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Skouteris, G.; Rodriguez Garcia, G.; Reinecke, S.; Hampel, U.;

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THE USE OF PURE OXYGEN FOR AERATION IN AEROBIC WASTEWATER TREATMENT: A REVIEW ON ITS POTENTIAL AND LIMITATIONS

G. Skouteris[1,*], G. Rodriguez-Garcia[1], S.F. Reinecke[1], U. Hampel[1,2]

[1]Department of Experimental Thermal Fluid Dynamics, Institute of Fluid Dynamics, Helmholtz Zentrum Dresden-Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany

[2]Chair of Imaging Techniques in Energy and Process Engineering, Technische Universität Dresden, 01062 Dresden, Germany

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Abstract

In aerobic wastewater treatment, aeration is the most critical element of the treatment system. It supplies microorganisms with the required dissolved oxygen, maintains solids in suspension and, in membrane bioreactors, it controls fouling. However, conventional activated sludge is limited to the treatment of low strength wastewaters, as higher loadings require both higher biomass and higher dissolved oxygen concentrations. By replacing air with pure oxygen, oxygen transfer rates increase at lower flowrates. In this work, the potential and limitations of pure oxygen aeration are reviewed. The effect of the system's operational parameters and the mixed liquor characteristics on oxygen transfer, and vice versa, is determined. Pure oxygen treats higher loadings without compromising effluent quality. Fine bubbles are more efficient in oxygen transfer due to their increased contact

Corresponding Author: G. Skouteris, email: g.skouteris@hzdr.de

area. However, pure oxygen use is not always essential, so we recommend that it be restricted to applications where air use is not adequate.

Key words: aerobic treatment, activated sludge, membrane bioreactor, pure oxygen, fine bubbles

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1. INTRODUCTION

Secondary wastewater treatment (WWT) is an effective and cheap method for removing organic pollutants from wastewaters. Conventional activated sludge (CAS), an aerobic suspended growth treatment process, is one of the most widely used secondary treatment technologies. CAS requires oxygen for the microbial consortia to assist them in degrading the organic matter in wastewater ensuring their maintenance and growth. This makes aeration systems an integral part of the CAS plants, (Zhang et al., 2019; Calderón et al., 2013). Aeration supplies the dissolved oxygen (DO) that is required by the biomass in both CAS and membrane bioreactor (MBR) systems with MBRs being activated sludge systems where membrane filtration has replaced gravitational sedimentation. It also maintains solids in suspension and, in membrane bioreactors, it additionally mitigates membrane fouling and improves membrane cleaning, (Calderón et al., 2012). However, it is an energy intensive process accounting for the largest fraction (40%-75%) of energy costs of a WWT plant. In addition, existing technologies are old, often operating at a standard oxygen transfer efficiency (SOTE) of <15% or at even lower values, after real DO concentrations are considered, (Syron et al., 2015).

In principle, treatment efficiency in CAS is controlled by the DO amount in the aeration tank. Low DO concentrations lead to lower effluent quality due to low growth rates of bacteria. Population of filamentous microorganisms in the sludge also increases, which causes sludge bulking or biological foam (froth) formation, (Xu et al., 2016; Jolis et al., 2006; Lee et al., 2003). When low strength wastewaters are treated, traditional oxygen supply with air does satisfy the need for oxygen. However, CAS fails to treat high strength wastewaters as any increase in either the oxygen transfer efficiency (OTE) or in the biomass concentration within the aeration tank, is limited, (Lee and Kim, 2003).

As such, when high strength wastewaters are to be treated, satisfactory oxygen demand requires an intensified oxygen supply technology, e.g. pressurized aeration, deep shaft technology, pure oxygen (PO) use. In doing so, high mixed liquor suspended solids (MLSS) concentrations are combined with high OTEs. Pressurization accelerates the oxygen transfer rates (OTRs), or the actual mass of oxygen transferred per unit time, by increasing the partial pressure of oxygen. This is a low cost and a small footprint technique that ensures improved oxygen transfer. Deep shaft aeration employs hydrostatic pressure in a deep well to achieve better OTEs and is a technique that does not need much energy or space. Mass transfer depends on the depth of the well and the technique is widely used in plants treating high strength wastewaters. Finally, regarding PO aeration, PO (>90%) replaces air in aeration and maintains good aerobic conditions even when high organic loadings are treated, (Zhang et al., 2019; Xu et al., 2016; Stenstrom and Rosso, 2010; Esparza-Soto et al., 2006a; Lee and Kim, 2003). In this work, we concentrate on PO aeration and analyze its effect on WWT.

PO was first proposed in 1940 as a replacement of air in CAS but it was not put into commercial use until the '70s in the USA. The first PO activated sludge (POAS) plants for municipal effluent treatment were introduced in 1968, (Zhang et al., 2019; Calderón et al., 2013, 2012). To date, various kinds of wastewater have been treated by treatment systems incorporating PO aeration, (Zhang et al., 2019), including industrial wastewaters that contain toxic and refractory contaminants, (Zhuang et al., 2016b), and landfill leachates, (Canziani et al., 2006). In addition, PO has already been used in MBRs, (Rodríguez et al., 2010), biofilms (BFs) and moving bed biofilm reactors (MBBRs), (Salvetti et al., 2006), or in aerobic granulation applications, (Pan et al., 2017). As of 2014, in the USA, as much as 15% of all municipal WWT was realized with the use of PO in aerobic WWT plants, (Larrea et al., 2014). Table 1 shows research studies based on PO aeration and Table 2 shows large scale POAS applications.

TABLE 1: Research studies based on PO aeration

TABLE 2: Industrial WWT POAS applications

Researchers have been comparing the performances of the two aeration types since 1976, (Esparza-Soto et al., 2006a). PO increases the driving force for oxygen transfer and the degree of oxygen saturation possible — its partial pressure is 4.7 times of that of its atmospheric counterpart. It improves the OTR and maintains high DO concentrations at lower flowrates even when high strength or toxic wastewaters have to be treated. Compared with the use of air, PO provides higher gas phase oxygen concentrations, improves biokinetics and allows for faster treatment rates at higher MLSS concentrations and shorter hydraulic residence times (HRTs). Designs of PO based systems are simple and compact and allow for easy gas storage and handling. They handle foul condensates without in-plant stripping, so they reduce odour and volatile organic compound (VOC) emissions, decrease sludge production, as more complete oxidization to CO_2 is achieved, and minimize sludge bulking and biomass foaming problems. PO ensures treatment system stability at reduced power consumption rates and lower sludge disposal costs, (Neerackal et al., 2016; Zhuang et al., 2016b; Larrea et al., 2014; Calderón et al., 2013; Rodríguez et al., 2012b; Paice et al., 2003; Brindle et al., 1998; Shelef and Green, 1980). However, both oxygen generation equipment and the purchase of oxygen tanks have high costs, so oxygen should be utilized at minimum wastage, (Calderon et al., 2012; Brindle et al., 1998).

We herein give an overview on the potential effect of the replacement of air with PO in aerobic treatment methods. We summarize the consequences of the applied operational parameters and the influent and biomass characteristics on oxygen transfer, and vice versa, i.e. the consequences of PO on the treatment process over time. This work deals with PO introduced into tanks in bubbly form, which then dissolves into the liquid. This review has covered much of the existing literature and has dealt with a topic that has not been thoroughly reviewed to date. In addition, it gives a detailed assessment of the use of PO in both CAS systems and MBRs, comparing the two technologies, where possible.

2. PURE OXYGEN AERATION IN AEROBIC WASTEWATER TREATMENT

2.1. Analysis of Pure Oxygen Aeration

Oxygen transfer rates depend both on the driving force, namely the difference of the DO saturation concentration in water minus the DO concentration in water, and on the liquid side mass transfer coefficient (k_La), (Stenstrom and Rosso, 2010; Gostick et al., 1992).

Oxygen transfer in wastewater is usually affected by the biomass characteristics and the design of the aeration system. Aeration and the three parameters that characterize biomass, i.e. particle concentration, particle size and viscosity, are interrelated. Aeration intensity affects particle size and viscosity. Any increase in viscosity has a negative effect on oxygen transfer with the solids concentration modifying it. Oxygen transfer is also affected by the particle size and the particle concentration, whose effects are interrelated. Finally, the design of the aeration system additionally affects oxygen transfer with the α-factor being the main parameter that determines the system's aeration capacity. Both β-factor, which accounts for the effect of salts and particulates in wastewater on oxygen transfer, and the temperature-factor have a lesser impact on mass transfer, so they usually need not to be considered. On the other hand, the α-factor indicates the effect of wastewater on oxygen transfer and it varies with wastewater quality, MLSS concentration and the intensity of mixing or the applied turbulence, (Rodríguez et al., 2014, 2012b, 2011, 2010; Germain et al., 2005).

PO aeration leads to higher OTEs at lower flowrates under higher organic loadings. As the partial pressure of oxygen in the gas phase increases, the driving force for mass transfer also increases. As such, PO provides higher gas phase oxygen concentrations than air, allowing faster treatment under higher MLSS concentrations and shorter HRTs, (Zhuang et al., 2016a; Rodríguez et al., 2013). However, in covered POAS systems in particular, the CO_2 produced may somehow affect oxygen transfer as each time an oxygen bubble is inserted into the mixed liquor, CO_2 enters it reducing the partial pressure of oxygen and the saturation concentration of DO, (Gostick et al., 1992).

In practice, the amount of oxygen that is transferred is highly related to the applied flowrate — the higher the flowrate, the better the oxygen transfer. For the same flowrate of air and

PO, the amount of oxygen transferred is higher for the PO due to its higher driving force. PO has a DO saturation concentration that is up to five times higher than that of air at the same flowrate, (Zhuang et al., 2016a; Rodriguez et al., 2013). Lee and Kim (2003), who monitored the DO concentration changes for PO flowrates of 0.0125 L min^{-1}, 0.025 L min^{-1}, 0.05 L min^{-1}, 0.1 L min^{-1} and 0.2 L min^{-1}, in a tank of a working volume of 21 L, found that when the flowrate changed from 0.0125 L min^{-1} to 0.025 L min^{-1}, a significant sudden increase in the OTR occurred. For the next flowrates, the DO concentrations continued increasing but more gradually. Similarly, Zhuang et al. (2016a) noticed that, when a PO flowrate of 0.5 L h^{-1} increased to 1.5 L h^{-1}, the DO concentration at a specific time of 300 s was 1.8 times higher. In addition, by comparing PO flowrates with air ones, they also found that at a PO flowrate of 2.5 L h^{-1} the DO concentration was five times higher than that for a similar air flowrate. However, there was a limit regarding the DO amount that could be sustained, so any further increase in the PO flowrate could not increase the OTE value further.

With regard to the α-factors, Rodríguez et al. (2014), who determined the α-factors in an MBR fed either with PO or air found that, at a constant HRT, the α-factor increased when MLSS concentrations decreased but, at the same MLSS concentration, the PO related α-factor values were higher. Similarly, Rodriguez et al. (2011) showed that, at a constant HRT (and SRT), an increase in the MLSS concentration from 3420 mg L^{-1} to 12600 mg L^{-1} in an MBR fed with PO decreased the α-factor from 0.426 to 0.022. However, despite the decrease in the α-factor at the high MLSS concentration, the removal efficiency of organic matter remained high. This suggests that PO did maintain the aerobic conditions within the MBR even at high MLSS concentrations. In practice, the effect of MLSS concentrations must be determined in combination with the applied HRT, as HRTs regulate the time period during which oxygen is in contact with the liquid. Rodríguez et al. (2012b) showed that the α-factor was highly affected by both MLSS concentrations and the HRTs, but the MLSS concentration was better correlated with the α-factor than the HRT. However, any data based on a specific case study is unable to be directly compared with any other data, as the α-factor strongly depends on the bubble size, the system's geometry and the EPS/SMP concentrations, although the EPS/SMP concentrations have a lesser effect. In addition, α-factors, which can sustain aerobic conditions at bench scale, must be treated with caution when full scale applications are to be considered, (Rodríguez et al., 2012b).

In addition, it must be mentioned that Zhuang et al. (2016b) also found that the presence of long chain and aromatic hydrocarbons, as well as of ester compounds, in their coal gasification wastewater, hindered oxygen transfer from the bubbles to the liquid in an MBR fed with air. This consequently led to low DO values in this MBR with the ability of microorganisms to biodegrade having been affected.

As a final comment, Rodríguez et al. (2013) also showed that less energy was consumed when PO is employed — the values for oxygen transfer as a function of energy supplied were 0.62 $kg_{O2}\ h^{-1}\ kW^{-1}$ for PO and 3.31 $kg_{O2}\ h^{-1}\ kW^{-1}$ for air. Previously, Oackley (1997) had similarly mentioned that the power related costs were 25% lower for PO. However, oxygen production entails an additional cost, which is not applicable when air is used.

2.2. The Effect of Bubble Size

The efficiency of the oxygenation process depends on the oxygen transfer from the bubbly to the dissolved phase with the total DO in a vessel being closely related to the bubble motion and the hydrodynamic pattern of the fluid flow, (Torti et al., 2013). Oxygen transfer also depends on the interfacial area across which it occurs. Factors, such as the bubble size and the bubble residence time, also have an effect on oxygen transfer, (Gostick et al., 1992). Fine bubbles or microbubbles are preferable due to their small size, large interfacial area, long stagnation time and lower bubble rising speed as well as they lead to better k_La values than usual air or PO bubbles. These bubbles also deal with higher loadings and, at the same time, they form less foam, (Zhuang et al., 2016a,b). Zhuang et al. (2016b) employed MBR technology under either air or PO conditions using both usual and fine bubbles and they found that air demonstrated a worse performance. Usual bubbles, made either of air or of PO, had also a worse performance on oxygen transfer, although usual PO bubbles still performed better. However, there was also a dramatic increase in the k_La, when they switched from usual PO bubbles to fine PO bubbles. When fine bubbles were used, the gas liquid interfacial area significantly increased due to the decrease in bubble size, (Zhuang et al., 2016b). Coarse bubbles though, may be more efficient in stripping CO_2 out of a POAS system reducing the need for use of sodium hydroxide solutions, (Gostick et al., 1992).

3. EFFECT OF PURE OXYGEN ON BIOMASS

3.1. EPS and SMP Production

The type of aeration in aerobic WWT affects the extent of production of extracellular polymeric substances (EPS). In batch experiments, EPS concentrations are lower when PO is used, (Zhang et al., 2019). Zhang et al. (2019) found that at low food to microorganism (F/M) ratios, e.g. 0.05 $kg_{TOC}\ kg^{-1}_{MLSS}$ (100 mg L^{-1} TOC and 2000 mg L^{-1} MLSS), EPS production remained the same regardless of the type of aeration. In both cases, EPS initially increased reaching a peak value and subsequently decreased. This trend is expected as EPS concentration during the substrate utilization phase increases, whereas during endogenous respiration, it starts decreasing as EPS now function as substrate. At the higher F/M ratio of 0.25 $kg_{TOC}\ kg^{-1}_{MLSS}$ (500 mg L^{-1} TOC, 2000 mg L^{-1} MLSS), they observed a similar increase/decrease trend, but the EPS amount in the bioreactor fed with air was still high at the end of the batch reaction due to the high amount of total organic carbon (TOC) that was still present in wastewater.

In general, Zhang et al. (2019), claimed that PO use had an enhanced effect on EPS reduction regardless of the initial TOC concentration as well as it better promoted EPS consumption, when that was required. By maintaining the substrate concentration constant at 500 $mg_{TOC}\ L^{-1}$ and by varying MLSS concentrations from 2000 mg L^{-1} to 5000 mg L^{-1} to 8000 mg L^{-1}**,** EPS concentrations initially increased once again and then decreased, except for the highest F/M ratio as applied within the bioreactor fed with air, where a constant increase occurred. EPS rapidly decrease when the growth of microorganisms moves from the exponential increase to the stationary phase where food starts depleting, a case that was never achieved in the air related bioreactor at the highest F/M ratio. As up to 50% of the produced EPS are utilized by bacteria, PO aeration not only accelerated substrate decomposition and increased organic pollutant removal rates, but also promoted decomposition of the produced EPS when that was required, (Zhang et al., 2019).

In addition, EPS production is also affected by the applied air or PO flowrates — Zhuang et al. (2016b) found that the higher air flowrates that need to be applied and consequently the higher shear forces in an MBR fed with air produced more EPS, as flocs that are exposed to high shear forces easily break. On the other hand, the lower PO flowrates, in combination with the enhanced enzyme activity due to the higher DO concentrations, resulted in less EPS. This occurred either because of a lower EPS production or because of an improved EPS biodegradation. Pressurization, which, as already mentioned above is another way of increasing DO concentration in tanks, also lowered EPS production in tanks fed with PO, (Zhang et al., 2019), confirming the important role of DO on EPS formation.

With respect to aerobic granulation, on the other hand, PO aeration may potentially produce more EPS than air. In this case, EPS are an important ingredient for stable granules as they ensure their integrity, (Pan et al., 2017). Pan et al. (2017) showed that at 4.57 kg_{COD} m^{-3} d^{-1}, the maximum chemical oxygen demand (COD) loading rate they applied, their PO granular bioreactor had the highest amount of EPS that was equal to 193 mg g^{-1}. These EPS were more than those in their PO activated sludge bioreactor, which were more than those in the air granular bioreactor, which were more than those in the activated sludge bioreactor.

Under high organic (COD) loadings, biomass grows rapidly. Positive correlations between biomass and EPS and between influent COD and EPS and vice versa were found, as they both increase EPS. PO formed aerobic granules with some great ability of treating heavily polluted wastewaters. This was due to the retention of biomass on an EPS matrix that helped granules avoid disintegration. These granules were difficult to collapse allowing PO passing through them, so they tolerated any shocks due to the high organic loadings.

In addition, EPS production in aerobic systems is also affected by the salinity, so the combined effect of the aeration type and salinity needs to be considered. As salinity increases, the specific oxygen uptake rate (SOUR) also increases, with microorganisms needing more energy to endure the high saline conditions. In saline wastewaters, the large mass transfer resistance makes it difficult for oxygen to diffuse from the gas interface to the cell membrane as well as salinity itself also decreases oxygen solubility. CAS systems cannot cope with the increased DO concentrations required, in particular when high loadings are to be treated. PO use instead of air is then a promising alternative, (Hu et al., 2019). Hu et al. (2019) found that, in sequencing batch reactors (SBRs), PO improved the TOC removal efficiency at salinities less than 3%. Increasing salinity to values over 3.5%, the TOC removal efficiency decreased regardless of the type of aeration, however, PO still performed better than air. As salinity kept increasing, even high DO concentrations had a negligible effect on metabolism, because of the strong shock on the bacteria. EPS under either type of aeration increased as salinity increased, with PO mostly producing more EPS than air at the same salinity. In detail, at low salinities up to 1%, for both aeration types, EPS amounts were similar. For salinities above 2%, EPS at PO aeration conditions increased. PO mitigated the effect of high viscosity on oxygen transfer and promoted EPS production as a measure to protect the microorganisms from salt suppression. At a salinity of 3%, polysaccharides increased over time in both aeration types, but not the proteins. Initial concentrations of polysaccharides in both systems were lower than those of proteins, but as salinity increased, their concentration exceeded the concentration of the proteins, which remained stable. The production of polysaccharides was then the bacterial reaction against the high osmotic pressure due to salt. As such, their concentration was higher when PO was used due to the higher partial pressure, (Hu et al., 2019).

In addition, Hu et al. (2019) found that the soluble microbial products (SMP) also increased under both aeration types, as salinity increased. At the highest salinity of 5%, SMP were maximum in both bioreactors — 63.28 mg L^{-1} (PO) and 62.5 mg L^{-1} (air). At low salinities of 0.5% or 1%, SMP in the bioreactor fed with PO were more due to the sufficient degradation rate. At salinities of 2%, 3% or 4%, the opposite happened. The increased EPS production to help bacteria tolerate the saline conditions reduced the production rate of biomass associated SMP in the bioreactor fed with PO. At a salinity of 5%, the effect of salinity was dominant, as high osmotic pressures started rupturing the cells leading to higher biomass associated SMP production rates due to the EPS hydrolysis at a rate that was proportional to the EPS production.

Finally, by running batch experiments, Zhang et al. (2019) showed that at a low F/M ratio of 100 mg_{TOC} L^{-1}, SMP concentration remained stable regardless of the type of aeration. On the other hand, at a higher F/M ratio of 500 mg_{TOC} L^{-1}, SMP concentrations were always higher when PO was used. Improved oxygen transfer improved the enzyme and biomass activity, so the production of utilization associated SMP at high F/M ratios increased.

3.2. Enzyme Activity and Microbial Diversity

The enzyme activity shows the ability of bacteria to adapt themselves to environmental changes. During the formation of activated sludge, microorganisms use their enzymes, e.g. catalase, dehydrogenase, phosphatase, protease, esterase, glucosidase, to hydrolyze and biodegrade organic matter, mostly consisting of proteins and carbohydrates. Based on their variations, the physiology of the bacterial community is assessed, (Calderón et al., 2012). Enhanced enzyme activity leads to better multiplication conditions for the living microorganisms and subsequently improves the pollutant removal efficiency. However, at low DO conditions and high organic loading rates (OLRs), the enzyme activity deteriorates. PO use may then accelerate it, so it will consequently improve the microbial biomass activity as well — the increased substrate utilization rates when PO is used are interrelated with higher enzyme activity. PO aeration demonstrated higher concentrations for many of the enzymes. Microorganisms can adapt to PO aerated environments, so that the secretion of enzymes stabilizes at high values, (Pan et al., 2017; Zhuang et al., 2016a; Doviral-García et al., 2014). In addition, Zhuang et al. (2016b), by measuring SOURs, showed that when PO was used, SOUR was 4.15 $mg_{O2}\ g_{MLSS}^{-1}\ h^{-1}$, or 28% higher than the SOUR that was measured when air was used.

However, the improved biomass activity in POAS may also have an adverse effect, which needs considering, as it results in rapid DO depletion in the secondary clarifiers, which usually lack any aeration facility — a condition which can be additionally favored under higher ambient temperatures, as these temperatures further improve bacterial metabolism. In more detail, to avoid septic sludge and poor separation of solids from liquid as well as foul odours, DO levels must be always maintained at the required level. As the mixed liquor approaches the last stage in the aeration tank, DO concentrations drop and CO_2 concentrations increase. Increased CO_2 levels decrease the pH promoting the growth of filaments (that also thrive at low substrate concentrations) and fungi, which consequently hinder sludge settling and compaction of sludge. Sludge bulking will finally increase the concentrations of total suspended solids (TSS) in the effluent and cause losses of active biomass. In case oxygen becomes the limiting substrate, glucose consumption rates also increase causing sludge deflocculation. Therefore, sludge exhibits poor settling properties in the secondary clarifiers resulting in effluents of deteriorated quality. Any use of the design of and of the operational guidelines for CAS systems to POAS systems is not advisable as their microbial populations and the metabolic rates may not be similar. The challenge above must then be considered when POAS systems are to be designed, (Kundral et al., 2013; Lee et al., 2003).

Gostick et al. (1992) had also observed that at low F/M ratios, the POAS plant under their study, which was treating a vegetable processing wastewater, confronted bulking issues, as filaments growth was higher than that of floc forming bacteria. Low F/M ratios may indeed lead to the production of filaments and poorly flocculated pin flocs due to aged sludge, (Paice et al., 2003; Marshall and Sousley, 1997). On the other hand, too high F/M ratios may lead to dispersed growth, (Paice et al., 2003).

Finally, Calderón et al. (2013), by comparing the effect of the type of aeration on the performance of hydrolytic enzymes, did not detect any difference. Any increase in the pollutant removal efficiency during PO aeration was found to be unrelated to any improvement of the depolymerization of the particulate matter, (Calderón et al., 2013). This field does require further research, as some enzymes are highly affected by the redox of a system, whereas others, like protease and esterase, are not, despite being very important for the hydrolysis of macromolecules and contaminants, (Doviral-García et al., 2014).

The aeration type may also affect the microbial diversity, as the former can promote different species composition of activated sludge communities, (Zhuang et al., 2016a,b). Zhuang et al. (2016b) claimed that the bacterial communities are not the same in an MBR fed with air and an MBR fed with PO. First, based on the ten most abundant genera present in MBRs, their total relevant abundance in the MBR fed with PO was found to be higher than that in the MBR fed with air — 35.35% and 28.03% respectively. The MBR fed with PO concentrated more genera in a small fraction, indicating their adaption to higher DO concentrations. Then, although both MBRs did share a large proportion of core bacterial population, some differences were also observed, i.e. concentrations of Ohtaekwangia, which are detected during coal mine wastewater treatment, Thauera, which degrade phenol and methyl-phenols and Comamonas, an aromatic compounds degrader, increased in the MBR fed with PO, explaining also the better effluent quality. Finally, increased amounts of Phycisphaera in the MBR fed with PO additionally demonstrated its potential for nitrogen removal, (Zhuang et al., 2016b). However, when Calderón et al. (2012, 2013) compared MBRs employing PO with MBRs employing air, they concluded that the aeration type had only a negligible effect on the diversity and functionality of the bacterial community, which was particularly true when this effect had to be compared with the effects of temperature or VSS concentration. However, they also supported the fact that that type of aeration affected the bacterial community structure and differences in the relative abundance of dominant populations were once again recorded, (Calderón et al., 2012, 2013).

3.3. Foam and Froth Formation

Excessive foam formation can affect the final water quality, as bacteria trapped in foam die, so the aerobic treatment performance and the system's operational stability deteriorate. In the case of air, the large amounts of air that have to be used for sufficient DO concentrations cause foam formation, which hinders digestion and promotes biomass washout. To avoid or control this, as low flowrates as possible have to be applied, but these flowrates may deprive bacteria of the required DO. The use of PO may once again be help, (Zhuang et al., 2016a,b; Zupančič and M. Roš, 2008; Lee and Kim, 2003). Zhuang et al. (2016a,b), concluded that the improved removal efficiency in MBRs fed with PO was attributed to the prevention of foam expansion. Zhuang et al. (2016a) achieved DO concentrations of 10 mg L^{-1}, with a foam to liquid (F/L) ratio of 6%-10% for their MBR that was fed with PO and of 30%-45% for the MBR that was fed with air. Since air is only 21% O_2 in volume, the air flowrate needed to be up to five times higher leading to excessive foam formation, (Zhuang et al., 2016a,b).

CAS applications also suffer from froth caused by Nocardioform organisms. These aerobic gram positive hydrophobic filaments preferentially concentrate at the air liquid surface and produce thick viscous froth in both the aeration basins and the secondary clarifiers. Froth causes a series of problems, related to either the liquid itself or the solids handling including deterioration of effluent quality. Both CAS installations, particularly those operated at long SRTs, and POAS installations are equally affected by froth. POAS plants are affected by froth due to them containing significant surface trapping of activated sludge. One way of avoiding froth in POAS proposed Jolis et al. (2006) was the application of a low SRT and selective wasting. They found that an SRT of 0.3 days resulted in complete removal of filamentous microorganisms in two days, which subsequently allowed the POAS plant to operate successfully at an SRT up to 3 days without confronting any return of the filaments. In a further analysis, Jolis et al. (2007) additionally highlighted the importance of using anaerobic selectors in POAS to promote growth of phosphorous accumulating organisms (PAOs), which outcompete filaments. Under operating conditions favoring enhanced biological phosphorous removal (EBPR), that was at an SRT<2 d and an HRT>55 min, a decrease in filamentous organisms occurred resulting in effective froth control as well, (Jolis et al., 2007).

To this point, it may also be worth mentioning that the use of PO, on the other hand, may also be able to stimulate the growth of PAOs in EBPR systems, which alternate anaerobic and aerobic environments. Wei et al. (2014), who operated laboratory scale pressurized oxygen aeration SBRs, found that PO increased the oxygen-reduction potential (ORP) from the highly negative value required during the anaerobic treatment to the positive value required for the production of the oxidative environment for phosphorous uptake but no more details were given.

3.4. Pure Oxygen Aeration and Temperature

Special mention should be made on the effect of the type of aeration in combination with temperature. Temperature is an operating parameter that heavily affects bacterial metabolism, so its effect on biomass is more dominant than the type of aeration — any temperature changes may lead to poor sludge settling, high turbidity of the final effluent, etc., (Rodríguez et al., 2014; Bernat et al., 2017). However, Rodríguez et al., (2011), observed that in an MBR fed with PO operating at an SRT of 39.91 days, the 0.4 $g_{SS}\ g^{-1}_{COD}$ of sludge that was produced was similar to those reported in other studies, where air had had been used, however, in their study that was achieved at a lower temperature. As temperature affects sludge production with the latter decreasing as the former increases**,** their improved value can be attributed to the PO, which maintained sludge production at low levels.

A case of interest though is the aerobic thermophilic process, which is carried out at temperatures higher than 45°C. Comparer to mesophilic processes, it leads to higher biodegradation rates, inactivation of pathogens and lower excess sludge production, but to effluent of poorer quality. Effluent's higher COD and turbidity are due to the large amount of dispersed free bacteria and colloids, which hinder thermophilic sludge to settle in secondary clarifiers, (Collivignarelli et al., 2015). Indeed, Cohen (1977) had already found that high biomass reduction in an uncovered POAS system had been achieved due to the heterotrophic mesophilic bacteria. Zupančič and Roš (2008) studied the degree of degradability of excess activated sludge at different temperatures including thermophilic values by operating either aerobic or combined anaerobic/aerobic digestion. To satisfy the aerobic step, either PO or air had to be used. However, PO aeration and high thermophilic temperatures were found not to be compatible.

Aerobic sludge digestion, or the extension of CAS process under endogenous respiration conditions, requires a lot of oxygen or the process is disturbed. In the thermophilic range, permanent lack of DO is monitored in excess sludge, as the potential of oxygen for absorption is low due to poorer solubility. In addition, the oxygen demand is higher due to much higher rates of sludge digestion. Even though the mixing of sludge water, that is to say any water remaining after sludge digestion, with the main feed, does not increase the COD in the feed, it does contribute to as much as 50% of the total ammonium, (Zupančič and Roš, 2008). Sludge treated with PO by Zupančič and Roš (2008) degraded between 22°C and 50°C, whereas sludge treated with air degraded between 32°C and 65°C. When PO was employed, no sludge digestion took place above 50°C — in such temperatures, such high DO concentrations do not occur in natural environments, so bacteria were unlikely to tolerate them. On the other hand, in the mesophilic range, the PO had a better performance. Zupančič and Roš (2008) showed that both types of aeration had both advantages and disadvantages, with temperature prevailing against the aeration type. High temperatures promoted better digestion with air, something that was impossible for the PO, which at lower temperatures performed better. Finally, Collivignarelli et al. (2015), by performing ammonia utilization rate tests at 49°C using thermophilic biomass taken from a bioreactor fed with PO, showed that low nitrification rates in the range of <0.01 $mg_{N\text{-}NO3}^{-1}$ gr_{VSS}^{-1} h^{-1} were obtained, so no biological oxidation of ammonium through nitrification occurred. This also supports the need for lower temperatures during PO use.

4. EFFECT OF PURE OXYGEN ON EFFLUENT QUALITY

4.1. Organic Carbon Removal

PO aeration achieves high pollutant removal efficiencies at low oxygen flowrates. Table 3 shows the pollutant removal efficiencies of selected studies, which are high, including those of organic carbon.

TABLE 3: Pollutant removal efficiencies as indicated in a number of selected studies

However, a proper direct comparison of the relative performance of the CAS and POAS has long since been not an easy task as it needs involvement of many factors and requires complex experimental designs, (Cohen, 1977; Dirk, 1981). Any chance for advanced performance of POAS had to be solely attributed to the higher partial pressures and its ability to transfer oxygen rapidly and not on any significant differences in the intrinsic kinetic parameters and settling characteristics, (Shelef and Green, 1980). Rempel et al. (1992), by operating a pilot CAS and a pilot POAS, treating mill effluent at different sets of F/M ratios, SRTs and HRTs, showed that under similar sets of operating conditions, biochemical oxygen demand (BOD) and COD removal efficiencies between the two systems were not very different. They also pointed that the effect of operating times on the removal efficiencies was more important than that of the type of aeration.

Esparza-Soto et al. (2006a) additionally claimed that although PO increases treatment capacity of the aeration stage and produces biosolids that settle better, its benefits against air are still under discussion. PO systems, at high F/M ratios, are very successful in removing five-day BOD (BOD_5) and suspended solids (SS) as well as in producing little sludge. In addition, COD removal efficiencies in these systems also remain high, about 80% for F/M ratios up to 2.8 mg_{BOD5}/mg_{MLVSS}. However, this COD removal efficiency was also the same when air was used as well. The difference was that in the case of air, the range of F/M ratios applied was shorter and equal to only one third of that one reached by PO. For F/M ratios up to about 1.25 mg_{BOD5}/mg_{MLVSS} or 1.5 mg_{COD}/mg_{MLVSS}, both aeration types remove similar COD amounts. Nonetheless, at higher F/M ratios, a fair comparison cannot be made, as the systems fed with air have never been run at such high F/M ratios due to their restricted oxygen transfer capacity.

In addition, Esparza-Soto et al. (2006b) determined the molecular weight (MW) distribution in wastewater samples from full-scale WWT plants using different aerobic treatments and concluded that the plants generated effluent organic matter (E_fOM) with different MW dissolved organic carbon (DOC) distributions. Systems operating with air generated E_fOM with centrally clustered distribution — the intermediate MW fraction contained most of the organic matter, i.e. 50%-60%, DOC: 0.5 kDa to 3 kDa. Systems operating with PO, on the other hand, generated E_fOMs with skewed distributions towards the high MW fraction, i.e. 40%-50%. Long SRTs reduced the DOC concentration in the effluent, but the MW distribution remained unmodified. PO generated organic matter of a more refractory character. That was due to the higher DO concentrations, which increased the endogenous respiration rate and promoted the production of EPS and SMP, which are high MW refractory biopolymers. Both aeration types were then equally efficient up to certain F/M ratios, although PO systems extended operation to higher F/M loadings. However, if we aim for the production of E_fOM of better quality — less DOC with lower

MW — the systems operating with air were more efficient within the range of F/M ratios where the use of both air and PO was applicable, (Esparza-Soto et al., 2006a,b).
On the other hand, there are also studies which clearly state that the use of PO improves effluent quality. For example, Zhang et al., 2019, found different TOC biodegradation levels for PO and air, with PO showing a better performance. In addition, an increase in the MLSS concentration increased the gap between the PO and air related removal efficiencies, from 16.8% of TOC removal at 2000 mg L^{-1} to 76.5% at 8000 mg L^{-1}. Pan et al. (2017) also observed more filamentous, actinophryids and nematodes when air was used as well as the percentage of the total aerobic bacteria was higher when PO was used. However, in both cases, they carried out batch tests at small scale and, as PO acts faster than air, its use may be more advantageous with respect to operation restricted to short times. Finally, Bernat et al. (2017) found that despite the abundance of Vorticella infusionum, whose presence indicates unfavorable treatment conditions, in the mixed liquor, a stable and acceptable effluent quality was achieved in their POAS system, but this is more related to the fact that PO manages to better maintain aerobic conditions. As a conclusion, either under air or PO conditions, there seems to be no significant difference in terms of organic carbon removal. However, PO may be of help in cases where operational parameters do not support the use of air.

4.2. Removal of Phenolic Compounds and Micropollutants

Regarding phenolic compounds, PO use is quite promising. Moerman et al. (1995), by operating a POAS plant treating pretreated diluted carbonization wastewater, proved that 98% of thiocyanate, a substance whose overloading inhibits any phenol degradation, had been removed. In addition, Li and Loh (2006) found that during the cometabolism of 4-chlorophenol in the presence of phenol in an immobilized cell hollow fibre MBR enriched with Pseudomonas putida, both 4-chlorophenol and phenol degradations improved when PO was used. Finally, PO was satisfactorily used in continuous flow fluidized bed reactors for the degradation of polychlorinated phenols that are included in wood preservation chemicals, (Puhakka and Järvinen, 1992).

In addition, PO use can also help with the removal of refractory pollutants and micropollutants. CAS cannot remove antibiotics, endocrine disrupting compounds, pharmaceuticals and residual personal care products, household and industrial chemicals, etc. as CAS plants have not been designed or operated for this purpose, (Batt et al., 2007). Levine et al. (2006) assessed the persistence of a number of micropollutants during primary treatment, biological treatment comprising POAS and nitrification/denitrification and finally disinfection. Several substances tested had lower concentrations in the denitrified effluent than in the influent or the primary treatment effluent, which means that biological treatment with PO additionally helped with their removal. For instance, acetaminophen, a non-antibiotic over-the-counter pharmaceutical that had been detected at the highest concentration of 10 μg L^{-1} in the influent, was eliminated during biological treatment. Batt et al. (2007) monitored the fate of four antibiotics, i.e. ciprofloxacin (CIP), sulfamethoxazole (SMX), tetracycline (TC) and trimethoprim (TRI), in four full scale WWT plants including POAS. The use of PO had a positive effect on the removal of the antibiotics, mainly with respect to removal of SMX and TRI, despite the short HRT of 1 h that was applied. In all plants tested, removal efficiencies were strongly related to the operating times. However, by comparing the performance of the POAS plant with that one of the rotating biological contactor (RBC) plant, it was found that, although similar removal efficiencies for all antibiotics were reported for both plants, the HRT at POAS was four time less than that at RBC.

Bae et al. (2015) also assessed the contribution of PO aeration of a combined biological and physicochemical treatment (POAS + Fenton Process) system to the removal of refractory pollutants from dyeing wastewater, which is not a readily biodegradable wastewater as well as any potential improvement with respect to biological treatment of dyes needs long SRTs and high MLSS concentrations. Although the biological treatment system suffered from low MLSS concentrations due to inefficient settling, it managed to remove 53% of soluble COD (SCOD) and 12% of color indicating that the microorganisms in the aeration tank were acclimated to the dyeing wastewater. As such, Bae et al. (2015) managed to obtain a cost effective pretreatment, which both helped the Fenton Process in becoming more efficient and decreased the consumption of chemicals.

Martín-Rilo et al. (2018) employed PO aeration for the removal of a benzotriazole based anticorrosive from dairy wastewater with benzotriazole being an aromatic compound used as a metal corrosion inhibitor and an emerging toxic that tends to bioaccumulate. PO was injected in the intermediate step (Step 2) of a treatment process — that step was preceded by wastewater neutralization under CO_2 injection (Step 1) and followed by coagulation/flocculation in a dissolved air floatation tank (Step 3). Steps 1, 2, and 3 removed 44%, 30%, and 25% of the total benzotriazole respectively for an overall removal of 99.7%. The respective removal efficiencies of each step considering the concentration of the contaminant entering each step was 44%, 53%, and 95.6% respectively, so more than half of the amount of the chemical that entered Step 2 was removed.

Finally, PO aeration may also be beneficial regarding the removal of endocrine disruptive compounds from wastewater. For example, bisphenol-A, the endocrine disrupting chemical of the greatest concern, due to its effects being more detrimental than those of other substances of the same kind, cannot be removed by CAS. In addition, its removal does not necessarily guarantee its complete degradation, as, due to its hydrophobic nature, it is also be stored in the sludge or, where possible, it can be adsorbed by membranes, (Doviral-García et al., 2014). Dorival-García et al. (2014) assessed then the fate of bisphenol-A by employing MBR technology to prolong SRT, and they found that bisphenol-A reached the background level in the effluent in five days when PO was used, whereas it took 10 days in the case of air. By switching from air to PO, they also improved the biodegradation percentages from 60.2% to 87.4% in 10 days. With air, bisphenol-A that was sorbed in the sludge, remained within it, so its availability for biodegradation was reduced and the need for further sludge treatment before sludge disposal in order to remove the sorbed contaminant increased. On the other hand, with PO, the sorption tendency diminished and desorption was facilitated — 67.4% of bisphenol-A in the sludge underwent desorption and consequently biodegradation.

4.3. Removal of Volatile Organic Compounds

PO aeration is also highly capable of handling foul condensates without stripping or of biodegrading VOCs. To this end, Freitas dos Santos and Livingston, (1993a,b) proposed a gas enclosed recirculation system based on a bioreactor fed with PO, whose design was similar to that of a an air lift bioreactor, for the aerobic degradation of the 1,2-dichloroethane in 1,2-dichoroethane contaminated wastewater. Air stripping of 1.2-dichloroethane that usually takes places during its aerobic treatment was avoided — VOC emissions are known to cause immediate toxicity and odor and promote undesired chemical reactions. In their conventional air lift bioreactor, 33% of the 1,2-dichloroethane was lost due to stripping, whereas in the system where PO was used, the whole of 1.2-dichloroethane was mineralized.

In addition, Paice et al. (2003) mentioned that one of the selling points of POAS systems for kraft pulp mills was their ability to treat their foul condensates without any stripping, even from the start-up. However, these systems were liable to some problems, which were mainly directly related to their enclosed design. As such, the potential for high concentrations of dissolved CO_2 in the effluent, the potential for combustible gas alarms caused by VOCs in the condensates and possible premature corrosion of the concrete, which was associated with the bacteria that reduce sulfuric compounds, had to be carefully considered. These problems can also be additionally intensified, as this design further complicates the inspection and maintenance of the systems, (Paice et al., 2003).

4.4. Effect of Pure Oxygen on Heavy Metals

Heavy metals are non-biodegradable toxic substances that interact with the biomass and have various inhibitory or toxic effects on bacteria, with nitrifying autotrophs being more sensitive to them than heterotrophs, (Avezzù et al., 1995). Avezzù et al. (1995) assessed the fate of heavy metals during treatment of leachates by monitoring their distribution in the solid or liquid phase of the biological process and showed that, despite their accumulation in the biomass, BOD_5 and COD removal efficiencies remained quite high. As such, PO aeration created a quite favorable living environment for the microorganisms, which, by making microorganisms more resistant to compounds like heavy metals, managed to maintain considerable removal of organic carbon, (Avezzù et al., 1992).

5. PURE OXYGEN AND NITRIFICATION

5.1. Introduction

Ammonia that is not removed during WWT can cause a number of adverse environmental impacts, when effluent is discharged into the receiving water bodies. These include eutrophication, DO depletion and toxicity to aquatic organisms. Removal of nitrogen occurs during nitrification and denitrification. Aerobic autotrophic nitrification is an oxygen demanding process comprising two phases: i) ammonium is oxidized to nitrite**,** usually by Nitrosomonas, and ii) nitrite is oxidized to nitrate by Nitrobacter. During denitrification, nitrite and nitrate are reduced to nitrogen gas, (Neerackal et al., 2016; Rodríguez *et al.*, 2012c). High DO levels reduce the competitive stress upon the autotrophic nitrifying bacteria, with a constant DO concentration of 4 mg L^{-1} being high enough to achieve nitrification, (Moerman *et al.*, 1995). Nitrification is a complex process where PO aeration is evaluated together with other parameters like pH and temperature. Nevertheless, DO concentration is still used to control nitrification, particularly in the cases of high ammonia loading rates or low temperatures, (Bonomo *et al.*, 2000). DO concentration also controls nitrification phases as concentrations <0.5 mg L^{-1} ensure stable inhibition of nitrite oxidizing bacteria, (Canziani *et al.*, 2006).

5.2. The effect of pH

POAS is more vulnerable to low pH inhibition of nitrification than CAS because of their closed headspace design, whose main role is to minimize oxygen losses by recycling headspace gas. Because of the aerobic treatment, the headspaces can contain elevated amounts of CO_2, which under slightly increased pressure moves into the mixed liquor reducing the pH, whose degree of reduction depends on parameters like the system's buffer capacity or the degree of venting. This decrease affects the kinetics of enzyme reactions, the bacterial species predominance and the physical properties of the organisms and particles. When nitrification has to take place, pH reduction additionally inhibits it, unless acclimation has already taken place. As nitrification proceeds, alkalinity is consumed, which further reduces the pH — entrapped CO_2 does not reduce alkalinity itself but it does reduce the pH at a given alkalinity. Many POAS systems were originally designed only for removal of organic carbon, therefore to operate at low SRTs and HRTs ranging from 1.5 h to 2.5 h. In practice, nitrification could proceed at lower pH values in covered POAS systems, however, this requires contact times up to 3.5 h-5 h, which are rather longer than those usually applied to handle carbon. To resolve this, another process step may then have to be added, (Garber, 1977; Dirk, 1981; Shelef and Green, 1982; Sear et al., 2003). In addition, to reinstate nitrification that had been hindered by a pH of 6.5, Mauret et al. (2001) either alternated aeration with air and PO or combined the two. Nitrification that was non-existent at pH values <6 could also have been held if acclimation had preceded, e.g. Sears et al. (1995) found that nitrification in POAS was stable at pH values between 5 and 5.5 provided that the required acclimation period had been applied.

As combined carbonaceous and nitrogenous removal in POAS systems that are designed to remove only organic carbon is difficult, particularly in enclosed ones, Bonomo et al. (2000) proposed as an alternative the use of MBBRs aerated with PO for tertiary nitrification of the secondary effluent. The extra treatment step eliminated any competition between heterotrophic and autotrophic bacteria and PO aeration increased nitrification without requiring a certain increase in the thickness of the biofilm. They then managed to acquire high nitrification rates both in ammonia and in oxygen limiting conditions. Maximum efficiencies were seen at lower ammonia loading rates. Efficiencies higher than 90% were also achieved at ammonia loading rates higher than 4 g_N m^{-3} d^{-1} provided that the DO concentration was higher than 10 mg L^{-1}-15 mg L^{-1}, clearly stating the positive contribution of the DO use. Finally, alteration of PO use with air use, and vice versa, depending on the pH value, may be able to simultaneously treat carbon and ammonia in one basin and this seems to be technically feasible, (Mauret et al., 2001).

5.3. The Effect of Temperature

The effect of temperature on nitrification is complicated, as low temperatures reduce nitrification but increase oxygen's solubility. During nitrification in a POAS system at 12°C, Sears et al. (1995) proved that the HRT did not have any effect on the specific nitrification rates but the temperature controlled the process instead. Increasing the temperature at 24°C, an HRT of 4 h functioned better than that of 2.5 h. By adding an anoxic reactor, the pH increased as the alkalinity managed to recover due to denitrification — i.e. 3.57 mg_{CaCO3} was produced per mg_{NO3-} that was reduced. The anoxic tank itself did benefit nitrification, however, even without it nitrogen loss happens in aerobic systems to some extent — anoxic conditions can occur inside the flocs or due to some bacterial species that can perform denitrification under aerobic conditions, Sears et al., 2003, 1995). Indeed, Neerackal et al. (2016), by using Alcaligenes faecalis strain No. 4, which have the ability to turn ammonium in wastewater to nitrogen in one single aerobic process, showed that within a batch operation of 24 h, total ammonium removal from their dairy wastewater was about

100% when the reactor's headspace was flushed with PO and 42% when it was flushed with air.

Salvetti et al. (2006) also operated MBBRs fed with PO to monitor the combined effect of the temperature and the type of aeration on nitrification. PO diffuses more deeply into the biofilm than air, so it produces higher nitrification rates, hence, requiring smaller reactor volumes, (Salvetti et al. 2006). At low ammonium concentrations, Salvetti et al. (2006) found PO was not essential as air could provide the required DO and the temperature did not have any significant effect on the nitrification rates. As ammonium concentration increased, DO became the reaction limiting substrate (this occurs even at DO concentrations as high as 5 mg L^{-1} or 10 mg L^{-1}), so PO use was preferred. These findings are also in line with the findings of Bonomo et al., (2000) mentioned in Section 5.2. Under oxygen limiting conditions, the specific biomass activity, as the ratio of nitrification rate to biomass content on the support media, was higher between 23°C-28°C than between 18°C-22°C. By removing the effect of DO on biomass, whose solubility also decreases as temperature increases, that was further attributed to the higher temperature that favors nitrification and to the reduced resistance to diffusion, which allows more biomass to have access to DO.

5.4. The Effect of Operational Times

The role of SRT is important for POAS, as longer SRTs favor nitrification, (Sears et al., 2003). As nitrifiers grow slowly, their growth is strongly related to the applied SRT. Due to the high treatment rates that are achieved thanks to their high OTRs, POAS plants are usually operated at short sludge ages ranging from 1 day to 3 days, which are not long enough for nitrification, (Neethling et al., 1998). To overcome this, Neethling et al. (1998) transferred aged waste activated sludge from a CAS plant to a POAS plant to seed it with nitrifiers. This increased the "apparent" sludge age and the high loaded POAS plant achieved nitrification. After the seeding process, they found that ammonium concentration in the effluent decreased to 5 mg L^{-1} within 3 days stabilizing to values below 0.5 mg L^{-1} after about a month. Similarly, Randall and Cokgor (2001) applied lower flows and continuous seeding to maintain nitrification to a full scale POAS system.

In addition, Rodriguez et al. (2012c) studied the effect of HRT on nitrification under either PO or air conditions by employing MBR technology for the treatment of primary clarifier effluent. They showed that, by using PO instead of air, nitrogen removal efficiency was improved by 8% at an HRT of 12 h and by 13.5% at 18 h. Kinetics**,** as per the Monod's model and with ammonium being the substrate, also supported that result. At similar temperatures and MLSS concentrations, the K value, or the half-saturation constant for the PO increased by 190.4% at 18 h and by 324.1% at 12 h. However, this research did not analyze the complex relationship between the K value, the MLSS concentrations and the applied HRT. When air was used, the k_H value, or the hydrolysis constant, decreased and the k_d value, or the decay constant, increased, which additionally stated the advantage of the PO use, which was further strengthened at longer HRTs. In addition, by employing oxidation of anaerobically treated excess sludge with PO, Zupančič and Roš (2008) showed that at an HRT of 5 days only 38% of ammonium was converted. This increased to <70% at HRTs of 6 days or 7 days, to 85.1% at 8 days and finally 85.6% at 10 days indicating that no further improvement was possible. As such, the nitrification bacteria needed longer contact times to process large loadings of ammonium.

6. PURE OXYGEN AERATION AND MEMBRANE FOULING IN PURE OXYGEN MEMBRANE BIOREACTORS

As membrane fouling continues being an important research field in membrane bioreactors, in this section, we mainly concentrate on the potential effect of PO aeration on its mitigation during WWT. Despite the fact that the performance of MBRs operating at low/medium COD loading rates, e.g. <1 kg m^{-3} d^{-1}, has been widely studied, operation at COD loading rates >2 kg m^{-3} d^{-1} is not common. This is because of the incapability of maintaining a healthy aerobic environment, due to limited oxygen transfer efficiency, (Lee and Kim, 3003). As such, PO aeration suits well MBRs where high MLSS concentrations have to be maintained and high organic loadings have to be treated, so OURs as high as 50 mg_{O2} L^{-1} h^{-1} - 150 mg_{O2} L^{-1} h^{-1} are needed, (Larrea et al., 2014).

Although MBRs have many advantages over CAS, membrane fouling is still a problem that increases running cost and potentially reduces treated water volumes and quality, (Zhuang et al, 2016b). Membrane fouling in MBRs is the systematic accumulation of suspended solids, colloids, and macromolecules on the membrane surface, or inside the pores, causing a reduction in membrane permeability. It is a complex phenomenon that is affected by the membrane itself, the wastewater under treatment, etc. with viscosity of wastewater, EPS/SMP production, size distribution of sludge flocs and the presence of colloids contributing to this. An accurate indicator of the degree of membrane fouling is the transmembrane pressure (TMP), (Zhuang et al., 20016b; Lee and Kim, 2003).

With respect to the aeration type, Zhuang et al. (2016b) showed that the degree of membrane fouling was 33% less (on day 90) when an MBR was fed with PO, instead of air. Even though it had been expected that the higher air flowrates and the larger bubble size would have improved scouring and led to less fouling, it was finally proved that the PO fine bubbles controlled EPS production and the biomass particle size distribution better mitigating membrane fouling. Despite the fact that EPS amounts increased under both aeration types, their amount was lower when PO was used. High shear forces and high aeration intensities when air was used released more EPS deteriorating membrane fouling. Shear forces affect floc size, strength and compactness affecting sludge filterability. When PO was used, the flocs were constantly exposed to low shear forces, so they grew into loose weak ones. On the other hand, when air was used, the high shear forces broke flocs to fragments promoting membrane fouling, (Zhuang et al., 2016b).

Lee and Kim (2003) also operated an MBR fed with PO at 2 $kg_{COD}\ m^{-3}\ d^{-1}$ and the critical TMP (50 kPa) was reached in 50 days. By increasing the OLR to 4 $kg_{COD}\ m^{-3}\ d^{-1}$, it took 20 days, to 8 $kg_{COD}\ m^{-3}\ d^{-1}$ 10 days and finally to 10 $kg_{COD}\ m^{-3}\ d^{-1}$ less than 10 days. The membrane fouling rates demonstrated a trend twice as high as the organic loadings, however, after applying 8 $kg_{COD}\ m^{-3}\ d^{-1}$, the slope of membrane fouling rate decreased. As the organic loading increases, biomass also increases, so the F/M ratio is affected. The F/M ratio has additionally an effect on membrane fouling, but potential differences in MLSS concentrations and membrane fluxes at different F/M ratios have a complex effect, which is difficult to determine. For MLSS concentrations up to 10000 mg L^{-1}, Kim et al. (2003) found that the increase in TMP was smooth. However, it dramatically changed when the MLSS concentration exceeded 10000 mg L^{-1}, as the fouling rate was almost four times higher — it increased from 1.3 to 5.6. This became ten times higher, when the MLSS concentration exceeded 15000 mg L^{-1}, as it increased from 1.3 to 12.5.

MLSS concentrations directly affect viscosity. High viscosity suspensions require high cross flow velocities to create turbulence. If turbulence provided by aeration is not enough to scour solids off the membrane, membrane becomes fouled faster. An optimal MLSS concentration in relation with F/M, viscosity and structure/size of flocs seems to exist. In reality, membrane fouling is more affected by an increase in MLSS concentration than an increase in the organic loading. However, it is difficult to evaluate membrane fouling by disconnecting the two, as each time the organic loading increases, the biomass concentration increases as well. Therefore, it depends on whether membrane fouling is assessed from an organic loading or a biomass point of view. However, membrane fouling does not start building up soon after an increase in the organic loading but it takes some time due to biomass also needing time to increase and produce EPS at a sufficient amount. However, it must be pointed that the opposite can also be found in the literature, namely MLSS concentrations have no effect or even have a positive one on TMP. This can indeed happen, as the effect of MLSS concentrations on the filtration resistances is case-specific, (Lee and Kim, 2003).

Finally, Rodriguez et al. (2012a) quantified the influence of PO or air on the recovery of permeability of the membrane, namely the fraction of the difference of permeability, which is the quotient of flux over TMP, after cleaning minus the permeability before cleaning over permeability after cleaning. As such, a physical cleaning based on backflush of permeate and a chemical one, which becomes necessary when irreversible fouling needs to be removed, were tested. The mean recovery was found to be higher when PO was used regardless of the type of cleaning — 2% and 15% further improvement for the physical and the chemical cleaning respectively, which is also in line with the fact that physical cleanings are weaker than the chemical ones.

7. DISCUSSION AND FUTURE RESEARCH DIRECTIONS

PO aeration in aerobic WWT treatment, leads to higher OTEs at lower flowrates. It allows faster treatment under higher MLSS concentrations and shorter HRTs. In fact, it can promote treatment in cases where conventional aeration fails. EPS production was found to be affected by the type of aeration. Under substrate utilization conditions, PO leads to the production of more EPS, whereas under endogenous respiration, decomposition of produced EPS is promoted. The latter additionally supports the findings that MBRs fed with PO end up with less EPS than MBRs fed with air, as MBRs operate at longer SRTs to reduce sludge production. However, the less EPS in MBRs fed with PO are also due to the lower PO flowrates, compared with equivalent air flowrates in MBRs fed with air, as the lower PO flowrates reduce shear forces and flocs do not break. PO aeration under substrate utilization conditions additionally improves aerobic sludge granulation, as PO increases EPS production so that granules difficult to disintegrate are created. SMP production under high F/M ratios is also higher when PO is used, as higher amounts of utilization associated SMP are produced. On the other hand, the production of much larger MW biomass associated SMP, which mainly comprise the effluent soluble organic matter, at low/medium F/Ms, explains the higher refractory character of effluents, when PO is used. Enzymatic activity is accelerated during PO aeration. However, not all enzymes are benefited by the use of PO, important hydrolytic enzymes remain unaffected by the aeration type. In addition, the type of aeration does not significantly affect the bacterial diversity as well, however, it affects the relative abundance of the dominant bacteria. Finally, PO aeration also satisfactorily controls foam formation in aeration tanks, due to the lower flowrates needing to be applied.

Regarding removal of contaminants and treated water quality, PO aeration was proven quite efficient. However, after comparing performances of CAS and POAS systems with respect to carbonaceous matter removal, it was found that the type of aeration was not so critical as the HRT or the SRT. In addition, at low/medium F/M ratios, where both activated sludge systems can be equally applicable, no significant differences in terms of organic carbon removal are reported. The advantage of the use of PO though is that operation of POAS plants can be extended to higher F/M ratios where CAS plants are not usually designed to operate. Even though there is some research, which claims that, under similar operating conditions, PO aeration may lead to improved removal of organic carbon, this research is currently restricted to batch applications, e.g. SBRs, at small scale, where any improved removal efficiency can be attributed to the fact that PO acts faster than air. However, PO use may be more promising with respect to the removal of refractory pollutants and micropollutants. Under PO aeration, improved biological performance has been monitored with respect to removal of phenolic micropollutants, antibiotics, endocrine disrupting compounds, etc. Finally, organic matter removal in POAS plants remains unaffected by the presence of toxic heavy metals in the wastewater.

Regarding nitrification, it mainly depends on parameters other than the type of aeration, however, under conditions that undisputedly do not inhibit nitrification, higher DO concentrations due to PO, improved the removal of ammonium from wastewaters.

Nonetheless, POAS systems are designed to operate at short SRTs and remove only organic carbon, as the short SRTs hinder nitrification. This can be overcome by continually seeding the aeration tanks with aged sludge. In addition, under conditions where PO alternation with air, or vice versa, is possible, simultaneous removal of carbon and ammonia in one basin may also possible. If an extra treatment step for successful nitrification cannot be avoided, MBBRs fed with PO have been suggested for tertiary nitrification of secondary effluent. In any other event, MBRs, which, by definition, operate at longer SRTs, have to be considered, with MBRs fed with PO achieving better nitrification rates than those fed with air.

Regarding MBRs fed with PO, they may be proven to be quite useful in cases where high organic loadings are combined with high MLSS concentrations. MBRs fed with PO remove satisfactorily both carbonaceous and nitrogenous matter as well as micropollutants. In addition, the use of PO contributes to membrane fouling mitigation. In MBRs fed with PO, the lower flowrates that have to be applied control better any increase in TMP values, as the lower shear forces do not break flocs, so less EPS are produced. However, as organic loadings increase, MBRs fed with PO can become more vulnerable to membrane fouling. Finally, better recovery of permeability is achieved in MBRs fed with PO than in MBRs fed with air after application of a cleaning, with chemical cleanings having always a better performance than physical cleanings regardless the type of aeration.

Even though direct comparison of the two aeration types is not an easy task, there are cases where the use of PO does have a significant advantage over air, e.g. when high strength wastewaters have to be treated. In these cases, PO use is unavoidably recommended. However, the field still needs investigation, as the available literature is currently dominated by the use of air. Based on what most specifically has been found in this work, future research should concentrate on whether or not POAS can remove contaminants that usually are not removed by CAS and on analyzing further the effect of PO on the enzyme activity as all enzymes are not equally affected by the type of aeration. Finally, the potential for operation of MBRs fed with PO at higher organic loadings and higher MLSS concentrations has to be further determined.

8. CONCLUSION

PO achieves faster treatment rates at higher biomass concentrations and shorter HRTs. It better controls EPS/SMP production, accelerates enzyme activity, produces less sludge and minimizes foam. In MBRs fed with PO, it also better controls membrane fouling and improves recovery of permeability after cleanings. PO is recommended when high strength wastewaters are treated. However, PO use has also been connected with some problems, such as the pH drop in the mixed liquor in the closed headspace POAS systems. Finally, PO may also produce final effluents of a higher refractory character. It is recommended that PO be considered where air fails.

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TABLE 1: Research studies based on PO aeration

Case Study	Type of Wastewater	Scale
Bébin and Renaudin, 1976	Dairy Industry Waste Effluent	Pilot
Calderón et al., 2012	Primary Effluent	Pilot
Canziani et al., 2006	Leachate	Pilot
Collivignarelli et al., 2015	Industrial Wastewater	Full
Dorival-García et al., 2014	Domestic Wastewater	Pilot
Esparza-Soto et al., 2006	Primary Effluent	Bench
Hu et al., 2019	Hypersaline Wastewater	Bench
Lee and Kim. 2003	Synthetic Wastewater	Bench
Lee et al. 2003	Municipal + Industrial Wastewater	Pilot
Mauret et al. 2001	Food Processing Wastewater	Full
Moerman et al. 1995	Pretreated Carbonization Wastewater	Full
Neethling et al., 1998	Municipal Wastewater	Full
Pan et al., 2017	High-Loading Wastewater	Bench
Peterson et al., 1978	Mill Effluent	Full
Rempel et al., 1992	Bleached Kraft Pulp Mill Effluent	Pilot
Rodríguez et al. 2010	Primary Effluent	Bench
Sears et al., 2003	Primary Effluent	Bench
Verstraete, 1980	Petrochemical Water	Full
Zhang et al., 2019	Synthetic Wastewater	Bench
Zhuang et al. 2016a,b	Coal Gasification Wastewater	Bench
Zupančič and Roš, 2008	Excess Sludge	Bench

TABLE 2: Industrial WWT POAS applications

Reference	Application	Flowrate or Capacity
Batt et al., 2007	Lackawanna WWT Plant, Erie County, New York, USA	17 034 m^3 d^{-1}
Brand et al., 2019	WWT Plant near Stanford, Northern California, USA	240 000 m^3 d^{-1}
Collivignarelli et al., 2015	WWT Plant, Northern Italy, Italy	280 m^3 d^{-1} (Average)
Confer et al., 1995; Esparza-Soto et al., 2006a	Ina Road Water Pollution Control Facility, Tucson, Arizona, USA	N/A
Gostick et al., 1992	Christian Salvesen (Food Services) Ltd Bourne, Lincolnshire, UK	3303 m^3 (Basin Volume)
Jolis et al., 2006	Oceanside Water Pollution Control Plant, County and City of San Francisco, California, USA	0.7 m^3 s^{-1} (Average DWF) 2.8 m^3 s^{-1} (Average WWF)
Karibayashi, 1992	Todoroki Sewerage Works, Kawasaki, Japan	395 500 m^3 d^{-1}
Kundral et al., 2015	South District WWT Plant, Miami-Dade County, Florida, USA	4.9 m^3 s^{-1}
Loiacono et al., 1992	Southeast Water Pollution Control Plant (SEP), San Francisco, USA	3.5 m^3 s^{-1}
Marshal and Sousley, 1997	Simpson Tacoma Kraft Effluent Treatment Facility	1.5 m^3 s^{-1} (Design Load) 1.7 m^3 s^{-1}(Max. Load)
Mauret et al., 2001	Food-Processing Industry WWT Plant (Slaughterhouse), France[a]	3500 PE
Mines, 1992	Main Street Wastewater Treatment Plant, Pensacola, Florida, USA	0.9 m^3 s^{-1}
Moerman et al., 2008	N/A	80 m^3 h^{-1} (after 50% Dilution)
Neethling et al., 1998	Rock Creek WWT Plant, Portland, Oregon, USA	3500 m^3 (Basin Volume)
Peterson et al., 1978	Longview Fiber's Mill Treatment System, Longview, Washington, USA	2.6 m^3 d^{-1}
Randal and Cokgor, 2001	HENRICO County, Virginia, Water Reclamation Facility, USA[b]	170 325 m^3 d^{-1}
Sears et al., 1995	North End Wastewater Pollution Control Centre, Winnipeg, Canada	N/A
Verstraete, 1980	BP Chemicals Belgium Works WWT, Belgium	3000 m^3 d^{-1}

Abbreviations: DWF: Dry Weather Flow, N/A: Non-Applicable, PE: Population Equivalent, UK: United Kingdom, USA: United States of America, WWF: Wet Weather Flow. [a]Alternating Anoxic-Aerobic Process, [b] Biological Nutrient Removal System

TABLE 3: Pollutant removal efficiencies as indicated in a number of selected studies

Case Study	Treatment Method	Pollutant	Removal Efficiency: Air (%)	Removal Efficiency: PO (%)
Bae et al., 2015	Activated Sludge + Fenton Oxidation Unit	SCOD		66
		Color		73
Bonomo et al., 2000	MBBR	NH_4^+-N		>80
Collivignarelli et al., 2015	MBR + NF Unit	COD		90
Dorival-Garcia et al., 2014	MBR	COD		90
		TSS		97
		TN		90
Hu et al., 2019	Activated Sludge[a]	TOC	28.8% (Salinity: 0.5%)	78.1 (Salinity: 0.5%)
			13.3% (Salinity: 5%)	17.2 (Salinity: 5%)
Lee and Kim, 2003	Anoxic Reactor + MBR	COD		up to 99.7 (Synthetic)
				up to 97.5 (Leachate)
Pan et al., 2017	Activated Sludge	COD (Granules)	~80	91
		NH_4^+-N (Granules)	>80	80
		COD (Flocs)	>80	<80
		NH_4^+-N (Flocs)	~75	~80
Rempel et al., 1992	Activated Sludge	BOD_5	79 - 96	71 - 94
		COD	27 - 50	29 - 56
Rodriguez et al., 2010	MBR	COD		>90
		BOD_5		>90
Rodriguez et al., 2012c	MBR	NH_4^+-N	65.5±11.9 (HRT:18 h)	79.1±9.7 (HRT:18 h)
			59±20.1 (HRT: 12 h)	67±8.7 (HRT: 12 h)
Sears et al., 2003	Activated Sludge	NH_4^+-N		>90
Zhang et al., 2019	Activated Sludge[a]	TOC	35.9	87.3
Zhuang et al., 2016a	MBR	COD	52	90
		TPh	50	95
Zupančič and M. Roš, 2008	Aerobic Treatment of Sludge	COD	55 (18 d), 55 (23 d)	55 (18 d), 60 (23 d)
		VSS	60 (21 d)	60 (39 d)

Abbreviations: BOD_5: Five-day Biological Oxygen Demand, COD: Chemical Oxygen Demand, HRT: Hydraulic Residence Time, MBBR: Moving Bed Biofilm Reactor, MBR: Membrane Bioreactor, NF: Nanofiltration, NH_4^+-N: Ammonium Nitrogen, SCOD: Soluble COD, TOC: Total Organic Carbon, TN: Total Nitrogen, TPh: Total Phenols, VSS: Volatile Suspended Solids. [a]Batch Reaction