Oxygen transfer efficiency in an aerobic granular sludge reactor: dynamics and influencing factors of alpha

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Abstract

In the pursuit of reducing carbon footprint and in view of increasing energy prices, energy efficiency is more important than ever before. Batch-wise operated aerobic granular sludge reactors consume up to 50% less energy compared to conventional activated sludge systems because pumping energy is reduced and mixing equipment is not needed. Further energy reduction efforts should therefore target aeration energy requirements. The alpha factor is an important factor influencing the oxygen transfer efficiency, however the dynamic behaviour of alpha has hardly been investigated in general and never for an aerobic granular sludge reactor. This study showed that alpha increases during the aeration phase of a cycle due to the influence of different process parameters. Through a data analysis study of 175 batch cycles of the Prototype Nereda® installation in Utrecht over the summer and winter period of 2020-2021, the exchange ratio and temperature were identified as the main influencing factors on the rate of increase of alpha in a batch cycle. A higher exchange ratio was related to a slower increase in alpha over the aeration phase, while a higher temperature was related with a faster increase in alpha. Moreover, alpha was characterized by a same minimal value at the beginning of every aeration phase, which could be explained by the adsorption of soluble biodegradable organic carbon described by a Langmuir adsorption model. Two mathematical models, a decreasing exponential and a first order model, were set up to unravel the dynamic behaviour of alpha. Both models were discussed in view of their practical implications for the design and performance optimization of aerobic granular sludge reactors and other batch-wise operated aerobic wastewater treatment systems.

Keywords

Aeration; oxygen transfer efficiency; alpha factor; aerobic granular sludge (AGS); batch-wise operation; wastewater treatment

Nomenclature

English alphabet

Abbreviation	*Definition*
AGS	Aerobic Granular Sludge
AOB	Ammonia-oxidizing bacteria
ER	Exchange ratio
MLSS	Mixed liquid suspended solids
NOB	Nitrite-oxidizing bacteria
OHO	Ordinary heterotrophic organisms
PAO	Phosphate accumulating organisms
WRRFs	Water and resource recovery facilities

Symbol	*Definition*	*Unit*
$bCOD_S$	Soluble biodegradable organic carbon	$g.m^{-3}$
C_{O_2}	Dissolved oxygen concentration in water	$g\ O_2\ m^{-3}$
$C^*_{O_2}$	Dissolved oxygen concentration in water at saturation	$g\ O_2\ m^{-3}$
F	Fouling factor	-
K_{eq}	Langmuir equilibrium constant	$g.m^{-3}$
$k_L a_{O2}$	Gas-liquid mass transfer coefficient in process water	h^{-1}
$k_L a_{O_2,clean}$	Gas-liquid mass transfer coefficient in clean water	h^{-1}
OTE	Oxygen Transfer Efficiency	-
Q_{in}	Influent flow rate	$m^3.h^{-1}$
$rbCOD_S$	Soluble readily biodegradable organic carbon	$g.m^{-3}$
$sbCOD_S$	Soluble slowly biodegradable organic carbon	$g.m^{-3}$

SRT	Sludge Retention Time	d
ΔT_{feed}	Length of the feeding phase	h
V	Water volume	m^3
V_{batch}	Volume of influent added during the feeding phase	m^3
$V_{reactor}$	Total volume of the reactor	m^3
W_{O_2}	Mass flow of oxygen fed by the blower to the aeration tank	g O_2 h^{-1}

Greek alphabet

α	Alpha factor	-
αF	Alpha factor for fouled diffusers	-
τ	First order time constant	h

Subscripts and superscripts

$_{ini}$	at the beginning of the aeration phase	
$_{end}$	at the end of the aeration phase	
Γ_{ads}	Amount of surface-active compounds adsorbed	mg surface active compounds.m^{-2} air bubble
Γ_{ads}^{max}	(Maximum) adsorption capacity	mg surface active compounds.m^{-2} air bubble

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1. Introduction

In the pursuit of reducing carbon footprint and in view of increasing energy prices, energy efficiency is more important than ever before. Wastewater treatment plants require about 1-3% of the global energy use [1] and are therefore often listed as significant energy consumers of the public sector. A new proposal for the EU Directive on energy efficiency forces public sector activities, including the treatment of wastewater, to reduce the annual energy consumption by 1.7% every year [2].

Reducing energy consumption fits in the ongoing paradigm shift in which wastewater treatment plants are increasingly regarded as water resource recovery facilities (WRRFs), which do not just provide clean water but also energy, nutrients and other recovered products. To increase the energy efficiency of a WRRF, many recent studies focus on opportunities to produce energy by harvesting the embedded energy in wastewater with the goal of having energy positive WRRFs [3–5]. These studies focus on an enhanced biogas yield through anaerobic digestion, sludge pre-treatments, on-site combined power and heat generation and co-digestion of sludge with food waste [6]. The goal of having energy producing WRRFs will not only be successful by an increase in the efficiency of energy production, a reduced energy consumption is needed as well.

Aeration is the most energy demanding process in a WRRF. It can take up to 75% of the overall energy expenditure of conventional wastewater treatment plants [7]. The introduction of the increasingly applied batch-wise operated aerobic granular sludge reactors reduces the energy up to 50% compared to continuous activated sludge systems [8]. The aerobic granular sludge process removes nutrients and organics in a single reactor instead of in separate reactor compartments which makes energy-intensive recycle pumps and mixers unnecessary. Due to the lower pumping and mixing energy need, the fraction of energy consumption spent on aeration is larger compared to continuous activated sludge plants (e.g. 83% [9]). Further energy reduction efforts of batch-wise operated aerobic granular sludge reactors should therefore target aeration energy requirements.

The aeration system provides oxygen to the water to meet the microbial oxygen requirements. The aeration system design and performance can be analysed by defining the oxygen transfer efficiency (OTE). It represents the fraction of oxygen provided by the blower that is transferred from the gas phase to the liquid phase (Eq 1.) [10].

$$\mathrm{OTE} = \frac{\alpha\, \mathrm{F}\, \mathrm{k_La_{O2}} \left(\mathrm{C}^*_{\mathrm{O_2}} - \mathrm{C_{O_2}}\right) V}{\mathrm{W_{O_2}}} \qquad \text{Eq.1}$$

The oxygen transfer efficiency is characterized by the gas-liquid mass transfer coefficient in clean water, $\mathrm{k_La_{O2}}$ (h^{-1}), decreased by a fouling factor F (-) to incorporate diffuser fouling and the alpha factor α (-) to introduce the dependency of operational and environmental conditions of the process [11]. $(C^*_{O_2} - C_{O_2})$ represents the difference between the dissolved oxygen concentration in water at saturation ($C^*_{O_2}$, g O_2 m^{-3}) and the dissolved oxygen concentration in water (C_{O_2} , g O_2 m^{-3}) which is the driving force for oxygen transfer. V is the water volume (m^3) and $\mathrm{W_{O_2}}$, the mass flow of oxygen fed by the blower to the aeration tank (g O_2 h^{-1}).

The alpha factor is an important factor describing the oxygen transfer efficiency of wastewater treatment plants [12]. Continuous well-mixed systems with activated sludge are characterized by rather constant alpha factors over the treatment length, while the alpha factor of continuous plug flow reactors is low at the inlet of the aerobic reactor and high at the outlet [13–16]. A gradual improvement of the aeration characteristics was also observed within cycles of batch reactors [17–20].

The gradual improvement of the aeration characteristics is likely due to slow degradation of surface-active compounds such as fatty acids, proteins, oils, soaps and detergents present in the wastewater (Rosso et al., 2006). Because of their amphiphilic nature, they accumulate at the air-liquid interface of rising bubbles, reducing the mass transfer of oxygen to the liquid [21]. The complex and frequently poorly defined mixture of surface-active compounds in the wastewater [22] makes it difficult to address the specific compounds which are responsible for the reduced alpha factor. A lot of research used the influent organics as a proxy for surface-active compounds to express the impact on the alpha factor [23–28]. Ahmed et al. (2021) were the first to investigate the real-time impact of the degradation of soluble organics on the increase of the alpha factor for an activated sludge batch plant. For the aerobic granule sludge process such an assessment has not yet been performed. Moreover, the influence of process conditions on the observed dynamic behaviour of the alpha factor is still unclear.

This research presents the first long term measurement campaign of the dynamic alpha factor in an aerobic granular sludge batch reactor. Off-gas data of 175 cycles were analyzed to investigate the explicit influence of the alpha factor on the oxygen transfer efficiency. The relation between the dynamic behaviour of the alpha factor and different process conditions (e.g. influent load, exchange ratio and temperature) was studied for the first time for an aerobic granular sludge plant. The impact of the degradation of soluble organics on the increase of the alpha factor for an aerobic granular sludge plant was investigated in more detail and theoretically explained by the Langmuir adsorption isotherm concept. Furthermore, a new approach was applied to describe the dynamic behaviour of the alpha factor by a first order relation with a time constant depending on the pollutant load at the start of the aeration phase. Finally, practical implications and further perspectives for the operation of aerobic granular sludge plants are discussed.

2. Materials and methods

2.1. Reactor under study

This research was done using the Prototype Nereda® in Utrecht, the Netherlands. This demo facility is owned by the district water authority Hoogheemraadschap de Stichtse Rijnlanden and is operated by Royal HaskoningDHV as full-scale AGS research facility. Nereda® is the trademark for the aerobic granular sludge technology owned by Royal HaskoningDHV. The reactor under study is preceded by influent screens and a grit and a sand removal chamber. The reactor treats on average 1000 $m^3.d^{-1}$ with respective average concentrations of total organics, total nitrogen and total phosphorus of 688 g $COD{\cdot}m^{-3}$, 67 g $N{\cdot}m^{-3}$ and 5 g $P{\cdot}m^{-3}$ for the period under study. The pH of the reactor varied between 6 and 7 which is expected to not have a major influence on the biological activity [29]. The total suspended solids in the mixed liquor (MLSS) varied between 6.5 and 9.5 g $TSS{\cdot}L^{-1}$ according to winter and summer conditions respectively. On average, 87% of the MLSS had a diameter above 0.2 mm. The microbial community of aerobic granular sludge consist mainly of relatively slow-growing bacteria such as AOB, NOB, and PAOs due to the high SRT of the granules [30]. The total reactor volume was 1050 m^3 (7.0 m process water depth and 150 m^2 surface). The bottom of the reactor is covered with fine-bubble diffusers. The aeration system consists of two positive displacement blowers of maximum 400 $m^3.h^{-1}$ each, working at 30 kW (Aerzen Blower Model GM 10-SG5/DN80).

The reactor was monitored for liquid and gas phase concentrations during the summer and winter period of 2020-2021 (exact periods are given in S.I. Table S1). The 175 batch cycles in which the off-gas analyser was active and exchange ratio met the predefined value (20, 25 or 30%, see further) were used to calculate the alpha factor over the aeration phase of every cycle. A typical reactor cycle is represented in Table 1. First, simultaneous upward feeding and discharge takes place during a time interval ΔT_{feed} (h), determined by the predefined exchange ratio (ER), which is the ratio of influent volume added to the reactor in a given batch, V_{batch} (m^3), over the total volume of the reactor, $V_{reactor}$ (m^3) (Eq. 2). V_{batch} is determined by ΔT_{feed} and a variable influent flow rate of municipal wastewater, Q_{in} ($m^3\ h^{-1}$). For the aerobic granular sludge reactor under study, the exchange ratio was fixed but the set value was changed over time (Table S1).

$$ER = \frac{V_{batch}}{V_{reactor}} = \frac{Q_{in}\ \Delta T_{feed}}{V_{reactor}} \quad \text{Eq. 2}$$

Next, intermittent aeration pulses are applied which mixes the influent with the remaining nitrate from the previous batch cycle to enhance denitrification. In the subsequent aeration phase a set-point of 1 mg $O_2.L^{-1}$ was maintained until almost complete nitrification and phosphate uptake (corresponding with a water quality of 3 mg NH_4^+-N.L^{-1} and 1 mg PO_4^{3-}-P.L^{-1}) was achieved. Subsequently, denitrification of the remaining nitrate was stimulated by turning off the aeration, except from intermittent pulses to keep the reactor contents mixed. The cycle ends with a settling period, during which also sludge is withdrawn. The variable length of the different phases makes that the total cycle length varies with the pollutant load conditions. Specifications on the applied ER, the influent flow rate, length of feeding, aeration, total cycle time and temperature of the 175 cycles under study are given in S.I. (Table S1).

Table 1. Typical operation cycle for the period under study.

PHASE	**Feeding and discharge**	**Intermittent aeration**	**Aeration**	**Intermittent aeration**	**Settling and sludge discharge**
DURATION	$\Delta T_{feed} = \frac{V_{reactor} \cdot ER}{Q_{in}}$	Variable	Variable	Variable	Fixed
CONVERSIONS	P release, COD storage, denitrification	Denitrification, COD removal	COD removal, nitrification, denitrificati	Denitrification, COD removal	

			on and P uptake		

2.2. Liquid phase measurements

Standard on-line monitoring data of the liquid phase were used to supplement the off-gas analysis. The liquid phase temperature (°C) (LDO, Hach), dissolved oxygen (g $O_2.m^{-3}$) (LDO, Hach), ammonium (g $N.m^{-3}$) (Amtax, Hach), nitrate plus nitrite (g $N.m^{-3}$) (Nitratax, Hach) and phosphate concentration (g $P.m^{-3}$) (Phosphax, Hach) were monitored. A delay of 20 minutes between analysing and detecting NH_4^+ and PO_4^{3-} concentrations was taken into account.

Soluble organic carbon (COD_S) was measured during the aeration phase of one specific cycle on 18 December 2020 at an exchange ratio of 50%. To measure COD_S, the samples were filtered through paper with pore sizes of 4-7 μm (Whatman Schleider & Schuell folded filter paper 595 1/2). A duplicate sample was taken every hour. During the first hour of aeration more samples were taken, three at the first 30 minutes and three at the first hour. The biodegradable COD_S fraction ($bCOD_S$) at each time instant was determined by subtracting the effluent concentration of COD_S (assumed to be the inert COD_S concentration) from measured total COD_S concentration at that time. This implicitly assumes that the effluent concentration of COD_S is inert, which is reasonable given by the long SRT, the plug flow regime in the reactor and full ammonium oxidation.

2.3. Gas phase measurements

A floating free-moving hood was placed on the water surface as in Baeten et al. (2021b). The hood had an area of 0.55 m^2 to collect off-gas from the 150 m^2 water surface. Even though the sampled surface area could be less than the 2% described by the ASCE 18-18 standard for oxygen transfer testing, it was deemed sufficient in this study, given the focus on dynamics and influencing factors rather than exact quantification. Moreover, aerobic granular sludge reactors are characterized by a uniform air distribution system at the bottom, while the same type of conversions take place throughout the reactor, all of which makes that less spatial variations over the water surface are occurring than for activated sludge systems, which are characterized by stronger spatial variations. The dead volume inside the hood was reduced with polyurethane foam in order to reduce the gas residence time inside the hood. Off-gas was sampled from the hood and sent through a cooler to dry the gas before entering an on-line analyser to measure the mole fractions of oxygen using specific paramagnetic sensors (NGA 2000 MLT1 by Rosemount, Emerson). A correction for time delay of 1.5 min was applied for the off-gas sampling (Fig. S1).

The on-site atmosphere was analysed for 5 minutes every hour to account for changes in the composition of the aeration air. The atmospheric temperature, atmospheric pressure and relative humidity were monitored (Bosch BME280). Because of the short tubing for the atmospheric sampling, time delays were neglected. During 25 December 2020 till 9 January 2021 and 22 January 2021 till 25 January 2021, the on-site atmosphere analyser was not working and thus data of the weather station nearby (de Bilt) was used [32]. The airflow rate of the blower was estimated based on the measured rotational speed of blower 1 and 2 (Eq. S1). The experimental set-up and measured variables are visualised in Figure S2.

2.4. Calculation of αF

The gas-liquid mass transfer coefficient $\mathrm{k_La_{O_2}}$ describes the transfer of oxygen from the gas phase to the liquid phase in process water. These $\mathrm{k_La_{O_2}}$ values were likely obtained under fouling conditions as the diffusers were about three years in operation since last cleaning [33]. The presence of contaminants, biomass and diffuser fouling causes a deviation in the gas-liquid transfer coefficient from the clean-water performance $\mathrm{k_La_{O_2,clean}}$. The ratio of $\mathrm{k_La_{O_2}}$ over $\mathrm{k_La_{O_2,clean}}$ is defined as αF (Eq. 3), combining the effect of the alpha factor (α) and the fouling factor (F). $k_La_{O_2,clean}$, which was determined for a temperature of 20°C, was corrected for the same liquid temperature of $k_La_{O_2}$(Eq. S6).

$$\alpha F = \frac{k_La_{O_2}}{k_La_{O_2,clean}} \qquad \text{Eq. 3}$$

Given that fouling is a slow phenomenon, it was reasonably assumed that F remained constant over the time period of this study, so changes in as αF could be attributed to changes in α. A beta factor of 0.95 was determined to correct for the effect of the dissolved solids concentration on the saturation concentration of oxygen [34].

The gas-liquid mass transfer coefficient of O_2 in clean water, $k_La_{O_2,clean}$ (h^{-1}) was determined on 13 December 2016 according to the standard protocol DWA-M 209, 2007 of the German Association for Water, Wastewater and Waste and is 6.16 ± 0.05 h^{-1} at 20°C. This value was obtained at the maximum aeration capacity of 800 $m^3.h^{-1}$ in absence of biomass, after cleaning the diffusers and with effluent water. To correct for the effluent water conditions and thus to derive $k_La_{O_2,clean}$, clean water oxygenation capacity measurements were performed in a separate 500 L water column and compared with the effluent water measurements. αF was calculated at all times at the same maximum airflow rate of 800 $m^3.h^{-1}$ (Fig. S3).

The gas-liquid mass transfer coefficient of O_2 in process water, $k_La_{O_2}$(h^{-1}) was calculated from the dissolved oxygen concentration and gas phase measurements. The calculation procedure, described by Baeten et al. (2021), is given in S.I. section S.1.1.3.

Calculation of αF, based on $k_La_{O_2}$ and $k_La_{O_2,clean}$, for the 18th of December, when COD_S measurements were performed, was done for the total aeration phase length as this specific cycle was operated continuously at the maximal aeration capacity (Fig. S4).

Finally, the smooth function '*smoothdata*' with method '*movmedian*' in MATLAB was used for all cycles studied to smooth the response data of αF and filter outliers in which a moving median of the αF data within a fixed window length of 10 datapoints (± 5 min of monitoring) was calculated (Fig. S5).

3. Results and discussion

3.1. Dynamics of alpha over a cycle

The alpha factor, αF in this study, characterizes the oxygen transfer efficiency and was calculated for 175 cycles. The aeration phase length differed for each cycle, because of varying loads fed to the reactor. The aeration was switched off once the effluent quality was reached, namely 3 mg NH_4^+-N.L^{-1} and 1 mg PO_4^{3-} -P.L^{-1}.

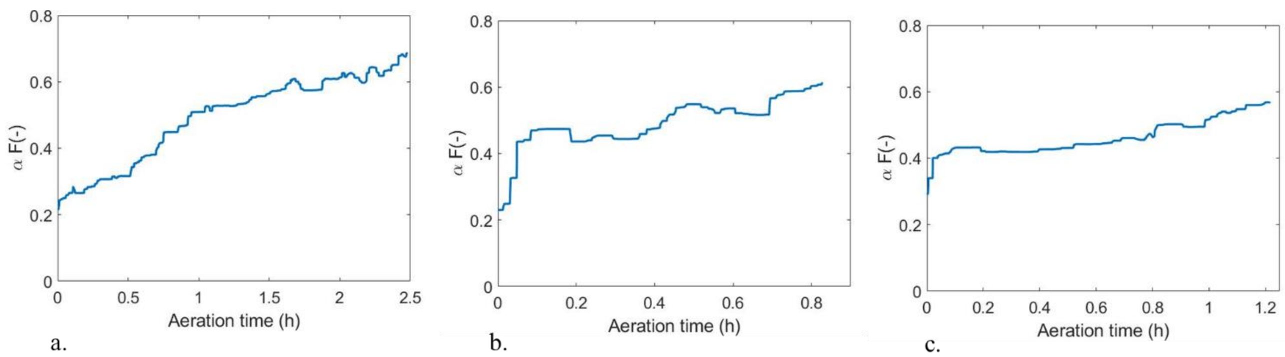

Figure 1. The increase of αF over the aeration time (at 100% aeration capacity) on (a) 22 June 2020, (b) 1 December 2020 and (c) 9 January 2021 (k_La_{O2} = 6.16 h^{-1}).

The dynamics of αF for three example cycles are displayed in Figure 1. The value of αF increased during the aeration phase. The increase in the oxygen transfer efficiency over the aeration phase in an aerobic granular sludge reactor is in agreement with Baeten et al. (2021) and Cecconi et al. (2020), who also observed an increase in the k_La_{O2} and the oxygen transfer efficiency, respectively. They related their observations to a presumed decrease in surface active compound concentrations over the batch cycle. The αF profiles determined in this study indicate an explicit influence of αF on the oxygen transfer efficiency. Furthermore, the rate of increase in αF over the aeration phase varied, as did the pattern of αF itself. This could be attributed to different concentrations or types of surface active agents at different days.

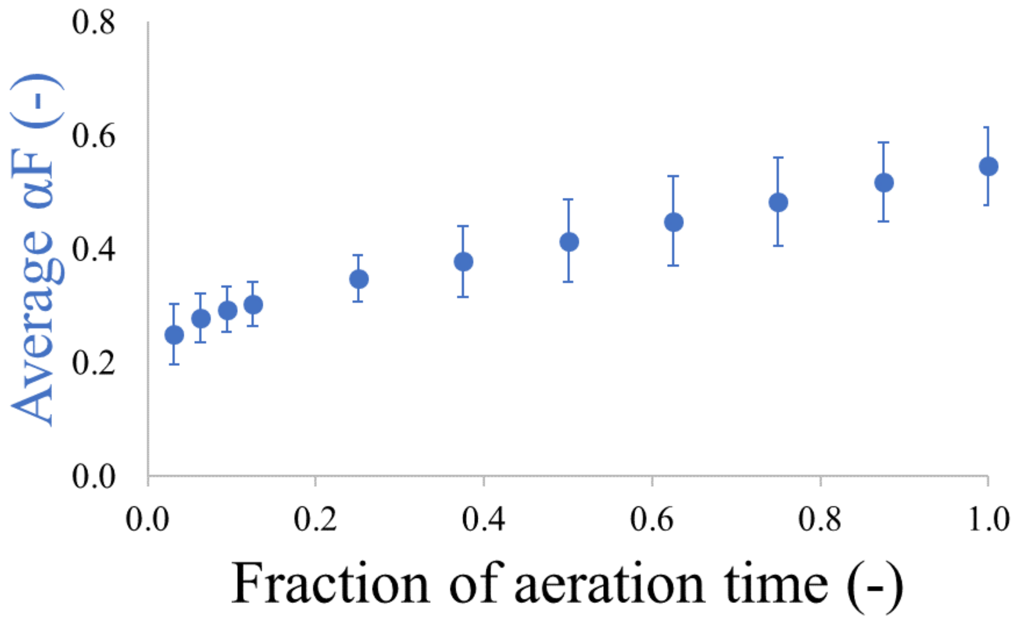

Figure 2. The average αF (at 100% aeration capacity) of 175 cycles with standard deviation over the fraction of the aeration time ($\frac{t}{T_{aeration}}$). Note that this average dynamic behaviour is specific for the installation under study.

The dynamic behaviour of αF was averaged out over the 175 cycles (Fig. 2, calculation detailed in S.I. section S.1.1.4.) and expressed as a function of the aeration time fraction (aeration time over total cycle time) to compensate for the different aeration phase length for each cycle. The duration of the aeration phase at 100% aeration capacity was 1h 25 minutes on average, with a minimum of 30 minutes and a maximum of 3h 30 minutes. The initial αF was on average 0.25 and increased up to 0.55 on average. The values of αF at the beginning (αF_{ini}) and the end of the aeration phase (αF_{end}) appeared quite independent of the aeration phase length and therefore independent of the variable influent concentrations. This observation was substantiated by investigating the αF_{ini} and αF_{end} for the variable influent ammonium concentrations of the 175 cycles (Fig. S9). This seemed to indicate a constant effect of surface-active compounds at the beginning and at the end of the aeration period.

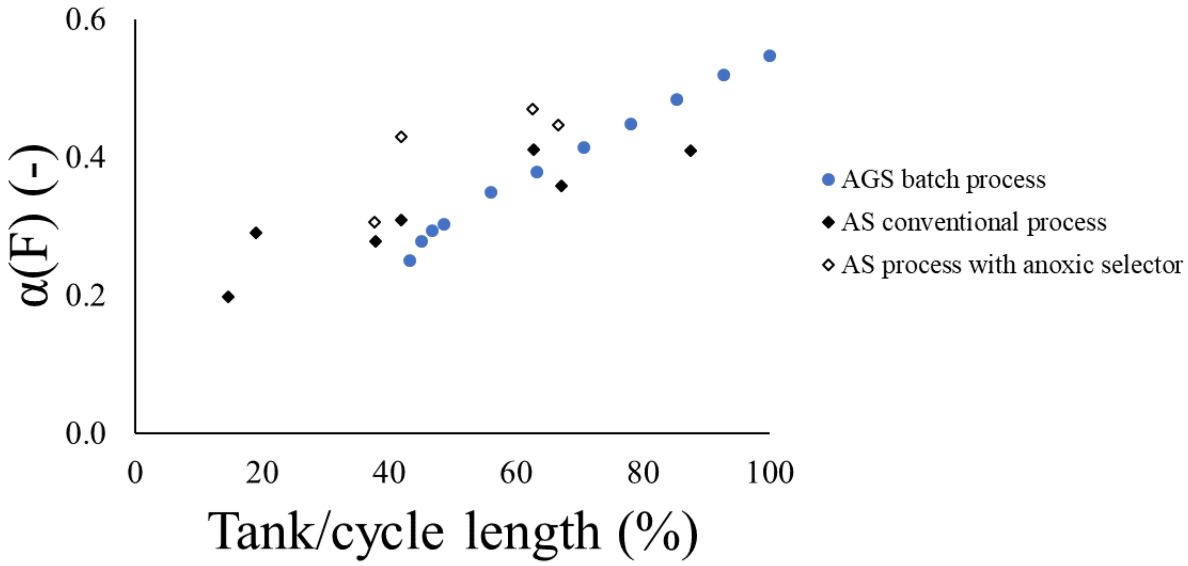

Figure 3. The average αF of 175 cycles over the fraction of the cycle length (%), including feeding and aeration phase, of an aerobic granular sludge (AGS) batch reactor compared to α over the fraction of the aerobic tank length (%) of a continuous activated sludge (AS) plug flow reactor with and without anoxic selector (data obtained from Rosso and Stenstrom (2007)).

Rosso and Stenstrom (2007) demonstrated an increasing oxygen transfer efficiency (α value) along the length of an activated sludge tank, both with and without anoxic selector. The few data points for continuous plug flow reactors with activated sludge from Rosso and Stenstrom (2007) are compared with the measurements of 175 cycles for a batch reactor with aerobic granular sludge obtained in this study (Fig. 3).

The tank length of a continuous plug flow reactor relates to the cycle length (feeding phase and aeration phase) of a batch reactor. The anaerobic feeding phase of an aerobic granular sludge batch reactor shows an analogy with an anoxic selector for activated sludge, which removes the readily biodegradable organic carbon (rbCOD) before entering the aerobic stage [35] thereby reducing the rbCOD accumulation at bubble surfaces [36,37] and thus increasing the alpha factor at the entrance of the aerobic stage. The initial αF of the aerobic granular sludge batch reactor (0.25 at the start of the aeration phase) in our study was lower than the initial α value of the continuous plug flow reactor with anoxic selector (about 0.3 at the entrance of the aerobic reactor) examined by Rosso and Stenstrom (2006b). This could be attributed to the fact that the α data of Rosso and Stenstrom (2007) originated from off-gas tests of new or recently cleaned diffusers, while the αF data of the aerobic granular sludge reactor under study incorporated fouled diffusers (F-factor < 1, exact value not determined).

The αF (or α) values of both systems displayed in Figure 3 increase along the tank or cycle length. The increase in alpha is higher for the batch-wise operated aerobic granular sludge reactor than for the continuous reactor with activated sludge, causing a higher end value for the aerobic granular sludge reactor. Cleaning the diffusers of the aerobic granular sludge reactor under study (F-factor = 1), would even further increase the values of the alpha factor compared to the one of the activated sludge reactor. Differences in alpha factor can be attributed to differences in the type of wastewater treated and in the prevailing SRT and MLSS concentrations between both systems, as well as to the process configuration as such, which makes that care must be taken not to over-interpret the comparison between both systems.

3.2. Relation of alpha with exchange ratio and temperature

As the exchange ratio determines the initial surface-active compound concentration in the aeration phase, the effect of exchange ratio on the profile of αF over the aeration phase was investigated for the 175 cycles under study (Fig. 4). A higher exchange ratio resulted in a longer aeration phase length, due to the higher pollutant load. The exchange ratio did not have a relevant effect on the average initial value of αF (αF_{ini} = 0.25, 0.23 and 0.24 for respectively an

ER of 20, 25 and 30%), neither on the average end value of αF (αF_{end} = 0.54, 0.52 and 0.58 for respectively an ER of 20, 25 and 30%), meaning that the exchange ratio influences the increase rate of αF. A higher exchange ratio implied a lower rate of increase in αF.

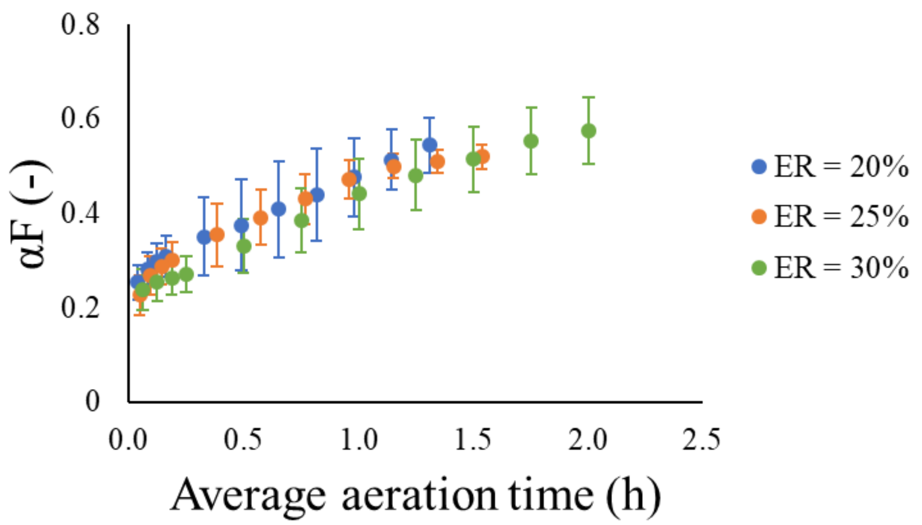

Figure 4. αF over the average aeration time (at 100% aeration capacity) for an exchange ratio (ER) of 20, 25, 30% of the 175 cycles.

As a result, the exchange ratio could be used as a manipulated variable to improve the rate of increase of αF and the related oxygen transfer efficiency. While manipulating the exchange ratio, it is also needed to take into account the incoming wastewater flow rate to be handled, the selected cycle time and other boundary conditions, e.g. respecting a maximum practical ER of 65% and keeping a maximum liquid upflow velocity of 5 $m.h^{-1}$ [38]. Moreover, the pollutant load and thus αF are not only dependent on the exchange ratio, but also on the wastewater strength. In practice, wastewater flow rate and pollutant concentration are correlated: rain weather is typically characterized by a high flow rate and low concentrations whereas dry weather conditions imply a relatively low flow rate and high concentrations. The optimal exchange ratio and cycle time to be applied in practice should consider all of these effects.

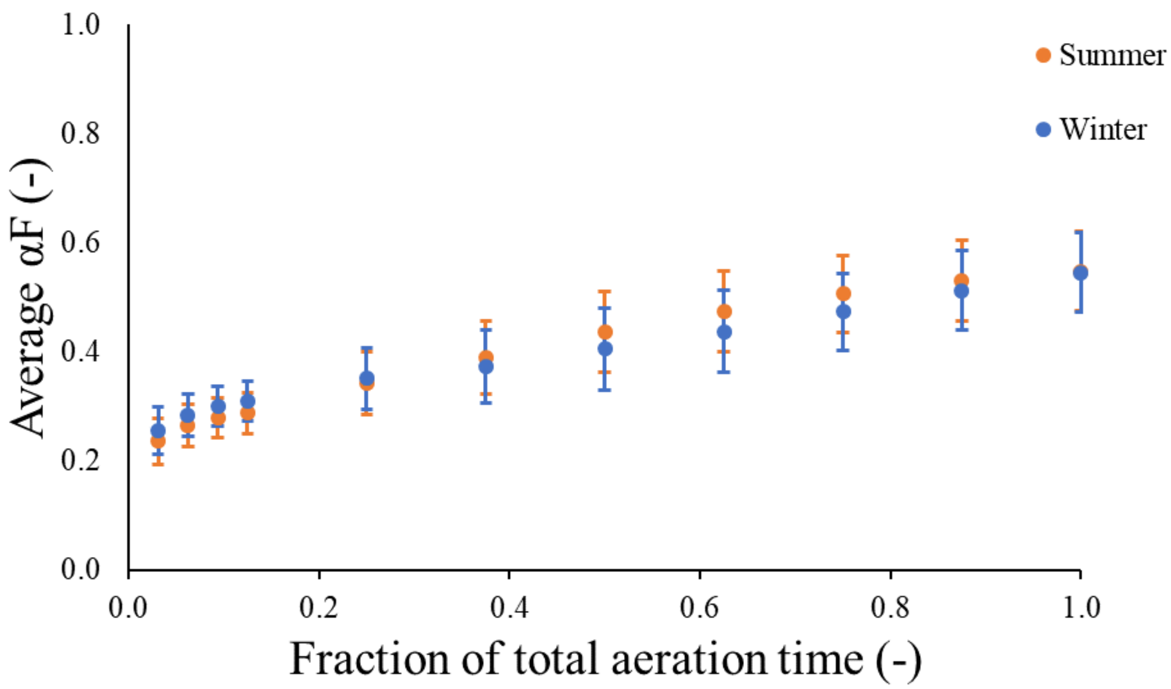

Figure 5. The average αF (at 100% aeration capacity) with standard deviation of 56 cycles during June-July 2020 ($T_{average}$ = 22°C) and of 124 cycles during December 2020-January 2021 ($T_{average}$ = 14°C) over the fraction of the aeration time ($\frac{t}{T_{aeration}}$).

The effect of temperature on the dynamic behaviour of αF is reflected in Figure 5. Temperature had no relevant effect on the average initial (αF_{ini} = 0.24 and 0.26 for respectively summer and winter) or final value (αF_{end} = 0.55 for both summer and winter) of the alpha factor. It was expected that during summer more surface-active compounds would be hydrolyzed in the sewer system and that more compounds would be taken up during the anaerobic feeding time, causing a lower surface-active compound concentration at the beginning of the aeration phase. However this did not seem to impact αF_{ini} .

In summer, a faster increase of αF compared to winter was observed. This could be due to the higher hydrolysis rates at higher temperature and/or due to the higher MLSS concentration (7.5 and 9.5 g $TSS{\cdot}L^{-1}$ for winter and summer respectively). Therefore, it took less time during summer to reach αF_{end} and thus to reach the most efficient oxygen transfer. The fact that the same αF_{end} is reached during winter and summer indicates that the removal of surface-active compounds had always stabilized by the end of the aeration phase, i.e., before the effluent criteria for N and P were met.

3.3. Relation of alpha with soluble biodegradable organic carbon and ammonium

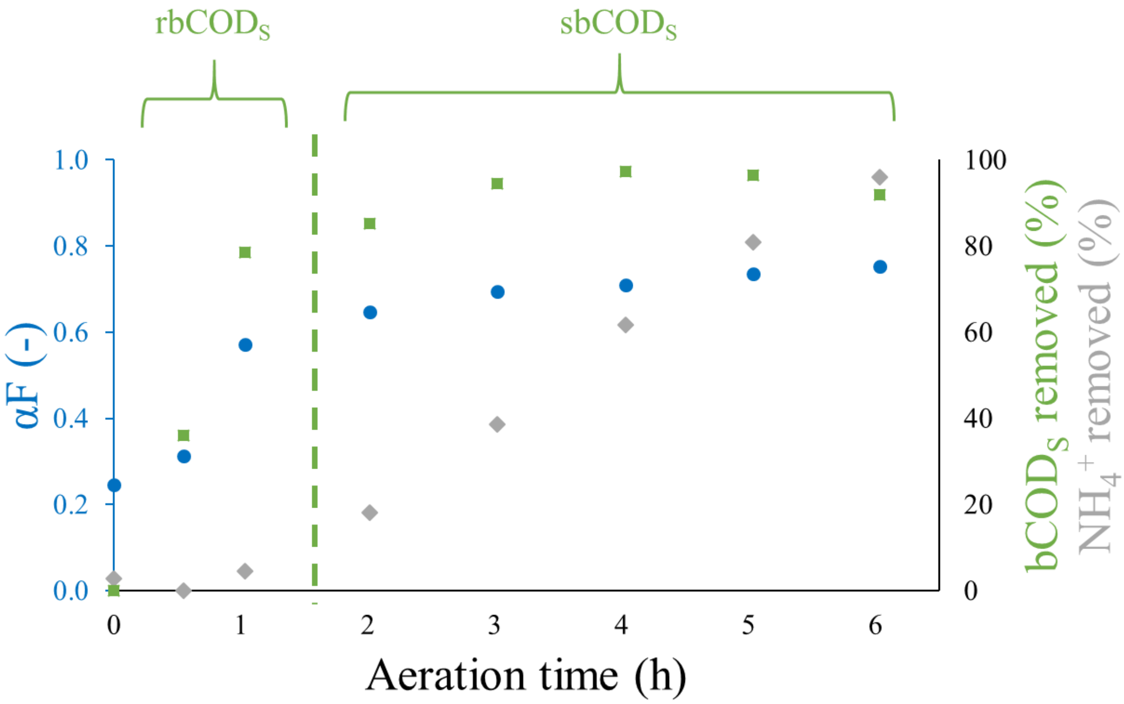

Figure 6. αF, $bCOD_S$ and NH_4^+ removed (%) over the aeration time of a single cycle (18 December 2020).

Detailed investigation of a single cycle (Fig. 6) showed that the increasing pattern of αF was very similar to the removal efficiency of soluble biodegradable organic carbon ($bCOD_S$), at least after one hour of aeration. While during the first hour and a half of the aeration phase a large amount of $bCOD_S$ was removed for a relatively small increase in αF (80% of all $bCOD_S$ was removed while αF increased from about 0.25 to 0.57, or $\frac{\Delta \alpha F}{\Delta bCOD_S} = \frac{0.32}{0.8} = 0.4$), less $bCOD_S$ was removed for a relatively higher increase in αF during the following five hours of aeration (remaining 20% of $bCOD_S$ while αF increased from about 0.57 to 0.72, or $\frac{\Delta \alpha F}{\Delta bCOD_S} = \frac{0.15}{0.2} = 0.8$). It could be expected that the fast $bCOD_S$ removal during the first hour of aeration mainly concerned soluble readily biodegradable organic carbon ($rbCOD_S$) while afterwards soluble slowly biodegradable organic carbon ($sbCOD_S$) was removed, at a slower rate. This suggests that αF increases more during the removal of certain amount of $sbCOD_S$ than during the removal of the same amount of $rbCOD_S$. Note that the end value of αF for this specific cycle (αF_{end} = 0.72) was remarkable higher than the one which was found for the other 175 cycles (αF_{end} = 0.55). This is likely due to the fact that this specific cycle was operated continuously at the maximal aeration capacity, allowing a higher dissolved oxygen concentration, which could have resulted in the removal of additional recalcitrant surface-active compounds.

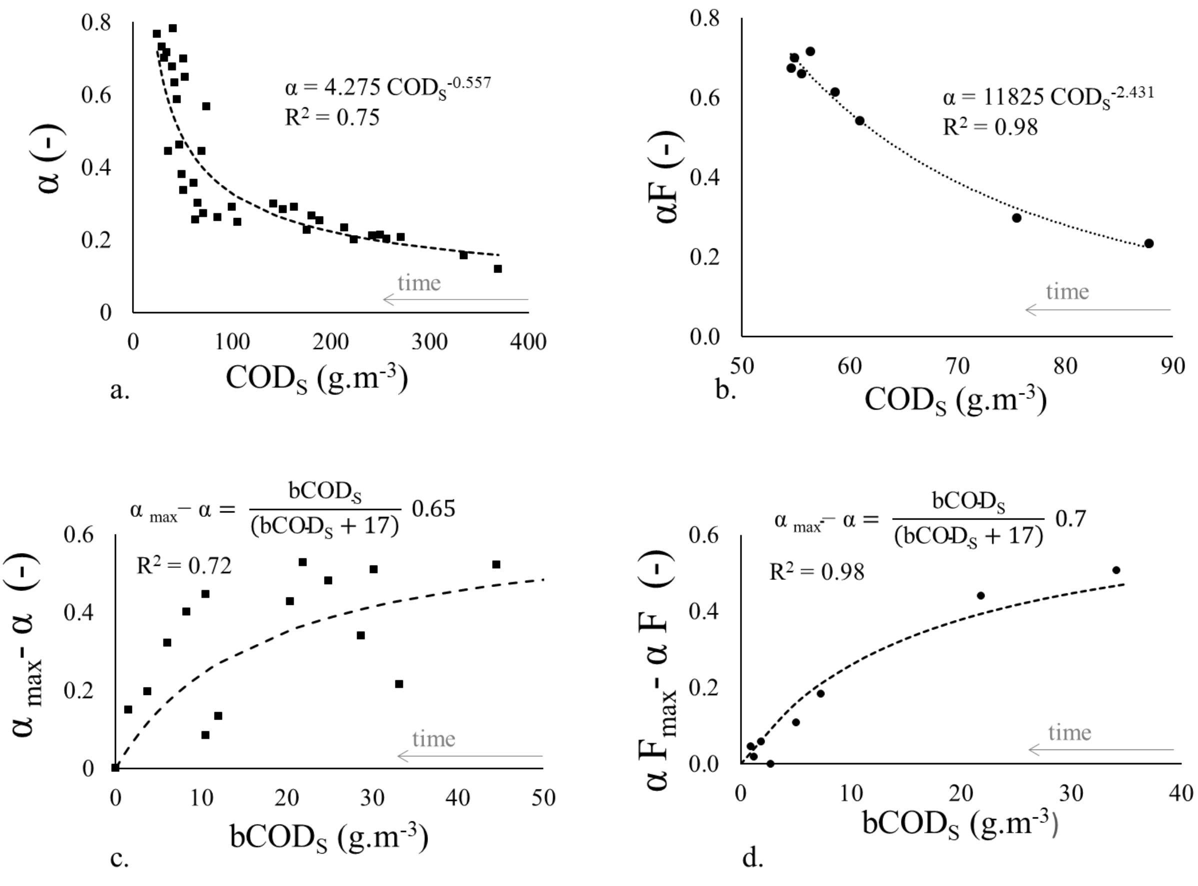

Figure 7. Relation between the alpha factor and soluble organic carbon (COD_S, $mg.L^{-1}$) (a) for 65 samples during the aeration time of an activated sludge batch reactor (data obtained from Ahmed et al. 2021) (b) for a single cycle (18 December 2020) during the aeration time of an aerobic granular sludge batch reactor. Adsorption isotherms of soluble biodegradable organic carbon ($bCOD_S$) with $\alpha(F)_{max}$-$\alpha(F)$ referring to the adsorption capacity of the air bubbles (c) based on data of Ahmed et al. (2021) and (d) data of the single cycle at 18 December 2020 in this study.

Ahmed et al. (2021) investigated the impact of biodegradation of soluble organic carbon (COD_S) on the alpha factor during the aeration time of an activated sludge batch reactor and showed a negative relation between alpha and COD_S ($R^2 = 0.75$) (Fig. 7a). Although this study was only based on one specific cycle, a similar decreasing exponential relationship between αF and COD_S was found according to Eq. 4 (Fig. 7b).

$$\alpha F = A\ COD_S{}^{-K} \qquad \text{Eq. 4}$$

where A and K are fitting parameters specific for the installation under study (Fig. 7a and b).

The obtained decreasing exponential relations reflect that, as more organic compounds are degraded (i.e., as time progresses), there is a relatively higher increase in the alpha factor for a given amount of organic compounds degraded. However, it is physically incorrect that alpha is infinite for a $COD_S = 0$ $mg.L^{-1}$ which makes that the model should be adapted to ensure realistic end alpha values. The different fitting parameters obtained in both studies (Fig. 7a and b) may be attributed to the different process configuration and operation, as well as to different soluble organic carbon fractionation ($sbCOD_S$ versus $rbCOD_S$). It could further be noted that COD_S comprises both soluble biodegradable and inert organic carbon. However, the inert fraction did not seem to have an effect since the final αF^* was about the same for all 175 cycles (with varying influent and consequently soluble inert organic carbon concentrations) (Fig. 2). As a result, instead of expressing the relationship between αF and soluble organic carbon in terms of total (= biodegradable + inert) soluble organic carbon (Fig. 7a and b), one could also opt to relate it to soluble biodegradable organic carbon (Fig. S10).

No distinct relation was found between αF and the amount of ammonium removed (Fig. 6). Nitrification started after one hour, which seems to coincide with the complete removal of $rbCOD_S$ or the moment when the $sbCOD_S$ started to be converted. The delayed nitrogen removal is attributed to oxygen limitation which is typical for installations with limited aeration capacity (under dimensioning), as was the case for the reactor under study and also observed in other studies (e.g. Ahmed et al. 2021). Still, the established relations between the alpha factor and COD_S indicate that oxygen limitation of nitrification at the start of the aeration phase is not only due to the oxygen demand associated with high initial $rbCOD_S$ concentrations, but also to a lower oxygen transfer efficiency (lower alpha factor).

3.4. Analogy to Langmuir adsorption isotherms

The relatively lower increase rate of the alpha factor at the start of the aeration phase was further investigated. The Langmuir adsorption isotherm describing the adsorption of solutes on interphases might be an interesting equation to describe the effect of $rbCOD_S$ on the alpha factor (Eq. 5).

$$\Gamma_{\mathrm{ads}} = \frac{\mathrm{bCOD_S}}{(\mathrm{bCOD_S}+\mathrm{K_{eq}})}\,\Gamma_{\mathrm{ads}}^{\mathrm{max}} \qquad \text{Eq. 5}$$

in which Γ_{ads} is the amount of surface-active compounds adsorbed, i.e., the occupancy (mg surface-active compounds.m^{-2} air bubble), $\mathrm{K_{eq}}$ is the Langmuir equilibrium constant (mg.L^{-1}) and Γ_{ads}^{max} is the (maximum) adsorption capacity (mg surface active compounds.m^{-2} air bubble). The Langmuir adsorption saturation behaviour is explained by a critical micelle concentration ($bCOD_S$ for $\Gamma_{ads} = \Gamma_{ads}^{max}$) which is the concentration of surface-active compounds above which all additional surface-active compounds added to the system will form micelles [39]. At $bCOD_S$ concentrations above the critical micelle concentration the adsorption capacity (and surface tension) remains relatively constant. At $bCOD_S$ concentrations below the critical micelle concentration, the adsorption capacity has rather a linear course.

We assumed the deviation from the maximum αF, namely αF_{end} in this study, as proportional to the amount of adsorbed surface-active compounds, Γ_{ads} (Eq. 5). The lowest value of Γ_{ads} is then obtained when the adsorption of surface-active compounds on the bubble surface area reached its minimum and when αF was the highest ($\alpha F = \alpha F_{end}$), which is at the end of the aeration phase. In the beginning of the aeration phase, the adsorption of surface-active compounds on the air bubbles was the highest and αF was the lowest ($\alpha F = \alpha F_{ini}$).

$$\alpha F_{end} - \alpha F = \frac{bCOD_S}{(bCOD_S + K_{eq})}(\alpha F_{end} - \alpha F_{ini}) \qquad \text{Eq. 6}$$

Eq. 6 was applied to the data of Ahmed et al. (2021), as well as the data of our study (Fig. 7c and d respectively), expressing the deviation from αF_{end} as a function of the $bCOD_S$ concentration. In the beginning of the aeration phase, at high $bCOD_S$ concentrations, a type of adsorption saturation behaviour was observed, causing a relatively large decrease in $bCOD_S$ concentrations to result in a relatively small change in alpha. This could be interpreted as an (almost) full occupation of the adsorption sites, for a $bCOD_S$ concentration higher than the critical micelle concentration. As time progressed, lower $bCOD_S$ concentrations were reached, causing a more rapid increase in alpha with decreasing $bCOD_S$ concentrations. Indeed, as more organic compounds were degraded, the critical micelle concentration was reached causing a higher increase in alpha, explaining why alpha increases more at the end of the process.

The $bCOD_S$ concentrations studied by Ahmed et al. (2021) were remarkable higher than the ones in our study, and that the alpha factor in the former study was relatively constant for a longer time upon the start of the aeration phase (Fig. S11). This could be explained by the $bCOD_S$ concentrations in the study of Ahmed et al. (2021) to be clearly higher than the critical micelle concentration. The $bCOD_S$ concentrations in our study are hypothesized to be closer to the critical micelle concentrations, leading to a faster decrease in adsorption of surface-active compounds on the air bubbles and corresponding steeper increase in the alpha factor with time.

3.5. First order dynamics of alpha – relation with pollutant load

The average dynamic behaviour of αF shows an increase over time, from a constant αF_{ini} to a constant αF_{end} (Fig. 2). Apart from the established relation between αF and $bCOD_S$, the dynamic behaviour of aF could also be described by a first order process characterized by a gain ($K = \alpha F_{end} - \alpha F_{ini}$) and a time constant, τ (h) (Eq. 7). The time constant, τ, is defined as the aeration time at which 63.2% of the end value (αF_{end} - αF_{ini}) is reached (S.I. section 1.2.5.).

$$\alpha F(t) = (\alpha F_{end} - \alpha F_{ini}) \cdot \left(1 - e^{\frac{-t}{\tau}}\right) + \alpha F_{ini} \qquad \text{Eq.7}$$

Given that all aeration cycles were characterized by the same αF_{ini} (= 0.25) and the same αF_{end} (= 0.55), the gain was determined as $K = \alpha F_{end} - \alpha F_{ini}$ = 0.3 specific for the installation under study. As a result, only the value of the time constant, τ, remains to be known for a full characterization of the αF dynamics.

αF was shown to be related to the concentration of soluble biodegradable organic carbon ($bCOD_S$) (Fig. 7). However, measuring the $bCOD_S$ concentration is not standard practice during the operation of a full-scale aerobic granular sludge plant. On the other hand, nutrient sensors are installed by default but no relationship between αF and NH_4^+ was observed (Fig. 6). It was assumed that the rate of increase in αF, characterized by τ, is related to the soluble biodegradable organic carbon concentration at the start of the aeration phase ($bCOD_{S,ini}$). As this concentration is not typically measured, other relations were sought.

Even though different conversion dynamics exist between the removal of $bCOD_S$ and NH_4^+, the influent wastewater composition is typically characterized by constant average ratios [40], in this case an average COD/TN ratio of 10. It was assumed that the ratio between $bCOD_S$ and NH_4^+ at the beginning of the aeration phase ($bCOD_{S,ini}$ and $NH_4^+{}_{ini}$ respectively) was constant as well. As $bCOD_{S,ini}$ is related to the time constant τ, it was expected that $NH_4^+{}_{ini}$ would also be related to τ. However, only a poor correlation between $NH_4^+{}_{ini}$ and the time constant τ was found (Fig. S13). This is explained by the different fractionation of $bCOD_{S,ini}$ per cycle, the effect of temperature on the kinetics of ammonium removal [41] and the adsorption of the influent NH_4^+ to the biomass [42], all influencing the ratio between the measured $NH_4^+{}_{ini}$ and $bCOD_{S,ini}$.

The phosphate peak at the beginning of the aeration phase ($PO_4^{3-}{}_{ini}$) is proportional to the uptake of soluble readily biodegradable organic carbon ($rbCOD_S$) in the anaerobic feeding phase prior to the aeration phase [43]. It was assumed that a higher $rbCOD_S$ uptake corresponded to a higher $bCOD_{S,ini}$ and on its turn to a larger time constant τ. No strong relationship between $PO_4^{3-}{}_{ini}$ and the time constant τ was found (Fig. S13 and S14).

The use of a first order relation with a fixed gain and a time constant depending on several process conditions is a novel and physically more correct way to describe the dynamic behaviour of αF compared to the one suggested by Ahmed et al. (2021) (Eq. 4). It is important to note that different installations will have other quantitative values for αF_{ini} and αF_{end} and as a result a different gain. The potential of proxies for the initial $bCOD_S$ concentration, reflected in the time constant needs further investigation. Overall, it is theorized that higher loads (~

higher $NH_4^+{}_{ini}$, $PO_4^{3-}{}_{ini}$) are related to a higher time constant, meaning that the time to reach αF_{end} will be longer. In contrast to the relation between αF and $bCOD_S$, the first order relation can only be established by first studying the dynamic behaviour of αF over a longer period of time to find a proxy for the $bCOD_S$ concentration. Afterwards, the first order relation could be part of a control strategy to select for the most optimal process performance depending on the incoming load of the specific aerobic granular sludge plant.

3.6. Practical implications and perspectives

The focus of this study was the dynamics of the alpha factor, influencing the oxygen transfer efficiency, in a batch-wise operated granular sludge reactor. The increase of alpha over the aeration phase was in this study attributed to the breakdown of surface-active compounds. Besides surface-active compounds, it is known that the alpha factor is affected by a wide range of other potential components and conditions in different ways and magnitudes [44]. These other components and conditions are part of the reactor design, such as diffuser type, distribution and tank depth [37] as well as related to operational aspects such as fouling [33], SRT [45], MLSS concentration and rheology [46], microbial activity [47] and soluble microbial metabolites. As these components are assumed to stay constant over a short term period, they were not taken into account as they will not likely influence the increase of alpha over the aeration phase of a specific cycle.

Reactor design and operational conditions are expected to influence the dynamic behaviour of the alpha factor. Even though the observed qualitative behaviour may be interpreted as generally valid, the numerical results are installation-specific. The initial value of alpha, found to be constant for the installation under study, will be numerically different for other installations and be determined by the origin of the incoming wastewater and thus the load of surface-active compounds, the degree of fouling of diffusers and the MLSS concentration and rheology. A different MLSS concentration and rheology can influence the bubble coalescence. The rheology of the sludge determines the possibility to create larger bubbles which reduce the specific area and thus reduce the alpha factor [27,46,48]. The effect of MLSS concentration and the biomass structure of aerobic granular sludge on the alpha factor is not yet investigated and could be a topic of further research. The numerical value of the alpha factor at the end of the aeration phase, on the other hand, will again be installation- specific and determined by above-mentioned components and conditions, as well as by the applied aeration strategy as was observed in this study. Besides, a faster increase in alpha is expected for a higher aeration capacity. A higher aeration capacity allows a faster increase in oxygen concentration which could compensate for the effects of the alpha factor.

Mathematical models are a powerful tool to evaluate, design, and optimize the operation of aerobic granular sludge plants [49]. Current mechanistic aerobic granular sludge models have however mainly focused on describing the biokinetic processes while the oxygen mass transfer is very simplistic modelled [50]. Indeed, typically constant alpha factors are used in these models [24,51–53]. Even though this is a realistic assumption for well-mixed reactors, it is in conflict with what has been observed in this study and as a consequence, might give wrong interpretation to the process design and optimization of batch-wise operated reactors. A potential improvement of current aerobic granular sludge models is to add a dynamic aeration model by using the relation between alpha and the $bCOD_S$ concentration over the aeration phase length, however the model should be adapted to ensure realistic end alpha values. This will give new insights on how process operation can optimize the oxygen transfer efficiency related to the dynamic behaviour of alpha.

The findings of this study can be seen as a starting point to further investigate new operational strategies that optimizes the oxygen transfer efficiency of an aerobic granular sludge plant. It was shown that a lower $bCOD_S$ concentration at the start of the aeration phase will promote a faster increase in the alpha factor and thus a higher oxygen transfer efficiency. It follows that a possible optimization strategy include the trade-off between the exchange ratio and cycle time to influence the $bCOD_S$ concentration at the start of the aeration phase. Another strategy could be the prolongation of the anaerobic feeding time to decrease the $rbCOD_S$ concentration at the beginning of the aeration phase and thus fasten the increase of the alpha factor. Furthermore, a different aeration strategy, e.g. the use of extra aeration capacity at the start of the aeration phase, could be developed to influence the removal rate of $bCOD_S$ and thus the rate of increase of the alpha factor.

It is also important to take the effect of temperature into account. A different operational strategy between summer and winter may be necessary. On the one hand, lower temperatures negatively impact the microbial removal rates. On the other hand, lower temperatures increase the oxygen saturation concentration and thus the related oxygen transfer efficiency [54]. It is worth investigating which of the two factors will predominate and thus determine the effect on alpha.

It is clear that the alpha factor holds a strong potential for the optimisation of the aeration energy. Although this research was performed in an aerobic granular sludge batch reactor, a large part will be applicable for conventional batch systems with flocculent sludge. In light of the growing awareness on energy efficiency, research efforts are underway globally to reduce the energy consumption of WRRFs. However, it is remarkable that such an important energy aspect as the alpha factor is hardly researched while it is well known to affect the aeration energy significantly. Time has come to further unravel the alpha factor and its practical implications on WRRFs.

4. Conclusions

The oxygen transfer efficiency in a batch-wise operated aerobic granular sludge reactor was scrutinized through the study of the alpha factor during the aeration phase.

- The alpha factor increases over the aeration phase length, analogous to the increase along the length of the aeration tank of a continuous fed plug flow reactor. All aeration cycles were characterized by the same alpha at the beginning of the aeration phase and by the same alpha at the end of the aeration phase cycle, independent of the influent load, exchange ratio and temperature.
- The alpha factor was related to the removal of soluble biodegradable organic carbon ($bCOD_S$). This relation between αF and the $bCOD_S$ concentration could be described by a decreasing exponential function. Alternatively, the inverse relation between $bCOD_S$ and the alpha factor could be explained by a Langmuir adsorption isotherm describing the adsorption of $bCOD_S$ at the air-liquid interface. The constant initial alpha value is in agreement with the associated concept of a maximum adsorption capacity.
- The dynamic behaviour of the alpha factor could be described by a first order relation, with a fixed gain for all cycles under study and a time constant depending on several factors, such as the initial $bCOD_S$ concentration (on its turn determined by the exchange ratio) and the temperature. The potential of proxies for the initial $bCOD_S$ concentration could be further investigated to predict the dynamics of alpha over the aeration phase.

The insights from this study can be used to further optimise the energy efficiency and operation of aerobic granular sludge reactors and other batch-wise operated aerobic wastewater treatment systems.

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