading-edge technologies and their integration with other wastewater treatment unit processes. Up till now, the HRAS-PNA system and/or similar systems (e.g. bioflocculation and two-stage PNA) have only been evaluated via rough mass and energy balance calculations [9] and steady state simulations [20–22]. A few studies applied life-cycle analysis for environmental assessment of systems with PNA in the mainline [23,24]. However, the feasibility and long-term stability of the HRAS-PNA system under practical dynamic conditions (e.g. temperature, hydraulic load, and substrate concentrations) remains to be evaluated.

In this contribution, the feasibility and long-term stability of the HRAS process for COD removal and capture, combined with a granular sludge PNA reactor for nitrogen removal, was evaluated through dynamic modelling and simulation. The effect of operating conditions (e.g. SRT, DO and biomass concentration) on the individual HRAS and PNA processes was investigated first, followed by steady-state and dynamic evaluations of the combined HRAS-
PNA system in a plant-wide context. Finally, the latter system was compared against a conventional activated sludge (CAS) system in terms of effluent quality and operational costs.

2. Materials and methods

2.1 The integrated HRAS-PNA system

The integrated HRAS-PNA system (Fig. 1) was put forward as proposed by Kartal et al. [3]. In this system, the HRAS stage is mainly responsible for organic carbon removal and redirection, whereas the PNA stage is mainly responsible for nitrogen removal. For the latter purpose, a compact granular sludge reactor was considered [3]. The sludge line (i.e. thickener, AD, dewatering and storage tank) was also included for a comprehensive plant-wide evaluation and was modelled as in the Benchmark Simulation Model no. 2 (BSM2) [25].

Figure 1. Schematic plant layout of the integrated HRAS-PNA system

2.2 High-rate activated sludge (HRAS) stage model

The possible biological COD conversions in a HRAS process are illustrated in Fig. 2. The short SRT in HRAS system compared to a CAS system results in different COD removal behavior, which requires system-specific modelling assumptions. The widely-used Activated
Sludge Model no. 1 (ASM1) which focuses on long-SRT (> 3d) activated sludge systems (i.e. CAS systems), is not directly applicable for the modelling of a short-SRT HRAS system, as clearly stated by the authors themselves [26] and demonstrated by Nogaj et al.[17].

The HRAS stage was modelled through an adapted ASM1, according to the approach by Smitshuijzen et al. [27]. In this approach, the efficiency of adsorption/bioflocculation (r9, Fig. 2) the HRAS unit and the efficiency of liquid/solid separation in the subsequent clarifier are lumped into a single parameter, $f_{\text{settle}}$, which represents the fraction of particulates removed and is estimated from plant data (detailed in A1.2, Appendix). All particulates are assumed to adsorb and settle equally well. The stoichiometric matrix and process rate expressions of the HRAS unit model are the same as the ASM1 implemented in the Benchmark Simulation Model no. 2 (BSM2) [25] and are detailed in Table A1 and A2, respectively. The associated parameter values of the HRAS unit model are detailed in Table A3.

The HRAS stage is composed of the HRAS unit and the subsequent clarifier (Fig. 1). The HRAS unit was modelled as five completely mixed aerated reactors in series to simulate a plug flow configuration as typically applied in practice [28]. The total volume of the HRAS reactors was calculated to be 2500 m$^3$, based on the mass concentration of mixed liquor suspended solids (MLSS) that was expected in the reactors. As the key operating parameters in the HRAS process, the SRT was controlled by manipulating the waste sludge flow rate (detailed in A1.2). The DO was controlled at a setpoint of 0.5 g O$_2$.m$^{-3}$ at the 4$^{\text{th}}$ bioreactor by applying a closed control loop [27]. The same oxygen transfer coefficient ($K_L a$) in the 4$^{\text{th}}$ reactor was then applied to all other bioreactors in the model. The model was implemented in Matlab & Simulink® (version R2016b).
2.3 Partial nitritation-anammox (PNA) model

The granular sludge PNA reactor was modelled as 1-D biofilm based on Mozumder et al.[29], including four bacterial groups: ammonium-oxidizing bacteria (\(X_{\text{AOB}}\)), nitrite-oxidizing bacteria (\(X_{\text{NOB}}\)), anammox bacteria (\(X_{\text{AN}}\)) and heterotrophic bacteria (\(X_{\text{H}}\)). The effect of temperature was taken into account (Table A7) according to the approach in Hao et al. [30]. The stoichiometric matrix, process rates and associated parameter values of the PNA model are detailed in Table A5-A7.

The granular sludge PNA reactor has a fixed total volume (granules + bulk liquid) of 2500 m³. This was determined based on the biomass concentration (4 kg VSS.m⁻³) and specific nitrogen loading rate (0.125 kg N.kg VSS⁻¹.d⁻¹) derived from a pilot-scale granular sludge PNA reactor treating real HRAS stage effluent [31] and the influent of the PNA stage (i.e. effluent of the preceding HRAS stage) (detailed in A2.2). The granules were assumed to be of equal size with a radius of 0.75 mm. The bulk DO concentration was controlled by a constant set-point and the biomass concentration was manipulated by changing the number of granules in the reactor. The PNA model was implemented in AQUASIM v2.1 [32].

2.4 Plant-wide comparison with conventional activated sludge (CAS) system

The behavior of the HRAS-PNA system was compared with the one of a conventional activated sludge (CAS) system. The latter was simulated using the closed-loop configuration of BSM2 (Fig. A4, [25]). The BSM2 CAS system also includes a primary clarifier, a thickener, an anaerobic digester, a dewatering unit and a storage tank for the reject water, allowing for plant-wide evaluation in terms of effluent quality, operational cost and energy recovery potential [25]. The same influent datasets from BSM2 were used for both systems for steady-state and dynamic simulations.
2.5 Evaluation criteria for plant-wide comparison

The influence of operating conditions on the HRAS and the PNA stages was evaluated based on COD distribution and nitrogen removal efficiency, respectively. As for the comparison between the HRAS-PNA and CAS systems, this study used the default evaluation criteria of BSM2, which provide a simple and objective means for comparison and have been developed specifically for comparing dynamic responses [25]. The evaluation criteria consists of effluent quality index (EQI, in kg pollution units.d⁻¹) and operational cost index (OCI) (detailed in A3.2). The EQI is a weighted average of relevant effluent concentrations (e.g. TN, COD and TSS). The OCI is calculated as a weighted sum of different costs, including aeration energy pumping energy, mixing energy, sludge production for disposal, external carbon addition, methane production and the heating energy. The aeration energy in the HRAS stage was calculated the same way as for the CAS system in BSM2, whereas the aeration energy in the PNA reactor was calculated based on the oxygen consumption and a typical engineering approach related to the SOTE (standard oxygen transfer efficiency, kg O₂.kWh⁻¹) (detailed in A3.3). The evaluation period was the last 364 days in the dynamic simulations.

2.6 Simulation strategies

The influence of operating conditions on the individual HRAS and PNA stages was investigated first, followed by plant-wide evaluation of the combined HRAS-PNA system. Finally, the latter system was compared against a CAS system in terms of effluent quality and operational costs. An overview of simulation strategies is summarized in Table 1.

The influent dataset of BSM2, which consists of both constant influent and full dynamic influent data (609 days), was used as input for simulations. The constant influent file contains the average values of one full year dynamic data that represents the influent of the municipal wastewater treatment plant in BSM2. The average influent concentrations of COD and total
nitrogen (TN) are 593 mg COD.L\(^{-1}\) and 55 mg N.L\(^{-1}\), with an average flow rate of 20 648 m\(^3\).d\(^{-1}\) (Table 1, detailed in Table A4).

### Table 1. Overview of simulation strategies. Values in parentheses are for the PNA stage.

<table>
<thead>
<tr>
<th>Scenario</th>
<th>COD(_{in})</th>
<th>TN(_{in})</th>
<th>SRT</th>
<th>DO</th>
<th>T</th>
<th>(f_{settler})</th>
<th>Biomass concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>g COD.m(^{-3})</td>
<td>g N.m(^{-3})</td>
<td>day</td>
<td>g O(_2).m(^{-3})</td>
<td>°C</td>
<td>-</td>
<td>kg VSS.m(^{-3})</td>
</tr>
<tr>
<td>HRAS stage*</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reference case</td>
<td>593</td>
<td>55</td>
<td>0.3</td>
<td>0.5</td>
<td>15</td>
<td>0.984</td>
<td>-</td>
</tr>
<tr>
<td>Factors influencing COD redirection</td>
<td>593</td>
<td>55</td>
<td>0.1-2</td>
<td>0.1-2</td>
<td>10-30</td>
<td>0.85-1</td>
<td>-</td>
</tr>
<tr>
<td>PNA stage**</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Reference case</td>
<td>(50)</td>
<td>(46)</td>
<td>-</td>
<td>(1)</td>
<td>(15)</td>
<td>-</td>
<td>4</td>
</tr>
<tr>
<td>Effect of biomass concentration and DO</td>
<td>(50)</td>
<td>(46)</td>
<td>-</td>
<td>(0.5-2)</td>
<td>(15)</td>
<td>-</td>
<td>(2-12)</td>
</tr>
<tr>
<td>Plant-wide: HRAS-PNA vs. CAS</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Steady state*</td>
<td>593 ()</td>
<td>55 ()</td>
<td>0.4 (-)</td>
<td>0.5 (0.5)</td>
<td>15 (15)</td>
<td>0.984</td>
<td>8</td>
</tr>
<tr>
<td>Dynamic***</td>
<td>variable</td>
<td>variable</td>
<td>0.4 (-)</td>
<td>0.5 (0.5)</td>
<td>variable</td>
<td>0.984</td>
<td>8</td>
</tr>
</tbody>
</table>

*B SM2 constant influent; **Effluent of the HRAS stage at reference scenario; ***BSM2 dynamic influent

For the HRAS stage, the effect of the operating conditions (DO, SRT and temperature) and combined adsorption/bioflocculation and settling efficiency \(f_{settler}\) on the performance was evaluated through steady-state simulations. For the PNA stage, the individual and combined influence of biomass concentration and bulk DO concentration on nitrogen removal was evaluated through steady-state simulations (Table 1). The effluent of the HRAS stage at reference scenario was used as the influent of the PNA stage for these simulations. For steady-state simulations, the HRAS stage model (in Matlab) was run with the constant influent data (Table A4) for 500 days, while the PNA stage model (in AQUASIM) was run with the effluent from the HRAS stage under reference case for 4000 days to ensure steady state was reached. The behavior of the HRAS-PNA system under dynamic conditions were evaluated using the full dynamic influent data for 609 days also from BSM2. The last 364 days (i.e. day 245-609) of the dynamic simulation was used for performance evaluation.
3. Results and discussion

3.1 HRAS stage for COD redirection

3.1.1 Modelling the HRAS stage

The biological conversions in activated sludge systems are presented in Fig. 2. The shorter SRT and HRT in a HRAS system compared to a CAS system results in different COD removal behavior in the two systems, which requires system-specific modelling assumptions.

Figure 2. The biological COD and nitrogen conversions in a high-rate activated sludge (HRAS) system. The state variables and processes with solid lines (r1-r8) are included in the model used in this study (i.e. ASM1 [26] and Smitshuijzen et al.[27]). The state variables and processes with dotted lines (r9-r11) are among those additionally included in the model of Nogaj et al.[17].

The readily biodegradable COD (Ss) is modelled as a single substrate with a single kinetic for CAS systems in ASM1[26]. However, the low SRT in HRAS process favors fast-growing bacteria which are only able to use the most readily biodegradable organic, i.e. part of the Ss in ASM1 [7,17]. Therefore, the effluent of HRAS process often has a higher soluble bridgeable COD compared to CAS process [7]. To describe this, three different approaches (S1-S3) have been proposed: (S1) spite the Ss into fast and slow fractions (i.e. dual soluble substrate)
and model them separately [7,17]; (S2) distribute a part of $X_S$ to $S_S$ [27] and (S3) increase the half saturation constant $K_S$ for $S_S$ [7].

The difference in modelling the removal of suspended COD (colloidal and particulate COD) lies in whether to model the adsorption/bioflocculation process explicitly. Suspended COD needs to be adsorbed by active biomass before being hydrolyzed and oxidized. The adsorption of suspended COD is a fast process and is considered instantaneously in ASM1[26]. However, due to the short HRT (i.e. contact/reaction time) in HRAS process, only a part of the suspended COD can be enmeshed by biomass [33]. To model the adsorption/ bioflocculation process, two different approaches (X1-2) have been proposed: (X1) integrate the production of extracellular polymeric substances (EPS), storage compounds (r10, Fig. 2) and adsorption of colloidal COD (r9, Fig. 2) into the ASM1[17,33] and (X2) lump adsorption/bioflocculation and settling efficiency into a single parameter ($f_{settler}$, detailed in A1.2) that can be identified with routinely measured data in WWTPs [27].

Currently, there are only few dedicated models available for HRAS system. Nogaj et al. [17] presented a mechanistic model (Approach S1&X1), in which new state variables (e.g. colloidal COD) and processes (e.g. r9-r11, Fig. 2) were added to ASM1. Despite its potential advantage of revealing more insights into the underlying processes, the introduction of new state variables and kinetic parameters requires a set of measurements (e.g. influent colloidal COD) that are usually not available for parameter estimation and model calibration. In contrast, Smitshuijzen et al. [27] presented a simpler model adaptation of ASM1 (Approach S2&X2) by implementing the combined adsorption/bioflocculation and settling efficiency ($f_{settler}$) and applying influent COD fractionation. The general applicability of the lumped $f_{settler}$ may require further validation in other WWTPs, while its simplicity allows for easier integration of the HRAS process model in plant-wide models, as is the case in this study.
3.1.2 Factors influencing COD redirection

The influent COD entering the HRAS system can go three possible routes: (i) COD loss due to mineralization i.e., oxidation of COD to CO$_2$ by biomass (r1-2, Fig. 2); (ii) end up in the HRAS effluent, fed to the PNA stage; (iii) COD capture in the sludge, fed to anaerobic digester for energy recovery. The COD distribution between these routes is crucial for the energy recovery potential and the performance of downstream autotrophic N removal process. A high COD capture in sludge is preferred for energy recovery, while a low effluent COD (i.e. low COD/N ratio) is demanded for subsequent mainstream PNA. The distribution was found affected by DO, SRT and temperature (T) as key operating conditions as well as by the combined adsorption and settling efficiency (characterized by the parameter $f_{settler}$), as summarized in Fig. 3. For the reference case, 22% of the influent COD was mineralized, 14% was in the effluent and 66% was captured in sludge, with an effluent COD/N ratio of 2.4 (g COD.g N$^{-1}$) (Fig. 3).
Figure 3. COD distribution as a fraction of influent COD and effluent COD/N ratio with different (A) DO, (B) SRT, (C) temperature and (D) f_settler in the HRAS stage (Influent COD=593 g COD.m⁻³, among which Xs=218 g COD.m⁻³ and Ss=204 g COD.m⁻³. The reference case is marked with dotted box.)

At DO concentrations lower than the reference case (<0.5 g O₂.m⁻³), the effluent COD decreased sharply with increasing DO concentration while the COD mineralization and COD capture increased accordingly (Fig. 3A). The lower effluent COD was mainly attributed to the improved soluble biodegradable COD removal at higher DO (r1, Fig.2). However, further increase of DO (>0.5 g O₂.m⁻³) showed marginal effect on influent COD distributions (Fig. 3A), indicating that DO was no longer the limiting-factor for COD removal in a HRAS process operated at a SRT of 0.3d. This trend is in line with the findings of the pilot-scale study from Nogaj et al.[17] and the full-scale investigations from de Graaff et al.[28]. The results imply...
that moderate DO set point (0.3-0.5 g O₂.m⁻³) should be applied in the HRAS stage for a sufficient COD removal and COD redirection for energy recovery while avoiding unnecessary aeration cost.

With increasing SRT, the effluent COD decreased while the mineralization of influent COD increased (Fig. 3B). Concerning the COD capture in sludge, it first increased (SRT of 0.1-0.3d) and then decreased gradually (SRT of 0.3-2d). The SRT determines the period during which particulates/biomass are retained within the system and was regulated by changing the waste sludge flow (Qₘₚ) in this model. At a low SRT (0.1d), the biomass concentration was too low and the removal of Ss was therefore limited, resulting a high effluent COD (40%, Fig. 3B). The Ss removal increased with increasing SRT and thus the mineralization of influent COD accordingly. However, higher mineralization also means more COD was wasted and less COD was left, to sludge formation (Fig. 3B). Similar trend was also observed in the pilot-scale HRAS systems by Jimenez et al.[6] where COD directed to sludge peaked at a SRT of 0.3d in the tested range of 0.1-2d. The results imply that a high SRT is desired to minimize effluent COD while a moderate SRT (e.g. 0.3d) is required to maximize the COD redirection for energy recovery. A SRT of 0.3-0.5d would be appreciate to balance these two needs. Noteworthy, the SRT needs to be chosen before building the HRAS unit, as it would affect the dimension of the bioreactors.

With increasing wastewater temperature, the effluent COD and COD capture in sludge decreased while the COD mineralization increased (Fig. 3C). Higher temperature improved biomass activities and overall process rates. This led to increased COD removal (low effluent COD) and mineralization through heterotrophic growth (r₁-2, Fig.2). The hydrolysis of Xs and biomass decay (r₇ and r₄₋₅, Fig.2) would be enhanced as well. Consequently, the COD capture in sludge decreased. These results imply that higher temperature may be favourable for COD removal of the HRAS stage but not necessarily for energy recovery. In regions that experience
large (seasonal) temperature variations, it could have a significant effect on the system performance throughout the year. Low temperature (e.g. in winter) may result in insufficient COD removal in HRAS stage and thus affect the autotrophic nitrogen removal in the subsequent PNA stage, as observed at the WWTP Dokhaven during winter [16]. However, temperature depends on the environmental conditions and cannot be easily or economically manipulated in full-scale WWTPs, which limits its potential use as a control strategy.

One of important parameters of the HRAS model adapted in the present study is the combined adsorption/bioflocculation and settling efficiency ($f_{settler}$). By definition (detailed in A1.2), removal of particulate COD would increase with increasing $f_{settler}$ and the effluent COD would decrease accordingly while the mineralization of influent COD would remain unaffected (Fig. 3D). The higher the $f_{settler}$, the better in terms of energy recovery. Therefore, more efforts are required on improving the adsorption/bioflocculation in the HRAS unit (e.g. by adding chemicals or pretreatment with chemically enhanced primary treatment) and the settling efficiency in the subsequent settler (e.g. by applying coagulation and dissolved air flotation).

Overall, the operation of a HRAS stage aims at maximizing COD capture in the sludge for energy recovery and minimizing effluent COD for the subsequent removal of PNA for nitrogen removal. A trade-off was observed for the selection of SRT and temperature, while improving the efficiency of adsorption/bioflocculation and settling was always desirable. Moderate DO (0.3-0.5 g O$_2$.m$^{-3}$) and SRT (0.3-0.5 d) were suggested for operating the HRAS stage.

3.2 The partial nitritation anammox (PNA) stage for nitrogen removal

3.2.1 Factors influencing nitrogen removal

Aiming at a high and stable N removal, the influence of several influent characteristics (e.g. NH$_4$-N concentration/load) and operating conditions (e.g. DO) on the N removal of
granular sludge one-stage PNA reactors have been evaluated through experiments and modelling and simulations (Table 2). For instance, increasing influent ammonium concentration (i.e. load) results in gradual decrease in N removal at fixed DO concentration [34], while increasing temperature improves N removal in granular sludge one-stage PNA reactor [15,34]. The N removal first increases with increasing influent COD (or COD/N) and then declines as it further increases [29,35], which is also observed for bulk DO concentrations [36,37]. Aeration pattern influences the N removal as well, with continuous aeration leads to higher N removal than that of intermediate aeration [38]. A minimum residual ammonium concentration in the PNA reactors is found necessary for NOB repression and therefore affects the N removal [39]. Granule size and its distribution have been shown to affect the nitrogen conversions as well as operation window of other parameters (e.g. DO) for optimal N removal [40,41]. The balance of microbial community and their activities is crucial for a stable and efficient PNA process. The AOB, NOB and heterotrophic bacteria preferentially grow in small granules and flocs, which leads to another control strategy in granular sludge PNA reactors, being selective removal of small granules and flocs [42,43]. Besides, the biomass concentration could also significantly affect the determination of other operating conditions (e.g. DO) and the performance of a bioreactor. However, its potential impact has not been explicitly studied for granular sludge PNA reactors.
Table 2. Overview of factors influencing the N removal from granular-sludge one-stage PNA reactors reported in literature. The second last column schematically shows a mini-graph (where applicable) representing how N removal reacts to changes of the influencing factors.

<table>
<thead>
<tr>
<th>Factors</th>
<th>Conditions</th>
<th>Approach</th>
<th>Graphic representation</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>T (°C)</td>
<td>NH₄-N (g N.m⁻³)</td>
<td>CODin (g COD.m⁻³)</td>
<td>Simulation</td>
<td>[34]</td>
</tr>
<tr>
<td>N/NH₄-N concentration</td>
<td></td>
<td></td>
<td></td>
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<td>[35]</td>
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<td>[15]</td>
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<td>20/30</td>
<td>1850</td>
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3.2.2. Effect of biomass concentration – specific nitrogen load

The impact of biomass concentration on the steady-state performance of the mainstream granular sludge PNA reactors is first investigated at a fixed DO concentration (Fig. 4). The interaction between the biomass concentration and the DO is assessed subsequently (Fig. 5).

With increasing biomass concentration, the nitrogen removal efficiency first increased and then decreased (Fig. 4A), as also reflected by the N₂ production (Fig. 4B). At low biomass concentration (< 4 kg VSS.m⁻³), ammonium conversion was still largely incomplete (33 g NH₄-N.m⁻³ remaining, 26.7% conversion). At intermediate biomass concentrations (4-8 kg VSS.m⁻³), the anammox fraction in the granules increased (Fig. 4A) and N₂ became the primary
nitrogen compound (Fig. 4B). As the biomass concentration further increased, the DO concentration at the center of the granules increased (Fig. 4A), which reflects the elevated oxygen penetration in the granules. Consequently, the NOB activity increased while the anammox activity got inhibited, as confirmed by the change of corresponding biomass fractions (Fig. 4A) and shift of the dominant nitrogen compound from N₂ and to nitrate (Fig. 4B). Noteworthy, nitrite accumulation was observed at intermediate biomass concentrations (Fig. 4A), implying an overcapacity of nitrite producer (AOB) over nitrite consumers (NOB, anammox and heterotrophic bacteria) at these conditions.

The increased DO penetration depth with increasing biomass concentrations (Fig. 4B) could be explained by the lower substrate (e.g. NH₄⁺) surface loads, as increasing biomass concentrations imply a higher biofilm surface. Previous studies have shown that ammonium surface load can inversely affect the oxygen penetration in granular sludge PNA reactors at sidestream conditions [37]. The influence of bulk DO concentration on the mainstream PNA reactor was evaluated as well and the results (Fig. S4) were consistent with previous studies (Table 2).
Figure 4. Influence of biomass concentration on the steady-state performance of the mainstream PNA process at a fixed DO concentration (1 g O₂, m⁻³): (A) particulate mass fractions, nitrogen removal efficiency and DO concentration at the center of the granules; (B) bulk concentrations of ammonium (sₙH), nitrite (sₙO₂), nitrate (sₙO₃) and nitrogen gas (sₙ₂).

The combined influence of biomass concentration and DO was studied by identifying the optimal bulk DO concentrations, corresponding with maximum N removal at different biomass concentrations (Fig. 5). Overall, the optimal bulk DO concentration for maximum N removal decreased with increasing biomass concentration. N removal efficiencies higher than 90% could be achieved with biomass concentration above 6 kg VSS.m⁻³ and DO concentration lower than 0.75 g O₂.m⁻³. The higher nitrogen removal efficiency than if there would be only anammox process for N₂ production (89%, according to the anammox stoichiometry [45]) was
a result of positive interaction between anammox bacteria and heterotrophic denitrifiers under the tested scenarios (bCOD/N ~1, Table 1). This was confirmed by labeling and tracking the N₂ production in the model and consistent with previous observations [29,35,46].

Figure 5. The optimal bulk DO concentrations at which maximum nitrogen removal was obtained in granular sludge PNA reactors with different biomass concentrations.

Biomass concentrations of 25-35 kg VSS.m⁻³ have been applied in full-scale granular PNA reactors for reject water treatment [47]. Therefore, the biomass concentration used for scenario analysis (2-12 kg VSS.m⁻³) should be practically feasible to maintain. The results from this study suggest that lower DO set point could be applied for mainstream granular sludge PNA reactors with higher biomass concentrations.

3.3 Plant-wide comparison of the HRAS-HRAS and CAS system

3.3.1 Steady state COD and N mass balances

The steady-state simulation results for the combined HRAS-PNA system and the CAS system were summarized in terms of plant-wide mass balances for COD and total nitrogen (TN) (Fig. 6&7). With a comparable effluent COD (i.e. COD removal) in both systems (Fig. 6A), the
HRAS-PNA system resulted in a lower COD mineralization and significantly more influent COD to be redirected to the anaerobic digester (AD) for energy recovery, compared to the CAS system (50.8% vs. 34.9%, Fig. 6A). As illustrated in Section 3.1.2, this was mainly due to the low SRT and DO applied in the HRAS stage. Moreover, less excess sludge was resulted in the HRAS-PNA system, even though more sludge directed to the anaerobic digester, indicating that the sludge from HRAS process was more biodegradable than that of CAS system.

**Figure 6.** Plant-wide steady-state simulation results – comparison between HRAS-PNA system and CAS system in terms of (A) Fate of influent COD and (B) Fate of influent TN. (conditions: HRAS: SRT=0.4 d, DO=0.5 g O\(_2\).m\(^{-3}\); PNA: Biomass concentration=8 kg VSS.m\(^{-3}\), DO=0.5 g O\(_2\).m\(^{-3}\); CAS: closed-loop BSM2)

As for nitrogen, the HRAS-PNA system had a higher TN removal (92%) than that of the CAS system (80%) at the tested conditions (Fig. 6B). No noticeable nitrogen removal was found in the HRAS reactors according to the mass balance (Fig. 7), indicating that there was no significant nitrification and consequently no nitrite/nitrate for heterotrophic denitrification at the tested HRAS stage conditions (i.e. SRT=0.4 d and DO=0.5 g O\(_2\).m\(^{-3}\)). Despite the low volumetric fraction (0.5%) of the reject water from the sludge line (sidestream), it accounted for a significant part (ca. 35%) of the nitrogen load to the PNA stage (Fig. 7).
The steady-state effluent concentrations of COD (63.6 g COD.m\(^{-3}\)) and TN (4.6 g N.m\(^{-3}\)) and TSS (27.6) of the HRAS-PNA system were well below the limits for urban wastewater treatment (EU council directive 91/271/EEC, Fig. 9A), demonstrating the feasibility of this system, which was further assessed with dynamic simulations.

**Figure 7.** Process flowsheet including flow and mass (COD, TN) balances for the HRAS-PNA system at steady-state. (Conditions: HRAS: SRT=0.4 d, DO=0.5 g O\(_2\).m\(^{-3}\); PNA: Biomass concentration=8 kg VSS.m\(^{-3}\), DO=0.5 g O\(_2\).m\(^{-3}\))

### 3.3.2 Dynamic performance – overall effluent quality and operational costs

The dynamic simulation results for the HRAS-PNA system and CAS system are summarized in Fig. 8 with respect to the effluent quality and operational cost index (OCI).
Figure 8. Comparison of dynamic simulation performance of the HRAS-PNA and CAS systems in terms of effluent quality and operational cost index (HRAS-PNA_SS and HRAS-PNA_DYN represent the results of steady-state and dynamic simulations, respectively. The limits are set according to the EU council directive 91/271/EEC. The operational cost indexes were calculated after taking weighting factors into account.)

The yearly average effluent concentrations of COD, TN and TSS of the HRAS-PNA system were all below the EU limits (Fig. 8A), agreeing with the results obtained from the steady-state simulations. The average effluent TSS concentration (30.5 g TSS.m⁻³) was slightly lower than the limit (35 g TSS.m⁻³) and was mostly likely a result of influent TSS (36.8 ± 11.2 g TSS.m⁻³). It should be noted that no secondary settler was assumed after the granular sludge PNA reactor. For a safety factor, one could add a secondary settler that is normally present in a CAS system or incorporate a settler within the PNA reactor [10]. The simulated effluent concentrations of the full-scale HRAS-PNA system are close to experimentally measured results of lab or pilot-scale mainstream anammox reactors. For example, average effluent TN concentration below 10 g N.m⁻³ was also achieved in two long-term lab-scale mainstream anammox reactors treating pre-treated sewage [14]. The average effluent TSS in a pilot-scale granular sludge PNA reactor treating real HRAS stage effluent was 30 g.m⁻³ [31]. However, the effluent quality under dynamic simulations was less pronounced than that under steady-state
simulation (Fig. 8A), implying the necessarily of performing dynamic simulation for feasibility
analysis of a novel system layout.

In terms of the operational cost, the HRAS-PNA system has a 107% lower OCI than the
CAS system (-667 vs. 9447, Fig. 8B). The significant reduction was mainly due to 52% higher
methane production (thus energy production), no carbon source dosage and 64% lower aeration
energy consumption in the former system (Fig. 8B and detailed in Table A11). The higher
methane production was a result of the higher COD captured in the HRAS stage (Fig. 6A),
while the savings on carbon source dosage was due to the autotrophic nitrogen removal in the
PNA stage. As for the lower aeration energy, it was a combined effect of the lower DO
concentration in the HRAS stage (i.e. less mineralization, Fig. 6A) and lower oxygen
consumption of the partial nitritation pathway in the PNA stage compared to the full
nitrification pathway in the CAS system. With respect to energy, the energy consumption in the
HRAS-PNA system for aeration, pumping, mixing and heating (AD) could be compensated by
the energy production from methane, with a net production of 4918 kWh.d⁻¹, in contrast to a
net consumption of 3179 kWh.d⁻¹ in the CAS system (Table A12). Even though the OCI does
not consider all WWTP energy consumptions exhaustively (e.g. building energy requirements),
this result illustrates the great potential of the HRAS-PNA to be energy-neutral or even energy-
positive. Besides, due to the short HRT in the HRAS reactors and the high biomass
concentration of the granular sludge PNA reactor, the HRAS-PNA system has a much lower
footprint than the CAS system (e.g. 5000 m³ vs. 12000 m³ for bioreactors in this study).

Overall, the HRAS-PNA system could achieve effluent quality that complies with EU
regulation with a significantly lower operational cost and aerial footprint, compared to the CAS
system.
3.3.3 Dynamic performance- feasibility of PNA process under dynamic mainstream conditions

To better investigate the feasibility of PNA process under dynamic mainstream conditions, the profiles of the influent temperature, COD/N ratio and effluent TN concentration during the dynamic evaluation period (364 days) were zoomed in and plotted in Fig. 9. It seems clear that low temperature had a negative impact on the N removal (shown as effluent TN) of the HRAS-PNA system (Fig. 9A). However, when it comes to the impact of the influent COD/N ratio, it is not straightforward to interpret from the dynamic profiles (Fig. 9A&B). Therefore, correlation analysis was performed to further gain insight into the relation between fluctuations of influent characteristics on the COD removal in the HRAS stage and N removal in the subsequent PNA stage (Table 3).
Figure 9. Profile during the dynamic evaluation period (364 days): (A) influent temperature, effluent TN concentration of the HRAS-PNA system (blue line) and the CAS system (black line); (B) Influent (i.e. effluent of the HRAS stage) COD/N ratios of the PNA stage.

Table 3. Pearson correlation analysis between influent characteristics, COD removal, nitrogen removal and effluent concentrations of the HRAS-PNA system during the 364 days dynamic evaluation period

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</table>

Note: All correlations are significant at the 0.01 level (2-tailed). N=34944. Green and red for positive and negative correlation coefficients (R), respectively. OLR represents organic loading rate (kg COD.d⁻¹); NLR represents nitrogen loading rate (kg N.d⁻¹); bCOD is the sum of Ss and Xs; Con_AN denotes the contribution of anammox bacteria in N removal (%).

The COD removal (%) in the HRAS stage during the dynamic simulations was found positively correlated with influent temperature (9.5-20.5°C, R=0.16), as observed before in steady-stage simulations of the individual HRAS stage (Fig. 3C). Consequently, the COD/N ratios of the HRAS stage effluent (i.e. PNA stage influent) was negatively correlated with influent temperature (R=-0.47). The N removal in the PNA stage was positively correlated with temperature (R=0.47) but negatively correlated the COD removal (R=-0.28) and effluent COD/N ratios (R=-0.35) of the preceding HRAS stage. Concerning the impact of the influent characteristics of the integrated HRAS-PNA system, the influent COD/N ratios was found rather unimportant for the COD removal in the HRAS stage (R=0.04) and therefore unimportant for the N removal in the PNA stage and the overall performance.
The results indicate that temperature could directly affect the PNA process per se and indirectly affect the PNA stage by influencing COD removal in the preceding HRAS stage. This implies that high effluent TN and thus poor reactor performance could be expected for PNA reactor at low temperature (e.g. winter), as reported in a pilot-scale reactor by Hoekstra et al.[16]. Furthermore, the results also suggest high influent COD/N ratios (1.3-4.3 g COD.g⁻¹ N) could be problematic for the PNA stage, in line with observations in previous experimental studies [44,48]. Organic matter can stimulate the growth of heterotrophic bacteria that compete with AOB for oxygen and with anammox bacteria for nitrite and thus affect the N removal. On the other hand, low influent COD/N ratios (e.g. < 0.5 g bCOD.g⁻¹ N) could also help improve the nitrogen removal in anammox reactors [35,44,49]. Moreover, stronger correlations were found between the bCOD/N ratios and all N-related parameters than that of COD/N ratio (Table 3), illustrating that the bCOD/N ratio could be a better indicator for predicting N removal in PNA reactors and for clarity when comparing results from different studies on the impact of organic matter.

Anammox process was found the primary for nitrogen removal in the PNA stage throughout the whole evaluation period, accounting for 75-95% of the nitrogen removal (Fig. A5). This implies that despite a negative impact of the influent COD/N ratios (1.3-4.3 g COD.g⁻¹ N, corresponding to 0.5-2.4 g bCOD.g⁻¹ N) on the nitrogen removal of the PNA process, the heterotrophic bacteria cannot outcompete anammox bacteria, indicating the resilience the PNA system under flocculating mainstream conditions.

The effluent quality of this combined HRAS-PNA system was shown to adequately comply with the current EU regulations for yearly average (Fig. 9A), despite the fluctuations of influent characteristic. However, the effluent TN concentration could violate the regulations on a daily basis during low temperature period (Fig. 9A). Moreover, the N removal in the HRAS-PNA system appears more sensitive to the temperature compared to the CAS system.
This raises questions on the long-term reliability of the combined system for municipal wastewater treatment; therefore, active real-time control would be needed to achieve a more stable and reliable system performance.

Temperature and influent COD/N ratios of the PNA stage (i.e. effluent COD/N ratios of the preceding HRAS stage) were shown crucial for system performance. Temperature is not an easily/economically manipulated variable but rather a disturbance variable in large scale applications; therefore, the COD/N ratios of HRAS effluent (i.e. COD removal in the HRAS stage) should be the focus for potential control strategies. This can be tackled in two ways: 1) proactive management of COD removal in the HRAS stage; 2) reactive/counteractive control in the PNA stage while receiving influent with high COD/N ratios. In this regard, many control strategies have been proposed or tested for PNA process for both sidestream (high-strength) and mainstream (low-strength) applications [38,50]. However, control strategies for HRAS system are still largely lacking [51]. SRT and DO were shown to be important operating parameters, a real-time control strategy which can adjust the SRT (e.g. via MLSS control and waste sludge flow) and/or DO (e.g. aeration intensity) in accordance with the effluent COD/N ratio would be most desirable.

3.4 Model implications and limitations

The model simulation results of this study show that the integrated HRAS-PNA system was capable of municipal wastewater treatment and producing effluent that adequately complies with the current EU regulations, with a much lower operational cost compared to the CAS system. It was found the advantages of the HRAS-PNA system was less pronounced under dynamic conditions than under steady-state conditions (e.g. effluent quality, Fig. 9A). The COD removal (in the HRAS stage) and N removal (in the PNA stage) were both reduced during low temperature period (Fig. 3C and Fig. 9A). Moreover, dynamic simulation also highlighted that the N removal in the HRAS-PNA system was more sensitive to temperature compared to the
CAS system (Fig. 9A). Therefore, it is important to use dynamic influents while evaluating the feasibility and performance of certain processes via modelling, as they can reproduce the behaviour of real influent and their effects on the effluent and sludge characteristics [52].

Concerning the HRAS stage model, the combined adsorption/bioflocculation and settling efficiency, $f_{settler}$, is a simple parameter to model the redirection of influent COD to the sludge line. However, it should be noted that in reality the removal of particulate COD through adsorption/bioflocculation is affected by the SRT and DO [6,17]. By lumping the efficiency of these two processes into the single parameter ($f_{settler}$), the general trend of the modelling results agrees with findings of experimental studies whereas the effect of SRT and DO on the adsorption/bioflocculation process cannot be fully reflected via this model. SRT and DO were shown to be important operating parameters, a real-time control strategy which can adjust the SRT and/or DO in accordance with the effluent COD/N ratio would be most desirable. However, more research is needed to understand the fundamental mechanisms of the bioflocculation processes and thus the influence of operating conditions. Moreover, influent COD fractionation would also affect the COD distribution in HRAS stage (Fig. A2 and [27]) and the model outcome. Dedicated influent characterisation is needed in this respect for the modelling of HRAS process [53].

Regarding the PNA stage, the results revealed the interactive impact of biomass concentration and DO on the N removal. A unified granule size was assumed in this modelling study. While considering granule size distribution in the model may give slightly different quantitative results, it is normally sufficient to predict overall nitrogen removal in PNA process without size distribution [41]. In this study, the 1-D granular sludge PNA model is implemented in AQUASIM while the HRAS stage (including the sludge line) models are implemented in Matlab & Simulink. To implement the PNA model also in Matlab would make it easier for future plant-wide evaluations.
It should be noted that optimization of the two systems (e.g. maximal COD or N removal) was not the purpose of this study. Therefore, the operational conditions used in the simulations for comparison were not the optimal but the commonly applied conditions in practice for each system. Even though the results should not be interpreted in a fully quantitative way, they nevertheless provide a valuable indication of the potential and shortcomings of the conceptual HRAS-PNA system, from an integrated system perspective.

4. Conclusions

The performance of a promising combined HRAS-PNA system was evaluated through modelling and simulations, from both individual and plant-wide perspective. Model results show that the operation of a HRAS stage often implies a trade-off between maximizing the COD capture in the sludge for energy recovery and minimizing effluent COD for the subsequent implementation of PNA process, moderate DO (0.3-0.5 g O₂.m⁻³) and SRT (0.3-0.5 d) were suggested for this purpose. For granular sludge PNA reactors, a higher biomass concentration would allow a lower DO set point for maximum nitrogen removal. The anammox process remained dominant in nitrogen removal throughout the one-year dynamic evaluation period with varying influent COD/N (1.3-4.3) and temperature (10-20 °C), indicating the resilience and long-term stability of the PNA system at mainstream conditions. However, both COD (in HRAS) and N removal (in PNA) were compromised at low temperature conditions. Overall, steady-state and dynamic simulations showed that the integrated HRAS-PNA system could achieve effluent quality that complies with EU regulations with a significantly lower operational cost, compared to the CAS system.
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References


