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Originally published:

May 2020

Bioresource Technology 312(2020), 123595

DOI: https://doi.org/10.1016/j.biortech.2020.123595

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

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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

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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

71 municipal effluent treatment were introduced in 1968, (Zhang et al., 2019; Calderón et al.,
72 2013, 2012). To date, various kinds of wastewater have been treated by treatment systems
73 incorporating PO aeration, (Zhang et al., 2019), including industrial wastewaters that
74 contain toxic and refractory contaminants, (Zhuang et al., 2016b), and landfill leachates,
75 (Canziani et al., 2006). In addition, PO has already been used in MBRs, (Rodríguez et al.,
76 2010), biofilms (BFs) and moving bed biofilm reactors (MBBRs), (Salvetti et al., 2006), or
77 in aerobic granulation applications, (Pan et al., 2017). As of 2014, in the USA, as much as
78 15% of all municipal WWT was realized with the use of PO in aerobic WWT plants,
79 (Larrea et al., 2014). Table 1 shows research studies based on PO aeration and Table 2
80 shows large scale POAS applications.
81
82 **TABLE 1:** Research studies based on PO aeration
83
84 **TABLE 2:** Industrial WWT POAS applications
85
86 Researchers have been comparing the performances of the two aeration types since 1976,
87 (Esparza-Soto et al., 2006a). PO increases the driving force for oxygen transfer and the
88 degree of oxygen saturation possible — its partial pressure is 4.7 times of that of its
89 atmospheric counterpart. It improves the OTR and maintains high DO concentrations at
90 lower flowrates even when high strength or toxic wastewaters have to be treated. Compared
91 with the use of air, PO provides higher gas phase oxygen concentrations, improves
92 biokinetics and allows for faster treatment rates at higher MLSS concentrations and shorter
93 hydraulic residence times (HRTs). Designs of PO based systems are simple and compact
94 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

143 PO, the amount of oxygen transferred is higher for the PO due to its higher driving force.
144 PO has a DO saturation concentration that is up to five times higher than that of air at the
145 same flowrate, (Zhuang et al., 2016a; Rodriguez et al., 2013). Lee and Kim (2003), who
146 monitored the DO concentration changes for PO flowrates of 0.0125 L min^{-1}, 0.025 L min^{-}
147 ^{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
148 that when the flowrate changed from 0.0125 L min^{-1} to 0.025 L min^{-1}, a significant sudden
149 increase in the OTR occurred. For the next flowrates, the DO concentrations continued
150 increasing but more gradually. Similarly, Zhuang et al. (2016a) noticed that, when a PO
151 flowrate of 0.5 L h^{-1} increased to 1.5 L h^{-1}, the DO concentration at a specific time of 300 s
152 was 1.8 times higher. In addition, by comparing PO flowrates with air ones, they also found
153 that at a PO flowrate of 2.5 L h^{-1} the DO concentration was five times higher than that for a
154 similar air flowrate. However, there was a limit regarding the DO amount that could be
155 sustained, so any further increase in the PO flowrate could not increase the OTE value
156 further.
157 With regard to the α-factors, Rodríguez et al. (2014), who determined the α-factors in an
158 MBR fed either with PO or air found that, at a constant HRT, the α-factor increased when
159 MLSS concentrations decreased but, at the same MLSS concentration, the PO related α-
160 factor values were higher. Similarly, Rodriguez et al. (2011) showed that, at a constant
161 HRT (and SRT), an increase in the MLSS concentration from 3420 mg L^{-1} to 12600 mg L^{-1}
162 in an MBR fed with PO decreased the α-factor from 0.426 to 0.022. However, despite the
163 decrease in the α-factor at the high MLSS concentration, the removal efficiency of organic
164 matter remained high. This suggests that PO did maintain the aerobic conditions within the
165 MBR even at high MLSS concentrations. In practice, the effect of MLSS concentrations
166 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

215 production remained the same regardless of the type of aeration. In both cases, EPS initially
216 increased reaching a peak value and subsequently decreased. This trend is expected as EPS
217 concentration during the substrate utilization phase increases, whereas during endogenous
218 respiration, it starts decreasing as EPS now function as substrate. At the higher F/M ratio of
219 0.25 $kg_{TOC}\ kg^{-1}_{MLSS}$ (500 mg L^{-1} TOC, 2000 mg L^{-1} MLSS), they observed a similar
220 increase/decrease trend, but the EPS amount in the bioreactor fed with air was still high at
221 the end of the batch reaction due to the high amount of total organic carbon (TOC) that was
222 still present in wastewater.
223 In general, Zhang et al. (2019), claimed that PO use had an enhanced effect on EPS
224 reduction regardless of the initial TOC concentration as well as it better promoted EPS
225 consumption, when that was required. By maintaining the substrate concentration constant
226 at 500 $mg_{TOC}\ L^{-1}$ and by varying MLSS concentrations from 2000 mg L^{-1} to 5000 mg L^{-1} to
227 8000 mg L^{-1}**,** EPS concentrations initially increased once again and then decreased, except
228 for the highest F/M ratio as applied within the bioreactor fed with air, where a constant
229 increase occurred. EPS rapidly decrease when the growth of microorganisms moves from
230 the exponential increase to the stationary phase where food starts depleting, a case that was
231 never achieved in the air related bioreactor at the highest F/M ratio. As up to 50% of the
232 produced EPS are utilized by bacteria, PO aeration not only accelerated substrate
233 decomposition and increased organic pollutant removal rates, but also promoted
234 decomposition of the produced EPS when that was required, (Zhang et al., 2019).
235 In addition, EPS production is also affected by the applied air or PO flowrates — Zhuang et
236 al. (2016b) found that the higher air flowrates that need to be applied and consequently the
237 higher shear forces in an MBR fed with air produced more EPS, as flocs that are exposed to
238 high shear forces easily break. On the other hand, the lower PO flowrates, in combination

239 with the enhanced enzyme activity due to the higher DO concentrations, resulted in less
240 EPS. This occurred either because of a lower EPS production or because of an improved
241 EPS biodegradation. Pressurization, which, as already mentioned above is another way of
242 increasing DO concentration in tanks, also lowered EPS production in tanks fed with PO,
243 (Zhang et al., 2019), confirming the important role of DO on EPS formation.
244 With respect to aerobic granulation, on the other hand, PO aeration may potentially produce
245 more EPS than air. In this case, EPS are an important ingredient for stable granules as they
246 ensure their integrity, (Pan et al., 2017). Pan et al. (2017) showed that at 4.57 kg_{COD} m^{-3} d^{-1},
247 the maximum chemical oxygen demand (COD) loading rate they applied, their PO granular
248 bioreactor had the highest amount of EPS that was equal to 193 mg g^{-1}. These EPS were
249 more than those in their PO activated sludge bioreactor, which were more than those in the
250 air granular bioreactor, which were more than those in the activated sludge bioreactor.
251 Under high organic (COD) loadings, biomass grows rapidly. Positive correlations between
252 biomass and EPS and between influent COD and EPS and vice versa were found, as they
253 both increase EPS. PO formed aerobic granules with some great ability of treating heavily
254 polluted wastewaters. This was due to the retention of biomass on an EPS matrix that
255 helped granules avoid disintegration. These granules were difficult to collapse allowing PO
256 passing through them, so they tolerated any shocks due to the high organic loadings.
257 In addition, EPS production in aerobic systems is also affected by the salinity, so the
258 combined effect of the aeration type and salinity needs to be considered. As salinity
259 increases, the specific oxygen uptake rate (SOUR) also increases, with microorganisms
260 needing more energy to endure the high saline conditions. In saline wastewaters, the large
261 mass transfer resistance makes it difficult for oxygen to diffuse from the gas interface to the
262 cell membrane as well as salinity itself also decreases oxygen solubility. CAS systems

263 cannot cope with the increased DO concentrations required, in particular when high
264 loadings are to be treated. PO use instead of air is then a promising alternative, (Hu et al.,
265 2019). Hu et al. (2019) found that, in sequencing batch reactors (SBRs), PO improved the
266 TOC removal efficiency at salinities less than 3%. Increasing salinity to values over 3.5%,
267 the TOC removal efficiency decreased regardless of the type of aeration, however, PO still
268 performed better than air. As salinity kept increasing, even high DO concentrations had a
269 negligible effect on metabolism, because of the strong shock on the bacteria. EPS under
270 either type of aeration increased as salinity increased, with PO mostly producing more EPS
271 than air at the same salinity. In detail, at low salinities up to 1%, for both aeration types,
272 EPS amounts were similar. For salinities above 2%, EPS at PO aeration conditions
273 increased. PO mitigated the effect of high viscosity on oxygen transfer and promoted EPS
274 production as a measure to protect the microorganisms from salt suppression. At a salinity
275 of 3%, polysaccharides increased over time in both aeration types, but not the proteins.
276 Initial concentrations of polysaccharides in both systems were lower than those of proteins,
277 but as salinity increased, their concentration exceeded the concentration of the proteins,
278 which remained stable. The production of polysaccharides was then the bacterial reaction
279 against the high osmotic pressure due to salt. As such, their concentration was higher when
280 PO was used due to the higher partial pressure, (Hu et al., 2019).
281 In addition, Hu et al. (2019) found that the soluble microbial products (SMP) also increased
282 under both aeration types, as salinity increased. At the highest salinity of 5%, SMP were
283 maximum in both bioreactors — 63.28 mg L^{-1} (PO) and 62.5 mg L^{-1} (air). At low salinities
284 of 0.5% or 1%, SMP in the bioreactor fed with PO were more due to the sufficient
285 degradation rate. At salinities of 2%, 3% or 4%, the opposite happened. The increased EPS
286 production to help bacteria tolerate the saline conditions reduced the production rate of

287 biomass associated SMP in the bioreactor fed with PO. At a salinity of 5%, the effect of
288 salinity was dominant, as high osmotic pressures started rupturing the cells leading to
289 higher biomass associated SMP production rates due to the EPS hydrolysis at a rate that
290 was proportional to the EPS production.
291 Finally, by running batch experiments, Zhang et al. (2019) showed that at a low F/M ratio
292 of 100 mg_{TOC} L^{-1}, SMP concentration remained stable regardless of the type of aeration. On
293 the other hand, at a higher F/M ratio of 500 mg_{TOC} L^{-1}, SMP concentrations were always
294 higher when PO was used. Improved oxygen transfer improved the enzyme and biomass
295 activity, so the production of utilization associated SMP at high F/M ratios increased.
296
297 **3.2. Enzyme Activity and Microbial Diversity**
298
299 The enzyme activity shows the ability of bacteria to adapt themselves to environmental
300 changes. During the formation of activated sludge, microorganisms use their enzymes, e.g.
301 catalase, dehydrogenase, phosphatase, protease, esterase, glucosidase, to hydrolyze and
302 biodegrade organic matter, mostly consisting of proteins and carbohydrates. Based on their
303 variations, the physiology of the bacterial community is assessed, (Calderón et al., 2012).
304 Enhanced enzyme activity leads to better multiplication conditions for the living
305 microorganisms and subsequently improves the pollutant removal efficiency. However, at
306 low DO conditions and high organic loading rates (OLRs), the enzyme activity deteriorates.
307 PO use may then accelerate it, so it will consequently improve the microbial biomass
308 activity as well — the increased substrate utilization rates when PO is used are interrelated
309 with higher enzyme activity. PO aeration demonstrated higher concentrations for many of
310 the enzymes. Microorganisms can adapt to PO aerated environments, so that the secretion

311 of enzymes stabilizes at high values, (Pan et al., 2017; Zhuang et al., 2016a; Doviral-García
312 et al., 2014). In addition, Zhuang et al. (2016b), by measuring SOURs, showed that when
313 PO was used, SOUR was 4.15 mg_{O2} g_{MLSS}^{-1} h^{-1}, or 28% higher than the SOUR that was
314 measured when air was used.
315 However, the improved biomass activity in POAS may also have an adverse effect, which
316 needs considering, as it results in rapid DO depletion in the secondary clarifiers, which
317 usually lack any aeration facility — a condition which can be additionally favored under
318 higher ambient temperatures, as these temperatures further improve bacterial metabolism.
319 In more detail, to avoid septic sludge and poor separation of solids from liquid as well as
320 foul odours, DO levels must be always maintained at the required level. As the mixed
321 liquor approaches the last stage in the aeration tank, DO concentrations drop and CO_2
322 concentrations increase. Increased CO_2 levels decrease the pH promoting the growth of
323 filaments (that also thrive at low substrate concentrations) and fungi, which consequently
324 hinder sludge settling and compaction of sludge. Sludge bulking will finally increase the
325 concentrations of total suspended solids (TSS) in the effluent and cause losses of active
326 biomass. In case oxygen becomes the limiting substrate, glucose consumption rates also
327 increase causing sludge deflocculation. Therefore, sludge exhibits poor settling properties
328 in the secondary clarifiers resulting in effluents of deteriorated quality. Any use of the
329 design of and of the operational guidelines for CAS systems to POAS systems is not
330 advisable as their microbial populations and the metabolic rates may not be similar. The
331 challenge above must then be considered when POAS systems are to be designed, (Kundral
332 et al., 2013; Lee et al., 2003).
333 Gostick et al. (1992) had also observed that at low F/M ratios, the POAS plant under their
334 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

359 removal, (Zhuang et al., 2016b). However, when Calderón et al. (2012, 2013) compared
360 MBRs employing PO with MBRs employing air, they concluded that the aeration type had
361 only a negligible effect on the diversity and functionality of the bacterial community, which
362 was particularly true when this effect had to be compared with the effects of temperature or
363 VSS concentration. However, they also supported the fact that that type of aeration affected
364 the bacterial community structure and differences in the relative abundance of dominant
365 populations were once again recorded, (Calderón et al., 2012, 2013).
366
367 **3.3. Foam and Froth Formation**
368
369 Excessive foam formation can affect the final water quality, as bacteria trapped in foam die,
370 so the aerobic treatment performance and the system's operational stability deteriorate. In
371 the case of air, the large amounts of air that have to be used for sufficient DO
372 concentrations cause foam formation, which hinders digestion and promotes biomass
373 washout. To avoid or control this, as low flowrates as possible have to be applied, but these
374 flowrates may deprive bacteria of the required DO. The use of PO may once again be help,
375 (Zhuang et al., 2016a,b; Zupančič and M. Roš, 2008; Lee and Kim, 2003). Zhuang et al.
376 (2016a,b), concluded that the improved removal efficiency in MBRs fed with PO was
377 attributed to the prevention of foam expansion. Zhuang et al. (2016a) achieved DO
378 concentrations of 10 mg L^{-1}, with a foam to liquid (F/L) ratio of 6%-10% for their MBR
379 that was fed with PO and of 30%-45% for the MBR that was fed with air. Since air is only
380 21% O_2 in volume, the air flowrate needed to be up to five times higher leading to
381 excessive foam formation, (Zhuang et al., 2016a,b).

382 CAS applications also suffer from froth caused by Nocardioform organisms. These aerobic
383 gram positive hydrophobic filaments preferentially concentrate at the air liquid surface and
384 produce thick viscous froth in both the aeration basins and the secondary clarifiers. Froth
385 causes a series of problems, related to either the liquid itself or the solids handling
386 including deterioration of effluent quality. Both CAS installations, particularly those
387 operated at long SRTs, and POAS installations are equally affected by froth. POAS plants
388 are affected by froth due to them containing significant surface trapping of activated sludge.
389 One way of avoiding froth in POAS proposed Jolis et al. (2006) was the application of a
390 low SRT and selective wasting. They found that an SRT of 0.3 days resulted in complete
391 removal of filamentous microorganisms in two days, which subsequently allowed the
392 POAS plant to operate successfully at an SRT up to 3 days without confronting any return
393 of the filaments. In a further analysis, Jolis et al. (2007) additionally highlighted the
394 importance of using anaerobic selectors in POAS to promote growth of phosphorous
395 accumulating organisms (PAOs), which outcompete filaments. Under operating conditions
396 favoring enhanced biological phosphorous removal (EBPR), that was at an SRT<2 d and an
397 HRT>55 min, a decrease in filamentous organisms occurred resulting in effective froth
398 control as well, (Jolis et al., 2007).
399 To this point, it may also be worth mentioning that the use of PO, on the other hand, may
400 also be able to stimulate the growth of PAOs in EBPR systems, which alternate anaerobic
401 and aerobic environments. Wei et al. (2014), who operated laboratory scale pressurized
402 oxygen aeration SBRs, found that PO increased the oxygen-reduction potential (ORP) from
403 the highly negative value required during the anaerobic treatment to the positive value
404 required for the production of the oxidative environment for phosphorous uptake but no
405 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.

453 **4. EFFECT OF PURE OXYGEN ON EFFLUENT QUALITY**

454

455 **4.1. Organic Carbon Removal**

456

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

460

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

462

463 However, a proper direct comparison of the relative performance of the CAS and POAS
464 has long since been not an easy task as it needs involvement of many factors and requires
465 complex experimental designs, (Cohen, 1977; Dirk, 1981). Any chance for advanced
466 performance of POAS had to be solely attributed to the higher partial pressures and its
467 ability to transfer oxygen rapidly and not on any significant differences in the intrinsic
468 kinetic parameters and settling characteristics, (Shelef and Green, 1980). Rempel et al.
469 (1992), by operating a pilot CAS and a pilot POAS, treating mill effluent at different sets of
470 F/M ratios, SRTs and HRTs, showed that under similar sets of operating conditions,
471 biochemical oxygen demand (BOD) and COD removal efficiencies between the two
472 systems were not very different. They also pointed that the effect of operating times on the
473 removal efficiencies was more important than that of the type of aeration.
474 Esparza-Soto et al. (2006a) additionally claimed that although PO increases treatment
475 capacity of the aeration stage and produces biosolids that settle better, its benefits against
476 air are still under discussion. PO systems, at high F/M ratios, are very successful in

477 removing five-day BOD (BOD_5) and suspended solids (SS) as well as in producing little
478 sludge. In addition, COD removal efficiencies in these systems also remain high, about
479 80% for F/M ratios up to 2.8 mg_{BOD5}/mg_{MLVSS}. However, this COD removal efficiency was
480 also the same when air was used as well. The difference was that in the case of air, the
481 range of F/M ratios applied was shorter and equal to only one third of that one reached by
482 PO. For F/M ratios up to about 1.25 mg_{BOD5}/mg_{MLVSS} or 1.5 mg_{COD}/mg_{MLVSS}, both aeration
483 types remove similar COD amounts. Nonetheless, at higher F/M ratios, a fair comparison
484 cannot be made, as the systems fed with air have never been run at such high F/M ratios
485 due to their restricted oxygen transfer capacity.
486 In addition, Esparza-Soto et al. (2006b) determined the molecular weight (MW)
487 distribution in wastewater samples from full-scale WWT plants using different aerobic
488 treatments and concluded that the plants generated effluent organic matter (E_fOM) with
489 different MW dissolved organic carbon (DOC) distributions. Systems operating with air
490 generated E_fOM with centrally clustered distribution — the intermediate MW fraction
491 contained most of the organic matter, i.e. 50%-60%, DOC: 0.5 kDa to 3 kDa. Systems
492 operating with PO, on the other hand, generated E_fOMs with skewed distributions towards
493 the high MW fraction, i.e. 40%-50%. Long SRTs reduced the DOC concentration in the
494 effluent, but the MW distribution remained unmodified. PO generated organic matter of a
495 more refractory character. That was due to the higher DO concentrations, which increased
496 the endogenous respiration rate and promoted the production of EPS and SMP, which are
497 high MW refractory biopolymers. Both aeration types were then equally efficient up to
498 certain F/M ratios, although PO systems extended operation to higher F/M loadings.
499 However, if we aim for the production of E_fOM of better quality — less DOC with lower

500 MW — the systems operating with air were more efficient within the range of F/M ratios
501 where the use of both air and PO was applicable, (Esparza-Soto et al., 2006a,b).
502 On the other hand, there are also studies which clearly state that the use of PO improves
503 effluent quality. For example, Zhang et al., 2019, found different TOC biodegradation
504 levels for PO and air, with PO showing a better performance. In addition, an increase in the
505 MLSS concentration increased the gap between the PO and air related removal efficiencies,
506 from 16.8% of TOC removal at 2000 mg L^{-1} to 76.5% at 8000 mg L^{-1}. Pan et al. (2017)
507 also observed more filamentous, actinophryids and nematodes when air was used as well as
508 the percentage of the total aerobic bacteria was higher when PO was used. However, in
509 both cases, they carried out batch tests at small scale and, as PO acts faster than air, its use
510 may be more advantageous with respect to operation restricted to short times. Finally,
511 Bernat et al. (2017) found that despite the abundance of Vorticella infusionum, whose
512 presence indicates unfavorable treatment conditions, in the mixed liquor, a stable and
513 acceptable effluent quality was achieved in their POAS system, but this is more related to
514 the fact that PO manages to better maintain aerobic conditions. As a conclusion, either
515 under air or PO conditions, there seems to be no significant difference in terms of organic
516 carbon removal. However, PO may be of help in cases where operational parameters do not
517 support the use of air.
518
519 **4.2. Removal of Phenolic Compounds and Micropollutants**
520
521 Regarding phenolic compounds, PO use is quite promising. Moerman et al. (1995), by
522 operating a POAS plant treating pretreated diluted carbonization wastewater, proved that
523 98% of thiocyanate, a substance whose overloading inhibits any phenol degradation, had

524 been removed. In addition, Li and Loh (2006) found that during the cometabolism of 4-
525 chlorophenol in the presence of phenol in an immobilized cell hollow fibre MBR enriched
526 with Pseudomonas putida, both 4-chlorophenol and phenol degradations improved when
527 PO was used. Finally, PO was satisfactorily used in continuous flow fluidized bed reactors
528 for the degradation of polychlorinated phenols that are included in wood preservation
529 chemicals, (Puhakka and Järvinen, 1992).
530 In addition, PO use can also help with the removal of refractory pollutants and
531 micropollutants. CAS cannot remove antibiotics, endocrine disrupting compounds,
532 pharmaceuticals and residual personal care products, household and industrial chemicals,
533 etc. as CAS plants have not been designed or operated for this purpose, (Batt et al., 2007).
534 Levine et al. (2006) assessed the persistence of a number of micropollutants during primary
535 treatment, biological treatment comprising POAS and nitrification/denitrification and
536 finally disinfection. Several substances tested had lower concentrations in the denitrified
537 effluent than in the influent or the primary treatment effluent, which means that biological
538 treatment with PO additionally helped with their removal. For instance, acetaminophen, a
539 non-antibiotic over-the-counter pharmaceutical that had been detected at the highest
540 concentration of 10 μg L^{-1} in the influent, was eliminated during biological treatment.
541 Batt et al. (2007) monitored the fate of four antibiotics, i.e. ciprofloxacin (CIP),
542 sulfamethoxazole (SMX), tetracycline (TC) and trimethoprim (TRI), in four full scale
543 WWT plants including POAS. The use of PO had a positive effect on the removal of the
544 antibiotics, mainly with respect to removal of SMX and TRI, despite the short HRT of 1 h
545 that was applied. In all plants tested, removal efficiencies were strongly related to the
546 operating times. However, by comparing the performance of the POAS plant with that one
547 of the rotating biological contactor (RBC) plant, it was found that, although similar

548 removal efficiencies for all antibiotics were reported for both plants, the HRT at POAS was
549 four time less than that at RBC.
550 Bae et al. (2015) also assessed the contribution of PO aeration of a combined biological and
551 physicochemical treatment (POAS + Fenton Process) system to the removal of refractory
552 pollutants from dyeing wastewater, which is not a readily biodegradable wastewater as well
553 as any potential improvement with respect to biological treatment of dyes needs long SRTs
554 and high MLSS concentrations. Although the biological treatment system suffered from
555 low MLSS concentrations due to inefficient settling, it managed to remove 53% of soluble
556 COD (SCOD) and 12% of color indicating that the microorganisms in the aeration tank
557 were acclimated to the dyeing wastewater. As such, Bae et al. (2015) managed to obtain a
558 cost effective pretreatment, which both helped the Fenton Process in becoming more
559 efficient and decreased the consumption of chemicals.
560 Martín-Rilo et al. (2018) employed PO aeration for the removal of a benzotriazole based
561 anticorrosive from dairy wastewater with benzotriazole being an aromatic compound used
562 as a metal corrosion inhibitor and an emerging toxic that tends to bioaccumulate. PO was
563 injected in the intermediate step (Step 2) of a treatment process — that step was preceded
564 by wastewater neutralization under CO_2 injection (Step 1) and followed by
565 coagulation/flocculation in a dissolved air floatation tank (Step 3). Steps 1, 2, and 3
566 removed 44%, 30%, and 25% of the total benzotriazole respectively for an overall removal
567 of 99.7%. The respective removal efficiencies of each step considering the concentration of
568 the contaminant entering each step was 44%, 53%, and 95.6% respectively, so more than
569 half of the amount of the chemical that entered Step 2 was removed.
570 Finally, PO aeration may also be beneficial regarding the removal of endocrine disruptive
571 compounds from wastewater. For example, bisphenol-A, the endocrine disrupting chemical

572 of the greatest concern, due to its effects being more detrimental than those of other
573 substances of the same kind, cannot be removed by CAS. In addition, its removal does not
574 necessarily guarantee its complete degradation, as, due to its hydrophobic nature, it is also
575 be stored in the sludge or, where possible, it can be adsorbed by membranes, (Doviral-
576 García et al., 2014). Dorival-García et al. (2014) assessed then the fate of bisphenol-A by
577 employing MBR technology to prolong SRT, and they found that bisphenol-A reached the
578 background level in the effluent in five days when PO was used, whereas it took 10 days in
579 the case of air. By switching from air to PO, they also improved the biodegradation
580 percentages from 60.2% to 87.4% in 10 days. With air, bisphenol-A that was sorbed in the
581 sludge, remained within it, so its availability for biodegradation was reduced and the need
582 for further sludge treatment before sludge disposal in order to remove the sorbed
583 contaminant increased. On the other hand, with PO, the sorption tendency diminished and
584 desorption was facilitated — 67.4% of bisphenol-A in the sludge underwent desorption and
585 consequently biodegradation.
586
587 **4.3. Removal of Volatile Organic Compounds**
588
589 PO aeration is also highly capable of handling foul condensates without stripping or of
590 biodegrading VOCs. To this end, Freitas dos Santos and Livingston, (1993a,b) proposed a
591 gas enclosed recirculation system based on a bioreactor fed with PO, whose design was
592 similar to that of a an air lift bioreactor, for the aerobic degradation of the 1,2-
593 dichloroethane in 1,2-dichoroethane contaminated wastewater. Air stripping of 1.2-
594 dichloroethane that usually takes places during its aerobic treatment was avoided — VOC
595 emissions are known to cause immediate toxicity and odor and promote undesired chemical

596 reactions. In their conventional air lift bioreactor, 33% of the 1,2-dichloroethane was lost
597 due to stripping, whereas in the system where PO was used, the whole of 1.2-
598 dichloroethane was mineralized.
599 In addition, Paice et al. (2003) mentioned that one of the selling points of POAS systems
600 for kraft pulp mills was their ability to treat their foul condensates without any stripping,
601 even from the start-up. However, these systems were liable to some problems, which were
602 mainly directly related to their enclosed design. As such, the potential for high
603 concentrations of dissolved CO_2 in the effluent, the potential for combustible gas alarms
604 caused by VOCs in the condensates and possible premature corrosion of the concrete,
605 which was associated with the bacteria that reduce sulfuric compounds, had to be carefully
606 considered. These problems can also be additionally intensified, as this design further
607 complicates the inspection and maintenance of the systems, (Paice et al., 2003).
608
609 **4.4. Effect of Pure Oxygen on Heavy Metals**
610
611 Heavy metals are non-biodegradable toxic substances that interact with the biomass and
612 have various inhibitory or toxic effects on bacteria, with nitrifying autotrophs being more
613 sensitive to them than heterotrophs, (Avezzù et al., 1995). Avezzù et al. (1995) assessed the
614 fate of heavy metals during treatment of leachates by monitoring their distribution in the
615 solid or liquid phase of the biological process and showed that, despite their accumulation
616 in the biomass, BOD_5 and COD removal efficiencies remained quite high. As such, PO
617 aeration created a quite favorable living environment for the microorganisms, which, by
618 making microorganisms more resistant to compounds like heavy metals, managed to
619 maintain considerable removal of organic carbon, (Avezzù et al., 1992).

620 **5. PURE OXYGEN AND NITRIFICATION**

621

622 **5.1. Introduction**

623

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

639

640 **5.2. The effect of pH**

641

642 POAS is more vulnerable to low pH inhibition of nitrification than CAS because of their
643 closed headspace design, whose main role is to minimize oxygen losses by recycling

644 headspace gas. Because of the aerobic treatment, the headspaces can contain elevated
645 amounts of CO_2, which under slightly increased pressure moves into the mixed liquor
646 reducing the pH, whose degree of reduction depends on parameters like the system's buffer
647 capacity or the degree of venting. This decrease affects the kinetics of enzyme reactions,
648 the bacterial species predominance and the physical properties of the organisms and
649 particles. When nitrification has to take place, pH reduction additionally inhibits it, unless
650 acclimation has already taken place. As nitrification proceeds, alkalinity is consumed,
651 which further reduces the pH — entrapped CO_2 does not reduce alkalinity itself but it does
652 reduce the pH at a given alkalinity. Many POAS systems were originally designed only for
653 removal of organic carbon, therefore to operate at low SRTs and HRTs ranging from 1.5 h
654 to 2.5 h. In practice, nitrification could proceed at lower pH values in covered POAS
655 systems, however, this requires contact times up to 3.5 h-5 h, which are rather longer than
656 those usually applied to handle carbon. To resolve this, another process step may then have
657 to be added, (Garber, 1977; Dirk, 1981; Shelef and Green, 1982; Sear et al., 2003). In
658 addition, to reinstate nitrification that had been hindered by a pH of 6.5, Mauret et al.
659 (2001) either alternated aeration with air and PO or combined the two. Nitrification that was
660 non-existent at pH values <6 could also have been held if acclimation had preceded, e.g.
661 Sears et al. (1995) found that nitrification in POAS was stable at pH values between 5 and
662 5.5 provided that the required acclimation period had been applied.
663 As combined carbonaceous and nitrogenous removal in POAS systems that are designed to
664 remove only organic carbon is difficult, particularly in enclosed ones, Bonomo et al. (2000)
665 proposed as an alternative the use of MBBRs aerated with PO for tertiary nitrification of
666 the secondary effluent. The extra treatment step eliminated any competition between
667 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

692 100% when the reactor's headspace was flushed with PO and 42% when it was flushed
693 with air.
694 Salvetti et al. (2006) also operated MBBRs fed with PO to monitor the combined effect of
695 the temperature and the type of aeration on nitrification. PO diffuses more deeply into the
696 biofilm than air, so it produces higher nitrification rates, hence, requiring smaller reactor
697 volumes, (Salvetti et al. 2006). At low ammonium concentrations, Salvetti et al. (2006)
698 found PO was not essential as air could provide the required DO and the temperature did
699 not have any significant effect on the nitrification rates. As ammonium concentration
700 increased, DO became the reaction limiting substrate **(**this occurs even at DO
701 concentrations as high as 5 mg L^{-1} or 10 mg L^{-1}), so PO use was preferred. These findings
702 are also in line with the findings of Bonomo et al., (2000) mentioned in Section 5.2. Under
703 oxygen limiting conditions, the specific biomass activity, as the ratio of nitrification rate
704 to biomass content on the support media, was higher between 23°C-28°C than between
705 18°C-22°C. By removing the effect of DO on biomass, whose solubility also decreases as
706 temperature increases, that was further attributed to the higher temperature that favors
707 nitrification and to the reduced resistance to diffusion, which allows more biomass to have
708 access to DO.
709
710 **5.4. The Effect of Operational Times**
711
712 The role of SRT is important for POAS, as longer SRTs favor nitrification, (Sears et al.,
713 2003). As nitrifiers grow slowly, their growth is strongly related to the applied SRT. Due to
714 the high treatment rates that are achieved thanks to their high OTRs, POAS plants are
715 usually operated at short sludge ages ranging from 1 day to 3 days, which are not long

716 enough for nitrification, (Neethling et al., 1998). To overcome this, Neethling et al. (1998)
717 transferred aged waste activated sludge from a CAS plant to a POAS plant to seed it with
718 nitrifiers. This increased the "apparent" sludge age and the high loaded POAS plant
719 achieved nitrification. After the seeding process, they found that ammonium concentration
720 in the effluent decreased to 5 mg L^{-1} within 3 days stabilizing to values below 0.5 mg L^{-1}
721 after about a month. Similarly, Randall and Cokgor (2001) applied lower flows and
722 continuous seeding to maintain nitrification to a full scale POAS system.
723 In addition, Rodriguez et al. (2012c) studied the effect of HRT on nitrification under either
724 PO or air conditions by employing MBR technology for the treatment of primary clarifier
725 effluent. They showed that, by using PO instead of air, nitrogen removal efficiency was
726 improved by 8% at an HRT of 12 h and by 13.5% at 18 h. Kinetics**,** as per the Monod's
727 model and with ammonium being the substrate, also supported that result. At similar
728 temperatures and MLSS concentrations, the K value, or the half-saturation constant for the
729 PO increased by 190.4% at 18 h and by 324.1% at 12 h. However, this research did not
730 analyze the complex relationship between the K value, the MLSS concentrations and the
731 applied HRT. When air was used, the k_H value, or the hydrolysis constant, decreased and
732 the k_d value, or the decay constant, increased, which additionally stated the advantage of
733 the PO use, which was further strengthened at longer HRTs. In addition, by employing
734 oxidation of anaerobically treated excess sludge with PO, Zupančič and Roš (2008) showed
735 that at an HRT of 5 days only 38% of ammonium was converted. This increased to <70% at
736 HRTs of 6 days or 7 days, to 85.1% at 8 days and finally 85.6% at 10 days indicating that
737 no further improvement was possible. As such, the nitrification bacteria needed longer
738 contact times to process large loadings of ammonium.
739

740 **6. PURE OXYGEN AERATION AND MEMBRANE FOULING IN PURE OXYGEN**
741 **MEMBRANE BIOREACTORS**
742
743 As membrane fouling continues being an important research field in membrane bioreactors,
744 in this section, we mainly concentrate on the potential effect of PO aeration on its
745 mitigation during WWT. Despite the fact that the performance of MBRs operating at
746 low/medium COD loading rates, e.g. <1 kg m^{-3} d^{-1}, has been widely studied, operation at
747 COD loading rates >2 kg m^{-3} d^{-1} is not common. This is because of the incapability of
748 maintaining a healthy aerobic environment, due to limited oxygen transfer efficiency, (Lee
749 and Kim, 3003). As such, PO aeration suits well MBRs where high MLSS concentrations
750 have to be maintained and high organic loadings have to be treated, so OURs as high as 50
751 mg_{O2} L^{-1} h^{-1} - 150 mg_{O2} L^{-1} h^{-1} are needed, (Larrea et al., 2014).
752 Although MBRs have many advantages over CAS, membrane fouling is still a problem that
753 increases running cost and potentially reduces treated water volumes and quality, (Zhuang
754 et al, 2016b). Membrane fouling in MBRs is the systematic accumulation of suspended
755 solids, colloids, and macromolecules on the membrane surface, or inside the pores, causing
756 a reduction in membrane permeability. It is a complex phenomenon that is affected by the
757 membrane itself, the wastewater under treatment, etc. with viscosity of wastewater,
758 EPS/SMP production, size distribution of sludge flocs and the presence of colloids
759 contributing to this. An accurate indicator of the degree of membrane fouling is the
760 transmembrane pressure (TMP), (Zhuang et al., 20016b; Lee and Kim, 2003).
761 With respect to the aeration type, Zhuang et al. (2016b) showed that the degree of
762 membrane fouling was 33% less (on day 90) when an MBR was fed with PO, instead of air.
763 Even though it had been expected that the higher air flowrates and the larger bubble size

764 would have improved scouring and led to less fouling, it was finally proved that the PO fine
765 bubbles controlled EPS production and the biomass particle size distribution better
766 mitigating membrane fouling. Despite the fact that EPS amounts increased under both
767 aeration types, their amount was lower when PO was used. High shear forces and high
768 aeration intensities when air was used released more EPS deteriorating membrane fouling.
769 Shear forces affect floc size, strength and compactness affecting sludge filterability. When
770 PO was used, the flocs were constantly exposed to low shear forces, so they grew into loose
771 weak ones. On the other hand, when air was used, the high shear forces broke flocs to
772 fragments promoting membrane fouling, (Zhuang et al., 2016b).
773 Lee and Kim (2003) also operated an MBR fed with PO at 2 $kg_{COD}\ m^{-3}\ d^{-1}$ and the critical
774 TMP (50 kPa) was reached in 50 days. By increasing the OLR to 4 $kg_{COD}\ m^{-3}\ d^{-1}$, it took 20
775 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
776 membrane fouling rates demonstrated a trend twice as high as the organic loadings,
777 however, after applying 8 $kg_{COD}\ m^{-3}\ d^{-1}$, the slope of membrane fouling rate decreased. As
778 the organic loading increases, biomass also increases, so the F/M ratio is affected. The F/M
779 ratio has additionally an effect on membrane fouling, but potential differences in MLSS
780 concentrations and membrane fluxes at different F/M ratios have a complex effect, which is
781 difficult to determine. For MLSS concentrations up to 10000 mg L^{-1}, Kim et al. (2003)
782 found that the increase in TMP was smooth. However, it dramatically changed when the
783 MLSS concentration exceeded 10000 mg L^{-1}, as the fouling rate was almost four times
784 higher — it increased from 1.3 to 5.6. This became ten times higher, when the MLSS
785 concentration exceeded 15000 mg L^{-1}, as it increased from 1.3 to 12.5.
786 MLSS concentrations directly affect viscosity. High viscosity suspensions require high
787 cross flow velocities to create turbulence. If turbulence provided by aeration is not enough

788 to scour solids off the membrane, membrane becomes fouled faster. An optimal MLSS
789 concentration in relation with F/M, viscosity and structure/size of flocs seems to exist. In
790 reality, membrane fouling is more affected by an increase in MLSS concentration than an
791 increase in the organic loading. However, it is difficult to evaluate membrane fouling by
792 disconnecting the two, as each time the organic loading increases, the biomass
793 concentration increases as well. Therefore, it depends on whether membrane fouling is
794 assessed from an organic loading or a biomass point of view. However, membrane fouling
795 does not start building up soon after an increase in the organic loading but it takes some
796 time due to biomass also needing time to increase and produce EPS at a sufficient amount.
797 However, it must be pointed that the opposite can also be found in the literature, namely
798 MLSS concentrations have no effect or even have a positive one on TMP. This can indeed
799 happen, as the effect of MLSS concentrations on the filtration resistances is case-specific,
800 (Lee and Kim, 2003).
801 Finally, Rodriguez et al. (2012a) quantified the influence of PO or air on the recovery of
802 permeability of the membrane, namely the fraction of the difference of permeability, which
803 is the quotient of flux over TMP, after cleaning minus the permeability before cleaning
804 over permeability after cleaning. As such, a physical cleaning based on backflush of
805 permeate and a chemical one, which becomes necessary when irreversible fouling needs to
806 be removed, were tested. The mean recovery was found to be higher when PO was used
807 regardless of the type of cleaning — 2% and 15% further improvement for the physical and
808 the chemical cleaning respectively, which is also in line with the fact that physical
809 cleanings are weaker than the chemical ones.
810
811

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.

836 Regarding removal of contaminants and treated water quality, PO aeration was proven
837 quite efficient. However, after comparing performances of CAS and POAS systems with
838 respect to carbonaceous matter removal, it was found that the type of aeration was not so
839 critical as the HRT or the SRT. In addition, at low/medium F/M ratios, where both
840 activated sludge systems can be equally applicable, no significant differences in terms of
841 organic carbon removal are reported. The advantage of the use of PO though is that
842 operation of POAS plants can be extended to higher F/M ratios where CAS plants are not
843 usually designed to operate. Even though there is some research, which claims that, under
844 similar operating conditions, PO aeration may lead to improved removal of organic carbon,
845 this research is currently restricted to batch applications, e.g. SBRs, at small scale, where
846 any improved removal efficiency can be attributed to the fact that PO acts faster than air.
847 However, PO use may be more promising with respect to the removal of refractory
848 pollutants and micropollutants. Under PO aeration, improved biological performance has
849 been monitored with respect to removal of phenolic micropollutants, antibiotics, endocrine
850 disrupting compounds, etc. Finally, organic matter removal in POAS plants remains
851 unaffected by the presence of toxic heavy metals in the wastewater.
852 Regarding nitrification, it mainly depends on parameters other than the type of aeration,
853 however, under conditions that undisputedly do not inhibit nitrification, higher DO
854 concentrations due to PO, improved the removal of ammonium from wastewaters.
855 Nonetheless, POAS systems are designed to operate at short SRTs and remove only organic
856 carbon, as the short SRTs hinder nitrification. This can be overcome by continually seeding
857 the aeration tanks with aged sludge. In addition, under conditions where PO alternation
858 with air, or vice versa, is possible, simultaneous removal of carbon and ammonia in one
859 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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1112 **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

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1119 **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}

1120 **Abbreviations:** DWF: Dry Weather Flow, N/A: Non-Applicable, PE: Population Equivalent, UK: United Kingdom,
1121 USA: United States of America, WWF: Wet Weather Flow. [a]Alternating Anoxic-Aerobic Process, [b] Biological Nutrient
1122 Removal System
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1129 **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)

1130 **Abbreviations:** BOD_5: Five-day Biological Oxygen Demand, COD: Chemical Oxygen Demand, HRT: Hydraulic
1131 Residence Time, MBBR: Moving Bed Biofilm Reactor, MBR: Membrane Bioreactor, NF: Nanofiltration, NH_4^+-N:
1132 Ammonium Nitrogen, SCOD: Soluble COD, TOC: Total Organic Carbon, TN: Total Nitrogen, TPh: Total Phenols, VSS:
1133 Volatile Suspended Solids. [a]Batch Reaction