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Increasing oxygen transfer efficiency through sorption enhancing strategies

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ABSTRACT

The link between aeration efficiency and biosorption capacity in water resource recovery facilities was extensively investigated, with special emphasis on wastewater characteristics and the development of strategies to maximize adsorption. Biosorption of oxygen transfer inhibitors (i.e., surfactants, colloidal, and soluble fractions) was examined by a series of pilot batch-scale experiments and full-scale studies. The impact of a sorption-enhancing strategy (i.e., bioaugmentation) deployed at full-scale over a five-year period was evaluated. Bench-scale experiments determined the inhibition coefficient (Ki) to measure the impact of surfactants and COD fractions as inhibitors of oxygen transfer efficiencies (αSOTE) in wastewater systems. The inhibition constant for surfactants Ki was found at 2.4 ± 0.4 mg L^{-1} SDS while for colloidal material was at 14 ± 1 mg L^{-1} (no inhibition for soluble fraction was found). Two enhancing biosorption configurations (i.e., contact stabilization and anaerobic selector) resulted in significant improvements in both aeration efficiency indicators (αSOTE) and surfactants removals. αSOTE improvements of 46% and 54% in comparison to conventional high rate activated sludge process (HRAS) were reported. Similarly, the removal of surfactants was increased by 27% and 56% using optimized enhancing-sorption strategies. Further analyses helped elucidate the underlying mechanisms of surfactants removal. Findings are expected to help full-scale applications increase their sorption potential as well as the concurrent aeration efficiency, which helps WRRFs to advance toward energy-positive wastewater treatments.

1. Introduction

With the objective of achieving energy neutrality for water resource recovery facilities (WRRFs), new technologies and operational strategies aimed to reduce energy demand while meeting effluent requirements are gaining interest. Limiting the oxidation of organics during the aeration process is being welcomed as a strategy to lower aeration costs while increasing carbon harvesting as useable energy, thus improving the overall carbon footprint (Jimenez et al., 2015; Meerburg et al., 2015a; Miller, 2015). This improvement is achieved through the combination of very short solids retention times (SRT) with high sludge-specific organic loading rates, which increases the sorption capacity of activated sludge. Short SRTs limit the oxidation of easily oxidizable organic fractions while capturing colloidal and particulate fractions

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through adsorption into the microbial flocs, which can then be further harvested as biogas resource (Jimenez et al., 2015).

Bioflocculation, or floc production due to extracellular polymeric substances (EPS), is essential in the solids-liquid separation phase during secondary settling (Forster, 1985; Liao et al., 2001; Nielsen et al., 1996; Urbain et al., 1993), but it also plays a key role in determining the adsorption capacity of flocs (More et al., 2014; Rahman et al., 2016; Schuler et al., 2001). New strategies based on maximizing bioflocculation and its concurrent adsorption capacity in high rate activated sludge (HRAS) systems are aimed at overcoming some of the current limitations arising from short SRTs (i.e., bioflocculation) are being developed (Meerburg et al., 2015b; Miller et al., 2017; Rahman et al., 2016; Van Winckel et al., 2019). A few strategies were described to enhance bioflocculation and settling, and were shown to be highly dependent on the loading conditions of the system, indicating that highly loaded systems at lab-scale (>10 kg COD/m^3/day) can enhance bioflocculation (i.e., settling, adsorption capacity, etc.) with an adequate F:M ratio (Rahman et al., 2017b; Van Winckel et al., 2019). On the other hand, low loaded systems benefit from implementing strategic feast-famine configurations (Van Winckel et al., 2019; Winckel et al., 2016). Even if different operational strategies are required to maximize bioflocculation according to the loading conditions of the system, all alternatives seem to indicate that bioflocculation is promoted by sustaining a minimum amount of EPS and, especially, by an adequate quality of the flocs (Mancell-Egala et al., 2016; Sesay et al., 2006). While no correlation between the EPS concentration and bioflocculation has been consistently proven in literature, there is evidence that EPS quality and composition (i.e., content ratio of protein to polysaccharide, glycoprotein content, etc.) is key to the formation of flocs and their adsorption capacity (Durmaz and Sanin, 2001; Liao et al., 2001; Sesay et al., 2006; Yu et al., 2008). Adsorption enhancing strategies can alter floc composition, either by adjusting critical processes (i.e., stabilization phases, promotion of feast-famine regimen, modification of solid retention time, etc.), or by the addition of a selected microbial population with its intrinsic EPS properties (i.e., bioaugmentation).

Bioaugmentation is one of the sorption enhancing strategies that is gaining interest due to its recent consideration as a cost-effective method to increase process stability, settling and enhance flocculation/adsorption capacity and even for being able to lower the required volumes for treatment (Limbergen et al., 1998; Mikhailova et al., 2014; Plaza et al., 2001; Stenström and La Cour Jansen, 2016). Bioaugmentation refers to the addition of specialized strains/mixed cultures (i.e., nitrifying/denitrifying bacteria) to wastewater reactors (Herrero and Stuckey, 2015; Jianlong et al., 2002; Limbergen et al., 1998). A bioaugmentation strategy, consisting of the addition of waste activated sludge returns from the nitrifying/denitrifying reactors performing biological nutrient removal (BNR) to the upstream high-rate activated sludge process (HRAS), was implemented to increase the nitrogen removal and to minimize the cost of external carbon dosing (methanol) for the downstream BNR process (Bailey et al., 2007), where otherwise the short SRT would be limiting nitrification. Improved settling and lower TSS after the implementation of the bioaugmentation strategy was observed in addition to the nitrogen removal intended. The improvements indicated an increase in bioflocculation, suggesting an enhanced potential for adsorption (Van Winckel et al., 2019). Trapping oxygen transfer inhibitors onto the floc (i.e., surface-active agents, or surfactants, and some of the COD fractions) may have been impacting positively the overall oxygen transfer efficiency, as suggested in recent literature (Stenström and la Cour Jansen, 2017; Stenström and La Cour Jansen, 2016; Tomczak-Wandzel et al., 2009).

Contact stabilization is among these new strategies aimed to enhanced sorption (Meerburg et al., 2016; Sarioğlu et al., 2004; Vasquez Sarria et al., 2011). The feast famine conditions induced in a contact stabilization layout are recognized as strategy to regulate EPS production and bioflocculation (Meerburg et al., 2016; Rahman et al., 2017a). The famine period during the stabilization phase is meant to induce starvation conditions for bacteria, turning them into the oxidation of the more easily available adsorbed material in the floc. This oxidation will free new sorption sites able to capture during feast conditions those substances associated with the reduction of oxygen transfer. However, the integrity and composition of the EPS forming the floc is also expected to suffer from bacteria scavenging during the starvation period, as under stress-famine conditions bacteria can used floc structural polymeric substances as substrate (Meerburg et al., 2016). Later, during the contact phase, the arriving biodegradable COD is expected to be transformed into new EPS, adding new sorption sites to the already obtained during the stabilization phase. Therefore, while the addition of a stabilization phase has proven to benefit biosorption, is still unclear the final balance on the overall production of sorption sites. The present study was designed to determine the potential to improve oxygen transfer in those cases or configurations with maximized bioflocculation/adsorption (e.g., contact stabilization, anaerobic selectors), while also trying to identify, through a set of specific experiments, the underlying mechanisms impacting oxygen transfer in WWRFs.

Enhanced removal of oxygen transfer depressors by sorption enhancing strategies could be considered a potential venue to improve aeration efficiency and reduce the concurrent operational costs. Maximizing sorption in activated sludge systems might, then, have two relevant direct beneficial impacts on aeration: 1) reduction of the overall oxygen requirements, for only the oxidation of readily biodegradable organics fraction is targeted; 2) maximization of the oxygen transfer efficiency as increased adsorption lowered the concentrations of mass transfer depressors (i.e., surfactants, colloids). Their collateral and untapped potential to maximize aeration savings should not be overlooked, as supplying oxygen during the biological oxidation is recognized as the highest energy use in wastewater treatment, accounting for more than 50–75% of the net power demand (Reardon, 1995; Rosso et al., 2005; Rosso and Shaw, 2015; WEF, 2009). The low standard oxygen transfer efficiency in process water (αSOTE, %), hence the low transfer of oxygen from the gas to the liquid phase per unit time (oxygen transfer rate; OTR, $kgO_2\ h^{-1}$) govern operating costs (Garrido-Baserba et al., 2016; Rosso and Stenstrom, 2007). The low efficiency is mainly caused by the effect of contaminants in wastewater, with surfactants being the most known contributors at depressing oxygen transfer (Eckenfelder and Barnhart, 1961; Gillot and Héduit, 2008; Hebrard et al., 2009; Henkel et al., 2009; Sardeing et al., 2006; Wagner and Pöpel, 1996). Other compounds of composition not characterized or unknown, such as the colloidal constituents which have been traditionally overlooked and aggregated with dissolved compounds, are also theorized to play an active role in the decrease of mass transfer efficiency in wastewater and its potential impact should be incorporated within the analysis.

The exact relation between the degradation/adsorption of oxygen transfer depressors (i.e., surfactants, colloids) due to sorption-enhancing technologies and the corresponding improvement in transfer efficiency is still unclear. This research compiled the impact of the sorption-enhancing strategy of bioaugmentation deployed at full-scale over a 5-year period. The observed improvement in bioflocculation and the overall WRRF performance was further assessed to elucidate the underlying mechanisms increasing oxygen transfer when biosorption-enhancing strategies were applied. The objective of this study was to quantify the effect of the wastewater matrix characteristics and plant layout on oxygen transfer efficiency, thus determining the effect of the main αSOTE inhibition contributors (i.e., surfactants, colloids and soluble fractions). Four biosorption-enhancing strategies: (i) High Rate Activated Sludge, (ii) Bioaugmentation, (iii) Contact-Stabilization, and (iv) Anaerobic Selector, were evaluated on their sorption potential and impact on oxygen transfer.

2. Materials and methods

The aeration studies were conducted at Blue Plains Advanced Wastewater Treatment Plant (AWWTP; Washington, DC) which treats over 1.1 million cubic meters of municipal wastewater per day, on average (~375 Mgal d^{-1}). The full-scale plant at Blue Plains treats raw wastewater through screening and aerated grit removal and then proceeds with chemically enhanced primary treatment (CEPT). The CEPT effluent is then treated through biological step-feed high-rate activated sludge (HRAS) with SRT of 1–3 days and is followed by biological nutrient removal (BNR) activated sludge for nitrification and denitrification (SRT = 15 d). The residual ammonia and phosphorus from the BNR system are further removed via enhanced nutrient removal (ENR) processes and then treated effluent is passed through multimedia filtration and, disinfection processes and finally discharged to the Potomac River. A full description of the reactors can be found in Mancell-Egala et al. (2017) and the typical process and wastewater characteristics of Blue Plains AWWTP can be found in the supplementary information (S.1).

Each of the HRAS secondary reactors is operated in step-feed configuration and has 4 channels/passes with anaerobic selectors. The DO is controlled at 1 mg O_2 l^{-1} with coarse bubble aeration and a targeted average mixed-liquor suspended solid (MLSS) of 2500 mg l^{-1}. Secondary reactors are divided in East and West system according to the received primary influent, which can present slight differences in wastewater characterization. Some channels in the secondary reactors at the Blue Plains AWWTP are bioaugmented with return sludge from the BNR reactor. This was implemented to allow for some nitrification in the secondary reactor, while maintaining the short SRT (1.32 ± 0.33 d), reducing the nitrogen load going to mainstream BNR systems. The level of bioaugmentation has been modified over a five-year span. Both East and West systems were operated under full bioaugmentation mode (0.7 ± 0.15 kg $TSS_{waste\ nitrifying\ sludge}$ kg TSS_{HRAS}^{-1} d^{-1} for West, 0.6 ± 0.07 kg $TSS_{waste\ nitrifying\ sludge}$ kg TSS_{HRAS}^{-1} d^{-1} for East), partial bioaugementation mode (0.35 ± 0.16 kg $TSS_{waste\ nitrifying\ sludge}$ kg TSS_{HRAS}^{-1} d^{-1} for both), and without bioaugmentation.

2.1. Sidestream column testing for evaluating bioaugmentation vs. non-bioaugmentation sludge on alpha factor using different diffuser materials

Two sidestream column studies were conducted to determine and compare oxygen transfer between different reactor channels. One column study was performed in a channel of reactor operated as a "step feed" reactor receiving waste sludge from the nitrification system (bioaugmentation), while the second study was performed in a plug-flow reactor (being fed 100% at the beginning of the first pass) without bioaugmentation. Both studies were performed in a column with 3.0 m of side water depth and 2.7 m of diffuser submergence (Fig. 1). The column was 0.80 m in diameter and 3.6 m deep in total (i.e., with a freeboard of 0.60 m). Five different fine-pore diffusers and one coarse bubble were tested in each column. Mixed-liquor was continuously pumped through the column at constant rate to produce a steady-state column DO concentration of 1–3 mg l^{-1} with the air diffuser operating within the range of air flows recommended by their respective manufacturers. The pump to feed continuously the column with mixed-liquor was centrifugal (Grainger, catalogue number 4HU75), delivering 265 l min^{-1} at 4.6 m of head and 250 l min^{-1} at 6.0 m of head. Initial clean water transfer efficiencies (SOTE, %) were measured in situ using the ASCE (2007) clean water testing standard protocol. For both studies, SOTE ranged from 2.4%/m for the coarse bubble to 5.2–6.5%/m.

2.2. Pilot reactor experiments to investigate the impact of surfactant and COD fractions on OTE

This study was performed in a pilot-system consisted of cylindrical reactors (227 l), secondary clarifiers (306 l) and a return activated sludge (RAS) buffer tank (50 l). Aeration and mixing were achieved with coarse bubble diffusers and a blower. The detailed description of the pilot reactor and the schematic diagrams of the investigated configurations are described in supplementary information (S.2). Different synthetic wastewater mixtures were prepared by adding different concentrations of surfactants, colloidal COD surrogate and soluble COD surrogate. Surfactant concentrations of 0, 1, 5, 10 and 15 mg l^{-1} of sodium dodecyl sulfate were added to the 227 l bulk volume of the pilot reactor. As a particulate/colloidal COD surrogate concentration of 5, 10, 20,50, 100 and 200 mg COD l^{-1} of powdered potato starch were used, and as a soluble COD surrogate acetate the concentrations ranged from 5, 10, 20, 50, 100–200 mg COD l^{-1}.

The reactor was mixed with 113.5 l of HRAS collected from a full-scale secondary reactor and 113.5 l of nitrification/denitrification effluent (collected from full-scale BNR reactors) to reach the total volume of 227 l and a final TSS concentration of 100 mg TSS l^{-1}. The batch procedure consisted in the aeration of a total volume of 273l, consisting in a mixture of 3l of sludge plus 270l of primary influent to ensure the right food to mass ratio F:M for the biomass (or TSS target of 150 mg l^{-1}).

The pilot reactor mixture was constantly aerated using an air blower, which supplied air for 2 h during the experiment. The air flow was controlled at 0.28 m^3 h^{-1} by acrylic rotameters (0.05–0.5 m^3 h^{-1} range) supplied by Cole-Parmer. A "YSI Pro plus DO meter" with probe placed at 50% of the water depth was used to measure DO concentrations logged in at 1 min intervals. This meter employed a polarographic sensor, appropriate for clean-water tests (Baquero-Rodríguez and Lara-Borrero, 2016; Philichi and Stenstrom, 1989). The off-gas measurements were conducted from a pilot reactor using the same procedure described in section 2.4.

2.3. Pilot-scale reactor experiments for investigating sorption enhancement and oxygen transfer efficiency under contact stabilization

The same pilot reactor was used to evaluate the impact of a contact stabilization reactor configuration on surfactant removal and aeration efficiency indicators. The pilot-scale batch reactor consisted of a 230 l cylindrical reactor column with diameter of 0.25 m and depth of 4.57 m. A EPDM fine-pore disc (0.23 m or 9in) was flanged at the bottom of the reactor column (i.e., 100% floor coverage). To replicate microbial starvation, 113 l of the bioaugmented HRAS sludge from the full-scale plant HRAS reactor was collected and aerated for 3 and 6 h in a 230 l pilot reactor column at a DO ≥ 6 mgO_2 l^{-1} without the addition of substrate. At the end of the corresponding stabilization period, the air was turned off and 113 l of CEPT effluent was added to the pilot reactor column to mimic the contactor phase. The contactor mixture was re-aerated to maintain the DO above 2 mgO_2 l^{-1} for carbon oxidation and to maintain aerobic conditions for the 1 h duration of the contact phase.

An air blower was used to supply air to the diffuser during the stabilization phase and also during the 1-h contact phase of

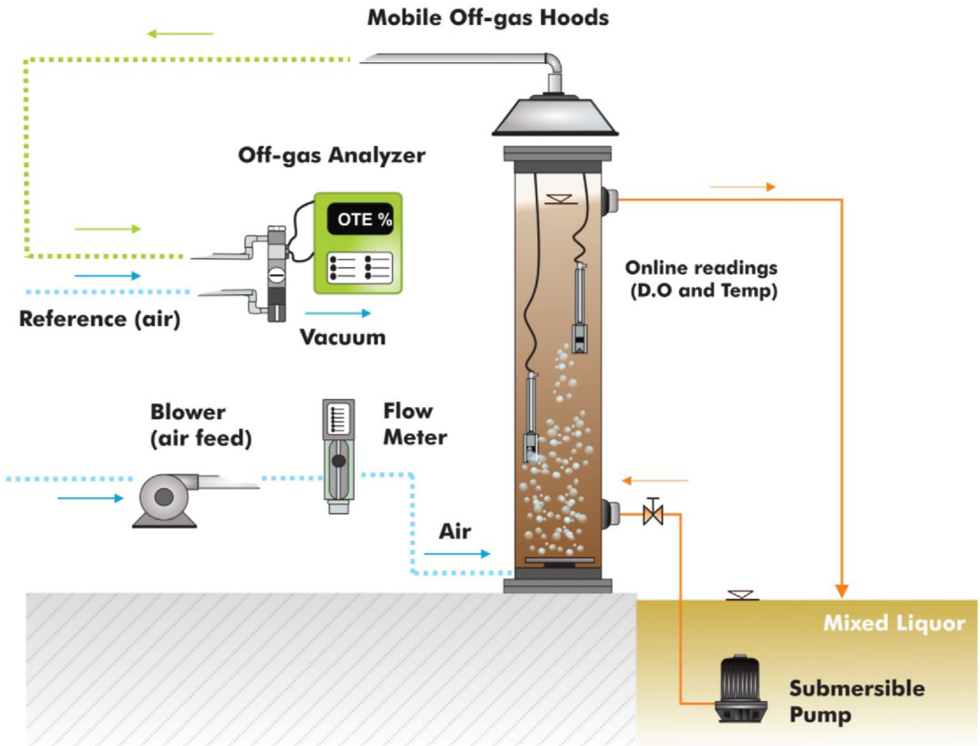

Fig. 1. Schematic representation of off-gas measurement in column testing.

aeration in the experiments. The contactor column was covered with a lid to collect the off-gas for analysis (see §2.5). The airflow was regulated at a rate of 0.28 m^3 h^{-1}, by acrylic rotameters (range = 0.05–0.5 m^3 h^{-1}) supplied by Cole-Parmer. An YSI Pro plus DO meter with the probe placed at 50% of the water depth was used to measure DO concentrations logged in at 1 min intervals.

Controlled experiments were also performed using the conventional HRAS configuration with only CEPT wastewater and HRAS (without pre-aeration) contact phase to compare the conventional plug-flow HRAS configuration to the CS system. Both experiments were repeated under anaerobic conditions to serve as the sorption control system. Samples were collected at 5, 30 and 60 min, and at the end of the stabilization period. The same procedure was applied during the contactor phase. In each sample, COD fractions, TSS, and surfactants concentrations were measured.

2.4. Pilot-scale batch reactor experiments for investigating sorption enhancement and oxygen transfer efficiency using anaerobic selector configuration

The 230 l pilot-scale batch reactor was filled with a mixture of chemically enhanced primary treatment effluent and bio-augmented high rate activated sludge (HRAS) in a ratio of 50:50 with final reactor mixture TSS level of 100–110 mg TSS l^{-1}. Three different HRT times for the anaerobic selector phase were used (15, 30, and 45 min). Anaerobic conditions were created by purging the reactor sample mixture with nitrogen gas to maintain DO at zero for 15, 30, and 45 min, correspondingly. Thereafter, air was added for 1 additional hour to restore aerobic conditions while also performing off gas measurements as described in §2.5.

2.5. Off-gas analysis

Oxygen transfer in process conditions was measured using the off-gas method following the ASCE testing protocol (ASCE, 1997). Off-gas analysis relies on a mass balance around the water column where the air is flowing (Redmond et al., 1992; Schuchardt et al., 2007). The measurements of oxygen partial pressure are used to calculate the standardized oxygen transfer efficiency in wastewater (αSOTE, %), the standardized oxygen transfer rate in wastewater (αSOTR, kgO2 h^{-1}), and of their respective airflow-weighted averages. When clean water tests are available, α can be calculated a posteriori as the ratio of αSOTE and SOTE.

The α factor was calculated as the ratio of process water αSOTE and clean-water SOTE. The latter was obtained for each diffuser from their corresponding manufacturers, as clean-water tests are unlikely at full-scale.

2.6. Analytics

The Hach® TNTPlus™ cuvette test TNT874 (Loveland, CO) was used for the measurement of anionic surfactants, which are a heterogeneous mixture of single components. Linear alkyl sulfonates (LAS) are anionic surfactants that are extracted in chloroform and evaluated photometrically. A cuvette test TNT874 was calibrated with sodium dodecyl sulfate (SDS) for the 0.1–4.0 mg L^{-1} and the results were reported as mgLAS l^{-1}.

TSS was measured according to the standard methods (APHA, 2005). COD was determined using Hach® (Loveland, Colorado, USA) kits.

2.7. Statistics

Statistical significance between treatments was determined with an unpaired *t*-test where unequal variances were assumed due to the small sample size. To determine the significance between slopes, three different slopes were calculated using linear regression and an unpaired *t*-test was performed on the resulting slopes.

3. Results and discussions

3.1. αSOTE in bioaugmented and non-bioaugmented full-scale reactors

3.1.1. Impact of different degrees of bioaugmentation on oxygen transfer

To determine the influence of the bioaugmentation process, aeration efficiency indicators (i.e. α factor and αSOTE) were monitored in the full-scale reactor during three key periods, associated with 3 different bioaugmentation levels, over a 5-year span. Conditions were altered between the East and West reactors to exclude the impact of potential differences in wastewater characterization. Both reactors operated under full bioaugmentation mode (0.7 ± 0.15 kg TSS_{WNS} kg TSS_{HRAS}^{-1} day^{-1} for West, 0.6 ± 0.07 kg TSS_{WNS} kg TSS_{HRAS}^{-1} d^{-1} for East), partial bioaugementation mode (0.35 ± 0.16 kg TSS_{WNS} kg TSS_{HRAS}^{-1} day^{-1} for both), and without bioaugmentation. Off-gas tests along the reactor system in bioaugmented passes consistently yielded an approximate 30–38% improved aeration efficiency in comparison to non-bioaugmented passes. A 37 ± 2% improvement in alpha factor was reported when the West reactor was fully bioaugmented. The latter was a little higher than the period where the East reactor was full bioaugmented (Fig. 2), which reported a 25 ± 4% higher aeration efficiency. Coinciding with the improvement of the aeration efficiency due to full bioaugementation, decreased surfactant levels of 4.6 ± 0.7 mg SDS l^{-1} and 5.2 ± 0.5 mg SDS l^{-1} for West and East reactors, respectively, were observed when bioaugemented (Fig. 3). Non-bioaugmented passes yield an average concentration of nonionic and anionic surfactants of 9.2 ± 1 mg l^{-1} and 8.7 ± 2 mg l^{-1}, respectively. During the sampling period, a 10–15% surfactant removal along non-bioaugmented channels was observed. On the other hand, surfactant concentrations in bioaugmented channels decreased an approximate 40–50% from the beginning of the first channel to the end. Removals from 57% to 90% for the non-ionic and 95–99% of the anionic surfactants were measured in the effluent of the secondary clarifier for non-bioaugmented and bioaugmented respectively.

During the third period (50:50), when the waste nitrifying sludge was equally split between the West and East reactors, both alpha factors and surfactant removal improved in comparison to non-bioaugmented periods (7 ± 1% and 20 ± 3%, respectively in West; and 6 ± 1% and 26 ± 4% in East). It should be noted that under partial bioaugmentation regimens similar improved results for alpha and surfactant concentrations were obtained when compared to full bioaugmentation periods. The threshold of flocculation (TOF) and sludge volume index (SVI) reported for the bioaugmentation condition improved by 31 ± 2% and 43 ± 4%, respectively, as reported by Van Winckel et al. (2019). This confirmed that the addition of activated sludge from long SRT configurations enhanced the bioflocculation properties of the system.

Operating at long SRT to allow nitrification/denitrification is recognized to have positive effects on oxygen transfer in WRRFs (Rosso, 2018; Rosso and Stenstrom, 2007). Traditionally, it has been assumed that longer SRT enhances the removal of refractory substrates (Germain et al., 2007; Gillot and Héduit, 2008; Groves et al., 1992; Henkel et al., 2011; Soliman et al., 2007) The addition of nitrifying sludge (>12 d) has shown in previous studies to improve the catabolism of specific compounds, e.g. surfactants (Sheng et al., 2017; Stenström and la Cour Jansen, 2017; van der Gast et al., 2004). Nevertheless, this may not be the case for short SRT at HRAS systems (<2 d) where the limited time may hamper the complete metabolization of refractory organics. More likely, almost-instant physical chemical processes, such as sorption, would better explain the removal of surfactants in the liquid phase at short SRT systems. This aspect was further studied in controlled experiments (see §3.3).

3.1.2. Influence of aeration equipment on improvements due to bioaugmentation

Four different types of diffusers were analyzed to quantify the impact on aeration efficiency indicators and their relationship with full bioaugmentation conditions. Transfer efficiencies in the

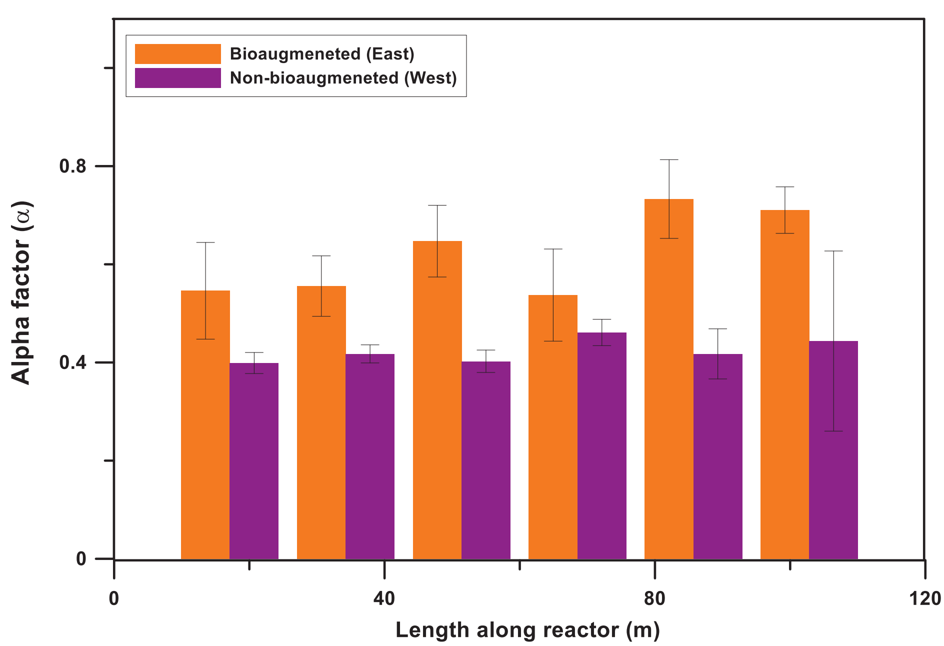

Fig. 2. Measured alpha factor at full-scale step-feed secondary reactors with and without full bioaugmentation. Each reactor was divided in two passes. Alpha values were measured along different lengths within each of the two different passes per reactor (orange bars represent full bioaugmented regimen and purple non-bioaugmented). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

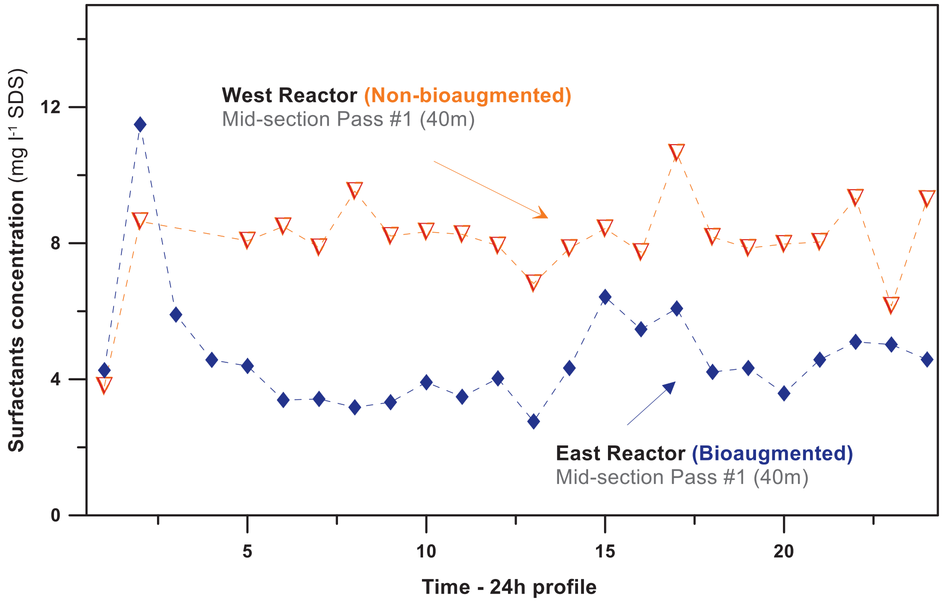

Fig. 3. Profiles over 24 h of surfactant concentrations in the West (non-bioaugmented) and East (bioaugmented) reactors.

bioaugmented testing column ranged from 5.2% αSOTE to 13.4% αSOTE, while the non-bioaugmented stalled between 2 and 6% αSOTE (Fig. 4). Similarly, the α factors for bioaugmented ranged from 0.67 to 0.73, which is more than 35% higher in relative terms than for the non-bioaugmented reactors. The high α factors for the fine pore diffusers did not include any effects from fouling since they were all new. It should be noted that differences between the two biological regimens (bioaugmentation vs. non-bioaugmentation) were minimal when coarse bubble diffusers were used.

The observed α factors in bioaugmented passes were significantly higher (approximately 30%, in relative terms) compared to those expected values for low SRT processes (0.40 ± 0.08) with carbon-only treatment (Rosso et al., 2005), indicating the impact of bioaugmented sludge. On the other hand, the oxygen transfer rates measured in the column fed by non-bioaugmented sludge (0.42 ± 0.07) were closer to those expected values for carbon-only treatment in both coarse and fine-pore diffusers.

Therefore, efficiency indicators (i.e. α factor and αSOTE) showed that bioaugmented sludge experienced an increase in efficiency of nearly 36 ± 0.5% in relative terms for the α factor and 45 ± 1% in relative terms for αSOTE. This result, although obtained in an open reactor, was similar to the results obtained at full-scale (see §3.1.1). The type of diffuser did not reveal significant impacts on the aeration efficiency when the two types of sludge were compared, thus indicating that the wastewater characterization variations due to bioaugmentation were the main cause for improved oxygen transfer.

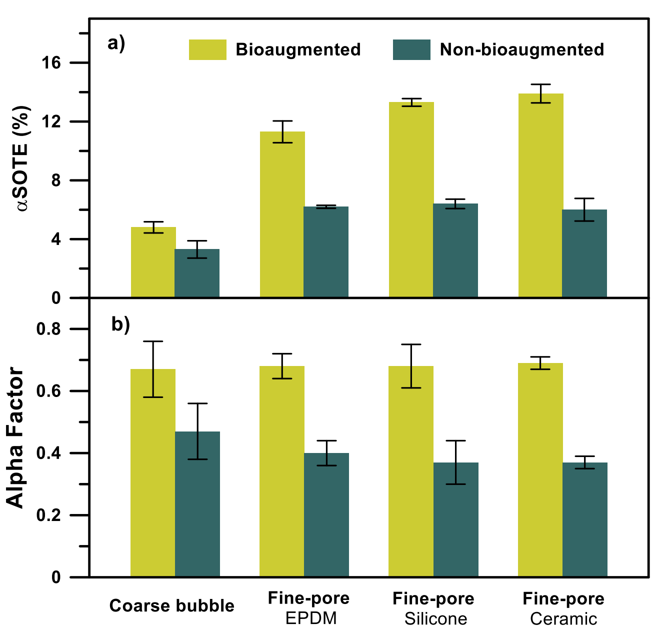

Fig. 4. Results of oxygen transfer tests performed on a side stream column continuously circulating the sludge from the full-scale reactors. The type of fine-pore diffusers corresponds to EPDM, Silicone and Ceramic, respectively.

3.2. Impact of wastewater characterization on OTE

Fig. 5 shows the progressive removal of organics along the main process stages, with the concurrent improvement in α.

To determine the exact influence of each of the wastewater constituents (COD fractions and surfactants) on oxygen transfer efficiency, a set of experiments in controlled environments were conducted (Fig. 6) using the following model compounds: acetate (as soluble COD source), starch (as colloidal COD source), sodium dodecyl sulfate (as surfactant) and harvested surfactant from the East reactor (as collected foam). To exclude the impact of suspended solids, all wastewater mixtures were diluted to have a TSS lower than 100 ± 10 mg l-1 and were then spiked with the model compounds.

Surfactant addition of both as synthetic as well as harvested form decreased oxygen transfer significantly (Fig. 6a). While the addition of 1 mg SDS l^{-1} resulted in a decrease in alpha factor (35 ± 3% and 36 ± 2% in relative terms for synthetic and harvested surfactant, respectively), increasing the surfactant concentration 10-fold (10 mg SDS l^{-1}) had little impact (47 ± 3% in relative terms). The obtained measurements are in accordance with previous studies reporting the inhibition of surfactants on mass transfer at the gas-liquid interface (Dereszewska et al., 2015; Liwarska-Bizukojc and & Bizukojc, 2006; Tomczak-Wandzel et al., 2009; Wagner and Pöpel, 1996). The fitting of a reverse Monod type inhibition curve to the data resulted in an

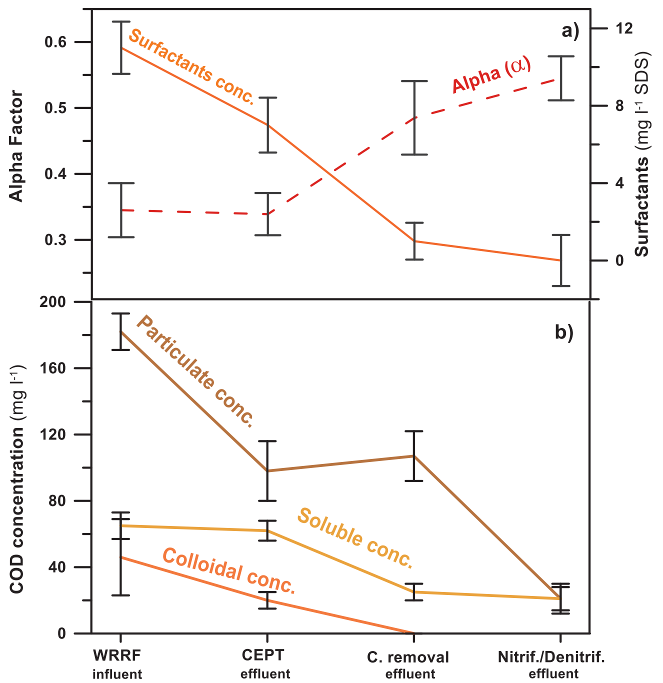

Fig. 5. a) Profiles of alpha factor and surfactants concentration; b) measured COD fractions of Blue Plains' different wastewater matrices [Influent; Influent after chemical enhanced primary treatment (CEPT effluent); effluent from the carbon removal stage (HRAS); and effluent from the biological removal nutrients stage (nitrification/denitrification)].

inhibition coefficient (Ki), expressing the concentration to obtain an inhibition of 50%, of 2.5 ± 0.5 mg SDS l^{-1} ($R^2 = 0.99$). The p-values associated with the unpaired Student's T-test for synthetic and harvested surfactants were $3.57 \cdot 10^{-4}$ and $8.40 \cdot 10^{-3}$, respectively, indicating a statistically significant correlation ($P < 0.05$) between surfactants and oxygen transfer efficiency. On the other hand, whilst still significant, the colloidal COD presented a much lower statistically correlation with aeration transfer efficiency ($3.96 \cdot 10^{-2}$). All estimated parameters for the reverse Monod inhibition fit of the data, for both synthetic and harvested surfactant, are given in Table 2. The reproducibility of controlled experiments using surfactants can be considered challenging as surfactants are very sensitive to background interferences and they need extremely pure solutions for the correct quantification (Nasiru et al., 2011; Racaud et al., 2010). Therefore, caution should be taken when compared to site-specific wastewater full-scale conditions (i.e., fibers, colloidal matter, microbial flocs, unknown SAA, adhering constituents, etc.). The reverse Monod curve was used to calculate the theoretical change in alpha factor considering the surfactants concentrations sampled during previous periods (Table 1) before and after the full bioaugmentation system. The calculations show a theoretical improvement in alpha of 0.23 ± 0.05% in relative terms (from 0.29 to 0.38 in absolute terms) when reducing the surfactants concentration from non-bioaugmented (8.5 ± 0.04 mg SDS l^{-1}) to bioaugmented conditions (5.4 ± 0.06 mg SDS l^{-1}). This theoretical improvement is similar to the measured at full-scale 19 ± 1% in relative terms (Fig. 2). This confirmed the validity of the estimated Ki value. K_i values ranging from less than 1–13 mg l^{-1} were recorded in literature for certain anionic surfactants (Chauhan and Sharma, 2014; Hait et al., 2003; Zhang et al., 2003). The obtain Ki in this study was towards the low compared to existing literature.

Fig. 6b showed a decline of 42 ± 3% in relative terms for alpha driven by the additions of colloidal material. A similar Monod inhibition model fitted the data (Table 2) and resulted in an estimated K_i of 7 ± 1 mg COD l^{-1} for colloidal material. The effect of colloidal material on oxygen transfer has been largely overlooked in literature, with some studies suggesting no relevant effect on K_{La} with the addition of a highly colloidal substances (Hwang and Stenstrom, 1985). Conversely, in this study we could confirm that colloidal and particulate material impacted oxygen transfer efficiency by changing wastewater rheological properties. This findings are in agreement with previous research reporting that an increased density or liquid viscosity by colloidal material can hinder the volumetric transfer rate as the movement of gaseous molecules is reduced (Amaral et al., 2016; Durán et al., 2016; Nittami et al., 2013). Moreover, since colloidal and particulate COD are slowly biodegradable, they could result in a potential suppression of oxygen transfer by inducing low oxygen uptake rate conditions (Reiber and Stensel, 1985).

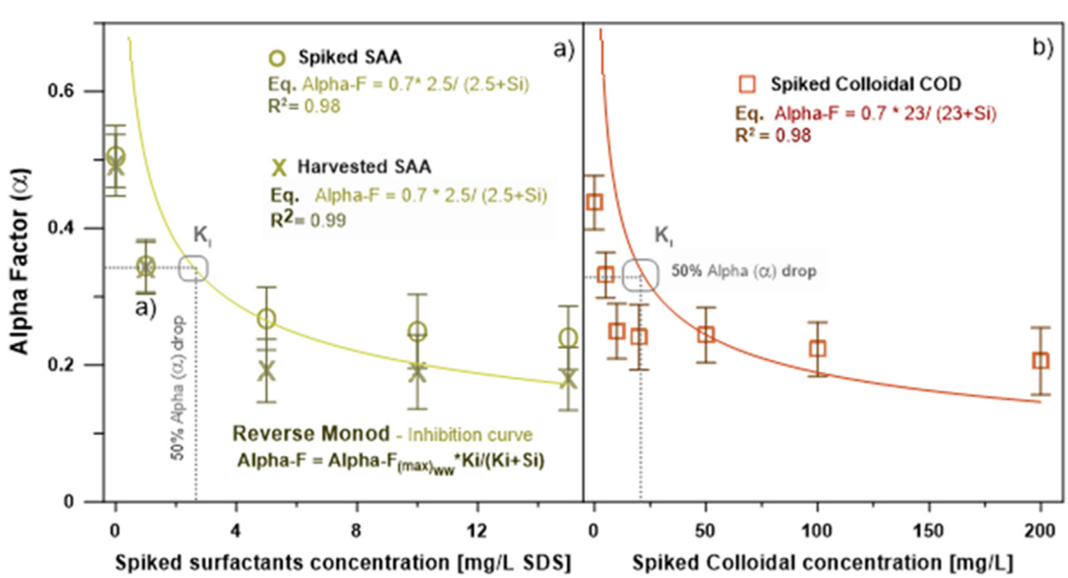

Fig. 6. Alpha factor (α) after the addition of surfactants (sodium dodecyl sulfate and surfactant present in the primary effluent), colloidal COD (powdered potato starch), and soluble COD (acetate). The same wastewater mixture and TSS dilution (100 mg l^{-1}) was applied to all batch-scale experiments.

Table 1
Aeration efficiency (alpha factor) and surfactant concentration over the three bioaugmented regimens for west and east reactors. Period I represents the interval when West reactors received the full amount of produced waste nitrifying sludge, while Period II represents the interval when East received the full amount. Period III refers to the time when the waste nitrifying sludge was split 50:50 between the two groups of reactors.

		Period I		Period II		Period III	
		East	West	East	West	East	West
Bioaugmentation rate	kg TSS kg TSS^{-1} d^{-1}	*nil*	0.70 ± 0.15	0.06 ± 0.07	*nil*	0.34 ± 0.18	0.37 ± 0.11
Surfactants	mg SDS l^{-1}	7.97	5.31	4.83	9.03	3.49	4.12
Alpha	(−)	0.35 ± 0.05	0.56 ± 0.04	0.52 ± 0.05	0.39 ± 0.02	0.45 ± 0.03	0.47 ± 0.04

Table 2
Monod inhibition equation fitting parameters and confidence on model.

	Ki	Alpha − max (in wastewater)	P-value model
Synthetic surfactants	2.5 ± 0.5	0.71 ± 0.04	3.57 10^{-4}
Harvested surfactants	2.5 ± 0.6	0.73 ± 0.07	8.40 10^{-3}
Colloidal COD	23 ± 4	0.68 ± 0.10	3.96 10^{-2}
Soluble COD	−	0.71 ± 0.05	−

No impact of soluble COD on the alpha factor was observed. Although it cannot be discarded that other types of soluble compounds found in wastewater can exert a detrimental effect on alpha, the current experimental setup demonstrated that none of the acetate concentrations applied (2−200 mg l-1) had a significant impact on decreasing alpha.

3.3. Increased sorption improves wastewater characteristics (and aeration efficiency)

Two alternative strategies to improve sorption, other than bioaugmentation of long SRT sludge were evaluated. Application of contact-stabilization and application of anaerobic selectors to improve sorption of surfactants and colloidal COD were proposed and tested in detail in pilot-scale batch tests.

3.3.1. High-rate contact stabilization as a strategy for improved oxygen transfer

Without any stabilization time, an alpha of 0.48 ± 0.20 and surfactant level of 2.8 ± 0.2 6 mg SDS l^{-1} was observed (Fig. 7b). Adding a stabilization time of 3 h resulted in an increase in α and surfactants removal by 16% and 22.5% in relative terms (Fig. 7b). These results are in accordance with previous studies demonstrating that the presence of a stabilization (famine) phase followed by low DO contact with the influent (feast) could improve biosorption capacity and obtain higher bioflocculation affinity and settleability (Meerburg et al., 2015a; Rahman et al., 2016; Vasquez Sarria et al., 2011). The increase in biosorption was not only suggested by an enhanced removal of surfactants, but also with a correlated increase in EPS production suggesting improved bioflocculation (Rahman et al., 2017a).

When increased stabilization times were used, decreased aeration efficiency and surfactant removal were observed. While the extension of the contact stabilization time to 6 h resulted in a significantly lower surfactant removal in comparison (12 ± 1.0%), extending the CS time to 15 h showed no improvement (Fig. 7b). Similarly, alpha values were even lower than those reported when there was no contact stabilization time (0.45 ± 0.06% and 0.39 ± 0.04% in relative terms). Although the depletion of biodegradable COD (freeing sorption sites) was expected to be further boosted during famine conditions by the release of microbial exocellular-enzymes improving the hydrolysis stage (Benefield and Randall, 1976; Sarioğlu et al., 2004), excessive stabilization periods may have induced the consumption of the required EPS for sorption, hampering the capacity to capture surfactant or other substances hindering oxygen transfer.

3.3.2. Anaerobic selectors as a strategy for improved oxygen transfer

To investigate the potential of anaerobic sorption of surfactants rather than a combination of sorption and oxidation, an anaerobic selector was applied. The implementation of an anaerobic contact phase with hydraulic retention times of 15, 30 and 45 min in the pilot-scale resulted in surfactant removal efficiencies of 74.5%, 87.3%, and 86.7%, respectively, and alpha factor values of 0.52, 0.62, and 0.58, respectively (Fig. 7a). This configuration yielded the highest results for both surfactants removal and alpha factor. About 117% improvement on surfactant removal was obtained in the anaerobic selector system when compared to the conventional HRAS system, while about 74% and 49% when compared to the bioaugmented reactor system and the CS configuration, respectively. Similarly, when compared to conventional HRAS, bioaugmented HRAS (50:50) and CS the alpha factor improved 63%, 35% and 20% in relative terms, respectively. The higher removal of surfactants obtained with the presence of an anaerobic selector indicates a clear increase in the biosorption capacity of the biomass.

As discussed previously, the observed biosorption capacity increase in the CS configuration may have corresponded to the enmeshment of surfactants and colloidal substances in the floc structure due to the increase in sorption sites (or free surface area) during the stabilization phase. Nevertheless, extended stabilization phases (or famine periods) may debilitate existing EPS as scavenging bacteria potentially turned to EPS in search for available substrate when the F:M in the reactor decreased during the famine period (Rahman et al., 2016).

The anaerobic selector configuration, on the other hand, may have benefited from a similar sorption mechanism, while a series of other physicochemical processes might have helped to further increase the sorption capacity.

Full-scale plants (except bioP configurations) usually locate anaerobic selectors after the returns of mixed liquor suspended solids coming from the recirculating activated sludge. The RAS is under famine conditions (end of aerobic zones) and had expectedly suffered minor losses of EPS as the anaerobic conditions in the clarifiers might have reduced the degradation rates of the EPS in comparison to aerobic conditions. Similarly, this study used bioaugmented sludge, also under famine conditions with potentially more free sorption sites, that in contact with rich influent may have

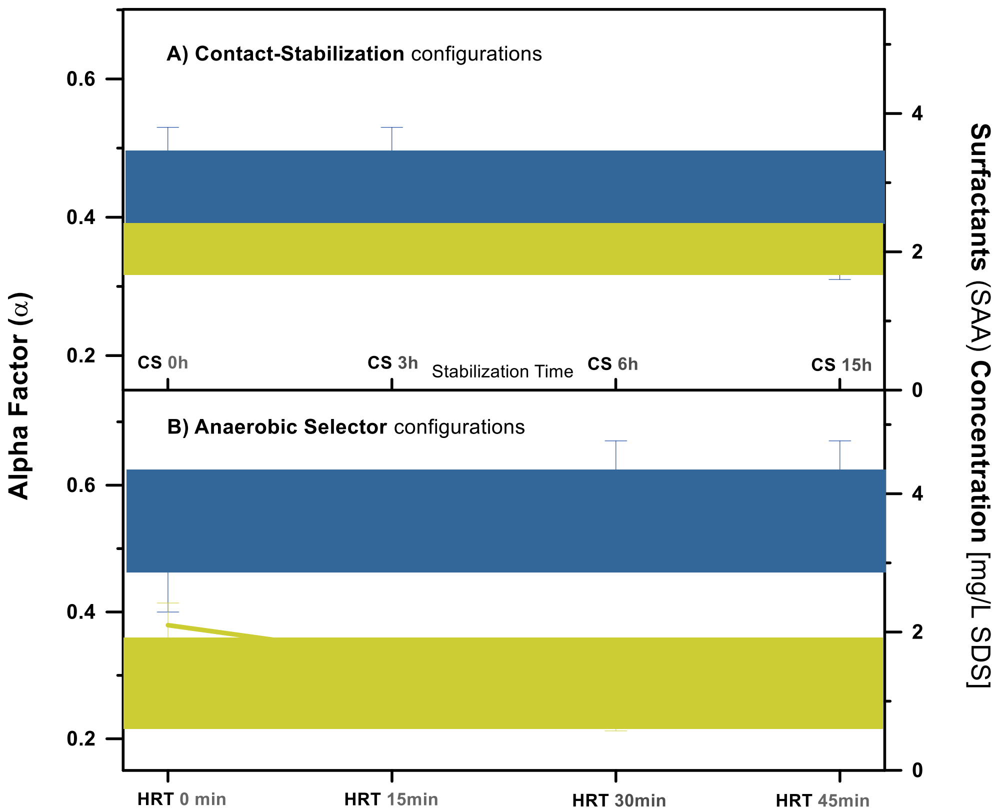

Fig. 7. Oxygen transfer efficiency (αSOTE) and surfactants removal obtained at the pilot reactor for the anaerobic selector and the contact stabilization configurations.

maximized the concurrent adsorption of mass transfer-reducing substances, while the anaerobic conditions might have maintained the integrity if the EPS to preserve biosorption.

Some recent studies, albeit under different operational conditions, have pointed out that the addition of anaerobic phases can increase not only the quantity, but also the quality of the EPS, by increasing the protein content in the EPS composition (Yao et al., 2019). The increase in protein concentration, which can require longer residence time, can decrease the effective critical potential on the surface of microorganisms and promote bioflocculation, enhancing both settling characteristics and sorption (Liu et al., 2014; Miao et al., 2018; Yao et al., 2019). Nevertheless, Rahman et al. (2016) reported no fresh EPS production during anaerobic phases and suggested that the EPS production was only improved when aerobically stabilized sludge (famine condition) was exposed to feast conditions, without significant changes on the EPS C:N ratio. The present study seems to align with the Rahman et al. (2016) as not relative increase in the EPS production was observed (Odize, 2018). However, it must to be noted that even if anaerobic conditions have the ability to increase EPS production and modify EPS characteristics, the rate of these changes under an anaerobic phase might have been too slow to have any significant impact sorption in the short term (Batstone and Keller, 2001; Nielsen et al., 1996).

Another factor that may have contributed to the highest observed surfactants removal, although to a minor extent due the short time of the hydraulic retention time (i.e., 30 min), is the intracellular storage of surfactants under anaerobic condition. Recent studies have shown that anaerobic conditions can promote the production of storage compounds as triacylglycerides, as bacteria accumulates them at low DO conditions, as they limit β-oxidation (Alvarez et al., 1997; Kinyua et al., 2017b). Similarly, other studies based on granular sludge have observed that anaerobic selectors might enhance storage mechanisms leading to increased density and settling stability (basis of granular reactors) especially when the F:M ratio reach a sufficient threshold (McSwain et al., 2005, 2004). Sludge with those mechanisms often exhibit increased protein content (Kinyua et al., 2017a, 2017b).

Similarly, even at lesser extent, is the potential biodegradation on of some non-ionic surfactants. Surfactants metabolization by bacteria is a slow process with degradation rates ranging from hours to a few days (Chang et al., 2005; Lu et al., 2008), which may have had little impact this type of short experiment, as discussed in the following section.

3.3.3. *Determination of surfactants removal mechanism*

The removal of surfactants in secondary treatment conditions has been consistently reported (De Wever et al., 2004; Terzic et al., 2005). Surfactants are assumed to either break down due to aerobic processes and/or being adsorbed on the activated sludge flocs. Several authors indicated that the mechanism of the breakdown for anionic surfactants (i.e., LAS) involves several relatively complex steps (Perales et al., 1999). These required steps may hamper the quick oxidation of surfactant in short SRT systems, indicating that surfactant removals in sorption-enhancing strategies systems is primarily due to adsorption phenomena. This suggests that removal during the contact phase or due to bioaugmentation is governed by physicochemical mechanisms such as sorption and flocculation, discarding any significant role of biodegradation as a removal mechanism. Indeed, it has been demonstrated that sorption of activated sludge is essentially 99% complete within 10 min (Wahlberg et al., 2012).

The CS configuration under optimized stabilization time and longer contact time, on the other hand, achieved 60% removal within the first 15 min, suggesting that biodegradation may have played a bigger role. The impact of biodegradation was already expected to be slightly higher under the anaerobic conditions as the selector provided favorable conditions for bacteria to metabolize soluble material. Nevertheless, it is still unlikely that the 40% removal difference between the CS and the anaerobic selector configurations (HRT = 15 and 30 min) is due to enhanced biodegradation rather than potential sampling errors for this type of reactor configuration (surfactants samples could only be taken at the end of the selector phase).

3.4. Research applicability on full-scale systems

The implementation of biosorption enhancing strategies such as bioaugmentation, contact stabilization, and anaerobic selectors have already shown their capacity to improve bioflocculation and settling characteristics (Mancell-Egala et al., 2017, 2016). This study confirmed that increasing sorption potential has clear benefits on the overall oxygen transfer efficiency and the concurrent operational costs, helping WRRFs to advance toward energy-positive wastewater treatments. The latter strategies mainly result in improvements of the wastewater matrix through surfactant and colloidal material sorption. This research provided evidence that aeration in the secondary system at Blue Plains benefitted from the implementation of a bioaugmentation strategy. While its unique setup, this bioaugmentation approach is not widely applicable to other resource recovery facilities. Nevertheless, the mechanism behind the success of bioaugmentation could be achieved to alternatives such as contact stabilization and related configurations (i.e., addition of an anaerobic selector phase). The necessary adjustments to adopt the discussed sorption-enhancing strategies in any WRRF will have to consider site-specific characteristics and wastewater characterization.

4. Conclusions

The link between aeration efficiency in WRRFs and wastewater characteristics, with a special emphasis on biosorption capacity, was extensively investigated. The biosorption of surfactants and colloidal COD fractions impacted the oxygen transfer significantly. This study provided Ki (50% relative decrease in alpha) levels for both surfactants and colloidal organics at 2.5+-0.6 mg SDS l^{-1} and 23+-4 mg COD l^{-1}, respectively. This showed the high sensitivity of alpha towards these wastewater fractions. In addition, bioaugmentation of long SRT sludge, application of contact-stabilization and implementation of anaerobic selectors, all promoted biosorption of surfactants and therefore significantly increased oxygen transfer rates. This study showed the convergence of strategies applied to improve settleability with improved aeration efficiency and provides practical guideline to improve oxygen transfer in WRRFs.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2020.116086.

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