

1                   **OPTIMIZATION METHODOLOGY FOR HIGH COD**  
2                   **NUTRIENT-LIMITED WASTEWATERS TREATMENT USING**  
3                   **BAS PROCESS**

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15                   **Keywords:** biofilm activated sludge, methodology, optimization, operational costs,  
16                   effluent quality, decision making.

17  
18                   **ABSTRACT**

19                   Optimization of biofilm activated sludge (BAS) process via mathematical modelling is  
20                   an entangle activity since economic, environmental objective and technical decision must  
21                   be considered. This paper presents a methodology to optimize the operational conditions  
22                   of BAS process in four steps by combining dynamic simulation techniques with non-  
23                   linear optimization methods and with operative decision making criteria. Two set of  
24                   variables are separately prioritized in the methodology: essential variables related to  
25                   physical operation to enforce established process performance, and refinement variables  
26                   related to biological processes that can generate risks of bulking, pin-point floc and rising  
27                   sludge. The proposed optimization strategy is applied for the treatment of high COD  
28                   wastewater under nutrient limitation using an integrated mathematical model for COD  
29                   removal that include predation, hydrolysis and a simplified approach to the limiting solids

30 flux theory in the secondary clarifier in order to facilitate the convergence of the  
31 optimization solver. The methodology is implemented in a full scale wastewater treatment  
32 plant for a cellulose and viscose fibre mill obtaining: i) improvement of the effluent  
33 quality index ( $\text{Kg pollution/m}^3$ ) up to 62% and, ii) decrease the operating cost index  
34 ( $\text{€m}^3$ ) of the process up to 30% respect the regular working operational conditions of the  
35 plant. The proposed procedure can be also applied to other biological treatments treating  
36 high COD nutrient-limited industrial wastewater such as from textile and winery  
37 production among others.

## 38 **1. Introduction**

39 Emission limits for industrial effluent are constantly being tightened up. Activated sludge  
40 (AS) process is a common system for biological treatment of industry effluents; however,  
41 more sustainable solutions require other technologies such as biofilm activated sludge  
42 process (BAS). BAS is composed of moving bed biofilm reactor (MBBR) and activated  
43 sludge reactor (AS) that are used as bacterial and predator stage respectively (Sointio et  
44 al. 2006, Revilla et al. 2016a). The overall result in BAS processes increases COD  
45 removal performance respect to other conventional treatment and, at the same time, lay  
46 out lower sludge production. An additional and determining benefit is the improvement  
47 sludge settleability in the final stage of secondary settling (Rankin et al. 2007) that allows  
48 that the activated sludge reactor to be operated at increased biomass concentration while  
49 simultaneously total suspended solids (TSS) concentration in the effluent can be reduced  
50 (van Haandel and van der Lubbe 2015). Moreover, this biologic double stage process can  
51 avoid the risk of bulking when it is operated under nutrients limitation (Rankin et al.  
52 2007). Predation is the powerful mechanism of the BAS process that allows achieving  
53 their main characteristic as the low sludge yield.

54 Modelling of biofilm stages and activated sludge provided a better understanding of the  
55 intrinsic connections between soluble and particulate compounds, biomass properties and  
56 process performance in terms of COD, BOD and TSS (Fan et al. 2017). Commercial  
57 simulation platforms, including GPS-X and BioWin can be used to describe the one  
58 dimensional (1-D) multi-species biofilm structure and biological behaviour of the  
59 treatment process (Li et al. 2016); however, none of these platforms include the  
60 description of the predator microorganism which plays a predominant role in a BAS  
61 process under certain conditions such as nutrient limited conditions (Revilla et al. 2016a).

62 The authors published recently the mathematical model of BAS process including  
63 hydrolysis and predation and uses wastewaters coming from viscose and cellulose  
64 production industrial process to validate the model. In Revilla et al. 2016a the whole BAS  
65 process was simulated and the evaluation of the role and contribution of predator  
66 microorganisms towards COD removal, nutrient requirements and sludge production is  
67 displayed.

68 More strict regulations are being imposed regularly in terms of COD, BOD and TSS  
69 removal that enforce wastewater treatment technologies to progress (Guerrero et al. 2011;  
70 Kamali and Khodaparast 2015). Optimization of an existing facility in terms of cost,  
71 operational improvements and removal efficiency is the most effective method of  
72 achieving the stricter compliance and the most effective method to overtake common  
73 trade-off between treatment results and operational costs. However, expectation of having  
74 to satisfy simultaneously a variety of objectives (environmental, economic and technical)  
75 increases the complexity of the problem and becomes a very difficult task that should be  
76 solved blending experience engineers and specific mathematical tools (Descoins et al.  
77 2012; Hakanen et al. 2013). In this context, practical experience of the operator in the  
78 prioritization of conflicting objectives or in the application of rules of thumb, needs to be

79 considered in the mathematical model since certain units at wastewater treatment plant  
80 (WWTP) may be exceptionally difficult or risky to operate.

81 Multiple objective optimization (MOO) has been widely applied in operation and design  
82 of municipal WWTP for different applications using interactive approaches between  
83 several optimization and decision making tools (Dai et al. 2016; Garrido-Baserba et al.  
84 2016; Hakanen et al. 2011; Rivas et al. 2008; Sweetapple et al. 2014). However, a  
85 systematic methodology of multiple objective optimization of a full-scale BAS process  
86 has not been addressed, to the best of our knowledge.

87 Generally, urban wastewater has high presence of nutrients that facilitates the biological  
88 process; however, some type of wastewater such as viscose and cellulose industry  
89 wastewater are poor in nutrients that must be added externally with the consequent  
90 increase in the operating costs of biological treatments (Rankin et al. 2007).

91 In this paper, an optimization methodology for BAS processes treating highly COD  
92 wastewater under limited nutrient is presented. Simultaneous optimization of effluent  
93 quality and operating cost under prioritized technical specifications is the main goal of  
94 this procedure. Applicability of the proposed methodology for biological treatment  
95 processes using biofilm is illustrated using two industrial-scale case studies from viscose  
96 and cellulose wastewaters.

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101 **2. Identification of the problem**

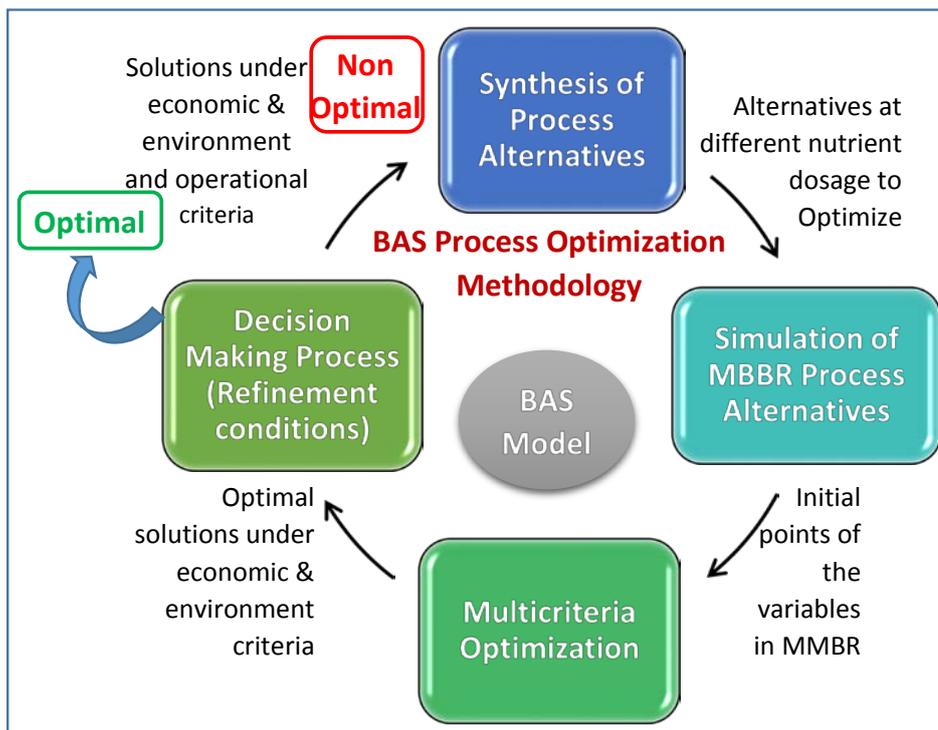
102 **2.1. Problem statement**

103 The problem of optimizing the operational conditions of the BAS plant can be stated as  
104 follows: given is a BAS plant with known design parameters and given is an influent  
105 stream with known flow rate that contain certain pollutants with known concentrations;  
106 the goal of this problem is to identify the operational conditions of the plant at minimal  
107 operational cost and minimal pollutant discharge in the effluent stream taking into  
108 account technical specifications.

109 To solve this problem a conceptual optimization methodology approach, based on four  
110 sequential stages showed in Figure 1, is proposed in this work. As explained previously,  
111 BAS process included two biological sequential steps: MBBR reactor and AS reactor. In  
112 MBBR reactor nutrient dosing is the unique variable that can be manipulated.  
113 Furthermore, the mathematical model of the MBBR reactor presented previously by the  
114 authors (Revilla et al. 2016b) is a multi-substrate biofilm and bulk liquid model and the  
115 mathematical model of the AS is continuous stirred-tank reactor. These two biological  
116 sequential steps described (MBBR and AS) are the base of the four sequential stages of  
117 the optimization methodology.

118 Starting from the regular operation conditions of an industrial plant, the first stage of the  
119 optimization methodology is the “Synthesis of Alternatives”; nutrient different dosage is  
120 the essential variable of the process (performance and cost) being used to generate  
121 different process alternatives. All the alternatives are simulated for MBBR reactor in the  
122 second stage “Simulation” using a previous MBBR mathematical method; besides, the  
123 results of “Simulation” allows obtaining the initial point of the variables of the AS  
124 process. The third step is the “Activated Sludge Multicriteria Optimization” that allows  
125 to obtain optimal solutions of the process variables under economic and environmental

126 evaluation criteria. This procedure (“Simulation of MBBR” and later “Activated Sludge  
 127 Multicriteria Optimization”) requires much less computation computational effort than  
 128 using an optimization software under dynamic and spatial conditions.  
 129 The last stage is the “Decision Making Process” where a set of refinement operation  
 130 bound let to obtain the optimal conditions under additional evaluation criterion. The  
 131 proposed methodology is iterative until the optimal solution is reached.



142 Figure 1.- Conceptual optimization methodology approach.

143  
 144 **2.2. BAS mathematical model**

145 A complete description for the mathematical models of four units involved in the BAS  
 146 process (MBBR, AS, secondary clarifiers and splitter) including the biological reactions,  
 147 stoichiometric and kinetic coefficients appears in previous papers presented by the  
 148 authors (Revilla et al. 2016a, b). In the present paper a significant modification has been  
 149 done in the mathematical model of secondary clarifier in order to evaluate the clarification  
 150 and thickening functions; a "simplified approach" proposed by von Sperling (2007) to the

151 limiting solids flux theory has been included in the model. Limiting solids flux concept  
 152 is widely used in the bibliography through the "non-differentiable minimum function"  
 153 (Amanatidou et al. 2015a, b) which requires iterative methods to solve it numerically and  
 154 can raise convergence issues when uses optimization algorithms (Hreiz et al. 2015a). The  
 155 use of the simplified approach facilitates the convergence of the optimization algorithms.

156 The simplified approach proposed by von Sperling (2007) takes into account four  
 157 variables i) hydraulic loading rate (HLR) which corresponds to the quotient between the  
 158 influent flow rate ( $Q_i$ ) and the surface area of the secondary settler (A) (equation 1), ii)  
 159 solids loading rate (SLR) which corresponds to the quotient between applied solids load  
 160 and the surface area of the secondary settler (equation 2), iii) sludge settling velocity ( $v$ )  
 161 (equation 3) and, iv) limiting solids flux (GL) (equation 4):

$$162 \quad \text{HLR (m}^3/\text{m}^2 \text{ hour)} = Q_i/A \quad (1)$$

$$163 \quad \text{SLR (Kg TSS/m}^2 \text{ hour)} = (Q_i + Q_R) \times \text{TSS}_{AS}/A \quad (2)$$

$$164 \quad v \text{ (m}^3/\text{m}^2 \text{ hour)} = (v_0) \times e^{-K \times \text{TSS}_{AS}} \quad (3)$$

$$165 \quad \text{GL (Kg TSS/m}^2 \text{ hour)} = m (Q_R/A)^n \quad (4)$$

166 where  $Q_R$  is the sludge recycle flow rate;  $\text{TSS}_{AS}$  are the total suspended solid  
 167 concentrations in AS reactor;  $v_0$ ,  $K$ ,  $m$  and  $n$  are specific correlation parameters.

168 In this work, "fair settleability", with sludge volume index (SVI) between 100-200 mL/g,  
 169  $v_0$  (8.6 m/hour),  $K$  (0.50  $\text{m}^3/\text{Kg}$ ),  $m$  (0.72) and  $n$  (8.41) is considered.

170 Finally, two new conditions are included in the previous BAS mathematical model  
 171 developed by the authors in order to ensure the clarification and thickening function.

- 172 • Clarification function: hydraulic loading rate (HLR) < sludge settling velocity ( $v$ )
- 173 • Thickening function: solid loading rates (SLR) < limiting solid flux (GL)

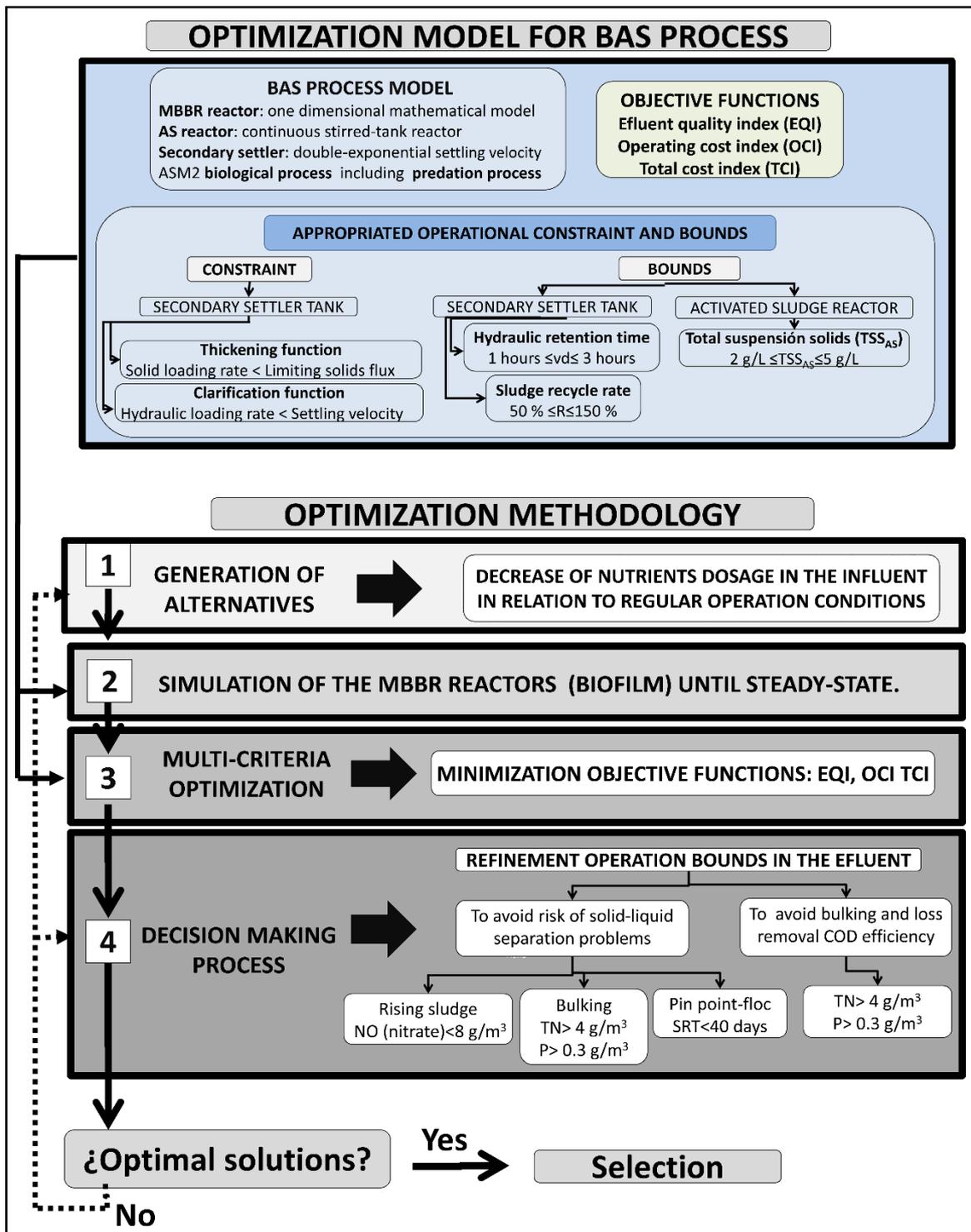
174 Blue section of the Figure 2 shows the essential attributes of the BAS process model  
175 including the appropriated operational variables and its constraints and bounds for the  
176 secondary settler tank and for the AS reactor apply in the present work. These suitable  
177 operational variables are included due to operational consistencies (Espírito Santo et al.  
178 2013) even though other authors can point out lightly different values (Henze 2008; Hreiz  
179 et al. 2015a; van Haandel and van der Lubbe 2015).

### 180 **3. Optimization methodology**

181 The complexity for simultaneous reduction of operational costs, reduction of the amount  
182 of pollutants discharged into sewer together with reduction of sludge generated, and the  
183 requirement of a technically well-operated full-scale wastewater plant have motivated  
184 this paper. Furthermore, this optimization process involves some other challenges such  
185 as, i) high number of (non-linear) equations and variables, and ii) dynamic and spatial  
186 distribution of the components into the length of the biofilm of the MBBR reactors.

187 The optimization methodology to fulfil the proposed objectives is divided into four  
188 consecutive steps detailed in the following sections. This approach simplifies the  
189 convergence of the mathematical models and the determination of the initial points of the  
190 variables. A detailed flowchart of the proposed optimization methodology and the relation  
191 with the mathematical model is shown in Figure 2.

192 This methodology can be used for wastewater under nutrient limitations coming from  
193 different industrial sector such as pulp and paper, petrochemical, pharmaceutical or food  
194 (Bakos et al. 2016; Freedman et al. 2005; Gray 2004; Hussain et al. 2015)



195

196 Figure 2. Flowchart of the optimization methodology.

197 **3.1. Generation of alternatives**

198 Nutrients must be dosed into the influent to ensure the proper growth of the  
 199 microorganisms because the BAS process under-study treats high COD wastewater under

200 limited nutrient condition. The optimization methodology starts generating alternatives  
201 by selecting one independent variable as additional constraint (Hreiz et al. 2015b). The  
202 selected variable in this work is the nutrient dosage in the influent; this variable affects  
203 the behaviour of MBBR reactors and the overall performance and cost of the BAS  
204 process. Different intervals of the nutrient dosage are considered to generate several  
205 alternatives depending on the precision degree required for the optimization process.

### 206 **3.2. Simulation of the MBBR reactors (biofilm) until steady-state.**

207 The mathematical model of the MBBR reactor previously developed by the authors  
208 (Revilla et al. 2016b) allows the determination of the concentration of the components in  
209 the reactor with time and the spatial distribution along the length of the biofilm. The AS  
210 model used by the authors for the AS step is simpler than MBBR model since it is  
211 modeled as a continuous stirred-tank (Revilla et al, 2016a).

212 The optimization methodology separates the BAS process into two stages: simulation of  
213 the MBBR reactor and optimization of the rest of BAS process. The simulation stage  
214 fulfils two aspects: i) solve a complex multi-species and multi-substrate biofilm and bulk  
215 liquid MBBR model and ii) facilitates the determination of the initial points of the  
216 variables for optimization process.

217 In the previous as well as in the present work, general chemical engineering process  
218 software (Aspen Custom Modeler) is used to simulate the behaviour of MBBR reactors  
219 since this software facilitates the creation of rigorous dynamic and spatial model. In this  
220 second stage of the optimization, this MBBR model is used to simulate the behaviour of  
221 the components for each alternative in the biofilm and in the reactor until steady-state.  
222 Once the steady-state is reached, the MBBR results of each alternative are sent to third  
223 stage of the methodology: optimization of the conditions of the BAS process.

### 224 3.3. Multicriteria optimization

225 Once MBBR behaviour is simulated until steady-state, the rest of the process is optimized  
226 for each alternative using three different index as objective functions: total cost index,  
227 effluent quality index or/and operating cost index as objective functions. The optimization  
228 of the BAS process is done by the software General Algebraic Modeling System (GAMS)  
229 using CONOPT as NLP algorithm (El Shorbagy et al. 2013).

#### 230 3.3.1. Objective functions

231 Three indexes are used as objective functions to be minimized in the present work:  
232 effluent quality index (EQI) that measure the presence of pollutant in the effluent, and  
233 operating cost index (OCI), that evaluates operation expenses in the plant. An additional  
234 index, the total cost index (TCI) linking effluent quality and operating cost indexes, is  
235 considered as objective function by weighting method (Flores-Alsina et al. 2008).

#### 236 *Effluent quality index (EQI)*

237 The effluent quality index (EQI) (Copp 2002; Foscoliano et al. 2016) quantifies into a  
238 single term, the effluent pollution load into a receiving water body (kg pollution/day)  
239 (Vanrolleghem and Gillot 2002). The discharged of different pollutants into the effluent  
240 is considered as a weighted sum of six evaluation criteria: total Kjeldahl nitrogen (TKN),  
241 COD, BOD, TSS, nitrate (NO) and total phosphorous (P). Equation 5 shows the weighted  
242 sum of each evaluation criteria where  $Q_e$  is the effluent flow rate.

$$\begin{aligned} 243 \text{EQI} \left( \text{Kg} \frac{\text{pollution}}{\text{day}} \right) &= \left[ 20 \times \text{TKN} \left( \frac{\text{g}}{\text{m}^3} \right) + 1 \times \text{COD} \left( \frac{\text{g}}{\text{m}^3} \right) + 2 \times \text{BOD} \left( \frac{\text{g}}{\text{m}^3} \right) + 2 \times \right. \\ 244 \text{TSS} \left( \frac{\text{g}}{\text{m}^3} \right) &+ 20 \times \text{NO} \left( \frac{\text{g}}{\text{m}^3} \right) + 100 \times \text{P} \left( \frac{\text{g}}{\text{m}^3} \right) \left. \right] \times Q_e \left( \frac{\text{m}^3}{\text{day}} \right) \times 10^{-3} \end{aligned} \quad (5)$$

245

#### 246 *Operating cost index (OCI)*

247 The operating cost index (OCI) that is used as the economic objective function, is an  
248 approximate measure of the plant's operational cost, involves the main operating costs,  
249 such as power consumption by aerators, sludge production, pumping energy and nutrient  
250 cost. The operating cost index (OCI) (equation 6) is calculated as the weighted sum of  
251 aeration energy associated to the oxygen consumption for the carbonaceous demand  
252 (CD), aeration energy associated to the oxygen consumption for the nitrogenous demand  
253 (ND), mixing energy (ME), pumping energy (PE), sludge production (SP) and nutrient  
254 cost (NC), using the weighting factors as Chen et al. 2015 and Zhou et al. 2015.

$$255 \text{OCI (€/year)} = 25 \times (\text{CD} + \text{ND} + \text{ME} + \text{PE}) + 75 \times \text{SP} + \text{NC} \quad (6)$$

#### 256 *Total cost index (TCI)*

257 Total cost index (TCI) is defined in equation 7 as the sum of the effluent quality index  
258 and operating cost index optimized (Vanrolleghem and Gillot 2002; Kim et al. 2015).

$$259 \text{TCI (€/year)} = 50 \times \text{EQI (Kg pollution/day)} + \text{OCI (€/year)} \quad (7)$$

260 Along the paper the values of EQI, OCI and TCI were calculated per m<sup>3</sup> of wastewater  
261 treated (Guerrero et al. 2011) in order to maintain the confidentiality of the industrial  
262 effluent characterization.

### 263 **3.4. Decision making process**

264 The proposed optimization strategy produces one optimal solution for each alternative  
265 and for each selected objective function. The variables included in the step 1 are related  
266 to operational conditions that are essential to obtain a minimum AS performance process.  
267 The last part of the methodology considers refinement conditions by including some

268 variables related to the biological processes to avoid risks of inefficiencies in the  
269 downstream liquid-solid separation processes; these variables can be used as decision  
270 making process. Even though most of the optimization models do not include priority  
271 levels such as essential operational variables and refinement operation variables, the  
272 incorporation of this type of prioritization allows extending the use of the methodology  
273 under circumstances where refinement variables or bounds can be skipped. The AS and  
274 clarifying units of the BAS process can run even without fulfilling the bounds of some of  
275 these refinement conditions but, generally, fulfilling them improve the behaviour of the  
276 BAS process.

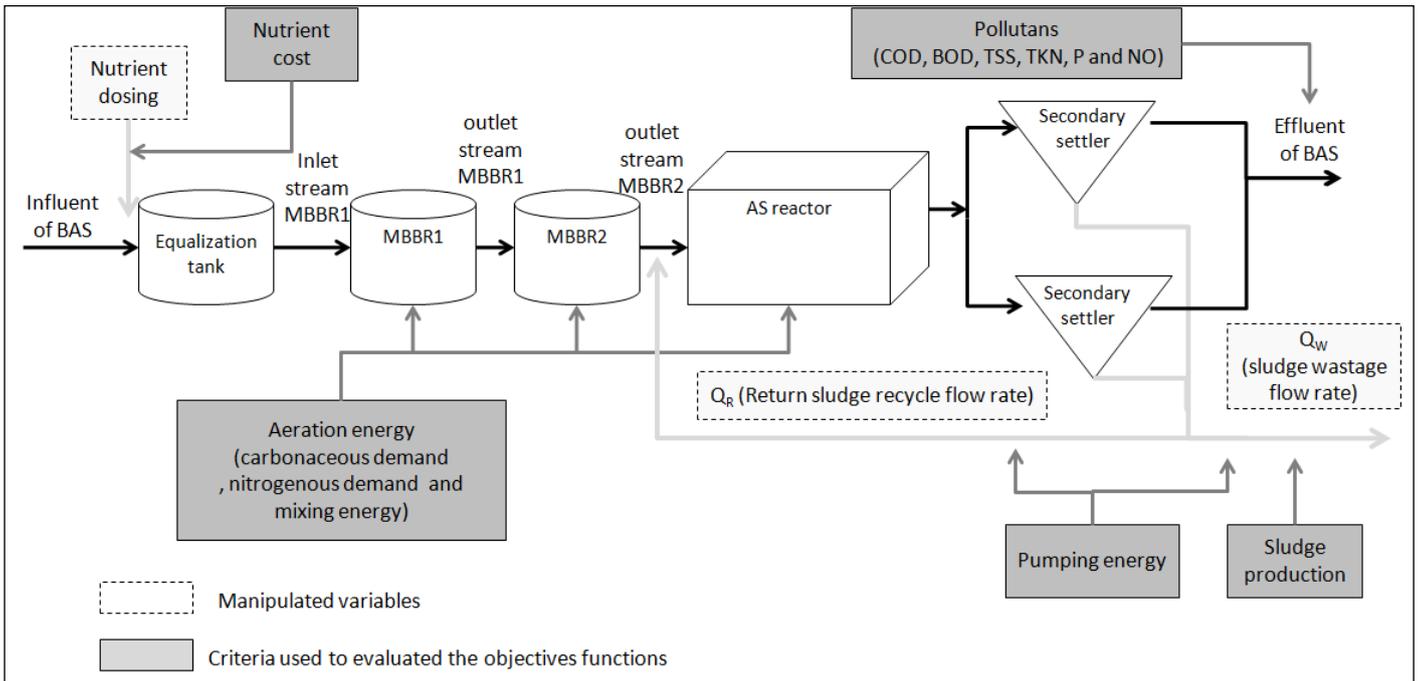
277 The last unit of the BAS process is the separation of sludge from wastewater in the  
278 secondary settler since most of the sludge is returned to AS tank while a minor part is  
279 taken out to be wasted. The separation of the sludge depends on the microorganism  
280 growing in large aggregate called flocs; however, the microorganisms do not always grow  
281 in the adequate form leading to sludge separation problems.

282 Avoiding the risk of separation solid-liquid problems in the secondary settler and loss of  
283 COD removal efficiency in the overall BAS process were selected as refinement o  
284 decision making requirements (Comas et al. 2008; Flores-Alsina et al. 2009). In the  
285 present work, these aspects have been tackled by considering some variables in the unit  
286 models and by adapting the upper or lower limits of these variables to the wastewater and  
287 process under study, as pointed out in the fourth step of Figure 2.

288 If after the decision making process none optimal conditions have been obtained it is  
289 necessary to come back to step 1 (generation of alternatives) to extend the number of  
290 alternatives or to come back to step 4 to extend the bounds of the decision-making  
291 variables.

292 **4. Industrial installation**

293 Veolia have installed more than 90 BAS plants worldwide to treat wastewater from pulp  
294 industry. In this work, the full-scale BAS process for biological COD removal consist of:  
295 an equalization tank (1,600 m<sup>3</sup>), two MBBR reactors in-series (5,331 m<sup>3</sup>), an AS reactor  
296 (47,000 m<sup>3</sup>), two parallel secondary settler tanks (volume 4,143 m<sup>3</sup> and 1,017 m<sup>2</sup> unit  
297 surface area) and one splitter to recirculate part of the sludge and to disposal the remaining  
298 part (Figure 3). The equalization tank is also used to dose nitrogen as urea (40% w/w) and  
299 phosphorous as phosphoric acid (72%). The MBBR reactors were filled with carriers type  
300 BiofilmChip P of Veolia's AnoxKaldnes™ to 10% of volume. The aeration system in  
301 MBBR reactors uses a blower of a unit air flow 31,600 Nm<sup>3</sup>/h for each MBBR reactor  
302 and perforated tubes in the bottom of reactor to produce medium bubbles (3 mm of  
303 diameter) with a high mixing capacity and medium oxygen transfer efficiency (16 %).  
304 The aeration system of AS reactor uses one blowers of a unit air flow 31,600 Nm<sup>3</sup>/h and  
305 diffusers of membrane to produce fine bubbles (1 mm of diameter) with a high mixing  
306 capacity and high oxygen transfer efficiency (45.5 %). Due to the high oxygen transfer  
307 efficiency in the AS reactor, the airflow supplied by the blower associated to the oxygen  
308 consumption for the carbonaceous (CD) and nitrogenous demand (ND) is lower than in  
309 the MBBR reactors resulting in a lower energy consumption (von Sperling 2007).



311 Figure 3. Flow sheet of the full-scale BAS process with detail of the manipulated variables  
 312 and criteria used to evaluate the objective functions.

313

314 The full-scale real BAS plant works under two different conditions (case-study A and B)  
 315 due to different industrial production requirements of the pulp integrated plant. The case-  
 316 study A treats wastewater from viscose and cellulose industry and the case-study B treats  
 317 wastewater from cellulose industry. The regular operational conditions of the studied  
 318 industrial BAS process (Table 1) are based on the industrial heuristic knowledge.

319 The low values of nutrient dosage used in this work in comparison with the “thumb rule”  
 320 (100 COD:5 N:1 P) are explained by the large amounts of nutrients that are regenerated  
 321 in the AS reactor (Comeau et al. 2003) since the biomass is consumed by predator  
 322 microorganisms.

323 Table 1 additionally lists a summary of the main outlet stream characteristics obtained  
 324 working at these regular operational conditions that reach a high COD removal

325 percentage of 76% and 85% for case studies A and B respectively. Besides, the values of  
 326 the objective indexes obtained working at regular operational conditions appear at the end  
 327 of Table 1.

328 Table 1.Characteristics of *regular working conditions* for the two case-studies of the BAS  
 329 process together with the variables and indexes obtained for these conditions.

Parameters of the treatment plant at regular operation conditions		Case-study A	Case-study B
<b>Influent conditions</b>	Wastewater origin $Q_i$ (m <sup>3</sup> /day)	Viscose & Cellulose 1.0 q*	Cellulose 0.59 q*
<b>Manipulated variables</b>	vd (hours) COD <sub>f</sub> :N:P R (%) SRT (Days) $Q_w$ (m <sup>3</sup> /day)	3.12 100:2.14:0.28 110 19 1,082	6.07 100:1.13:0.24 80 30 636
<b>Key variables</b>	COD removal percentage Sludge yield (Tn TSS/Tn COD removed) Efficiency (Tn COD removed/day)	76 0.207 35.4	85 0.155 28.3
<b>Objective indexes</b>	TCI (Total cost index) (€m <sup>3</sup> ) EQI (Effluent quality index (Kg pollution/m <sup>3</sup> ) OCI (Operating cost index) (€m <sup>3</sup> )	0.431 1.790 0.186	0.401 0.861 0.245

330 \*Reference value: q for  $Q_i$ .

331

332 It is important to remark that hydraulic retention time in the secondary settler (vd) for a  
 333 well-driven BAS plant should be between 1-3 hours (van Haandel and van der Lubbe  
 334 2015); however, vd is 6.07 hours in case-study B, therefore, in the present paper for  
 335 optimization purpose, only one secondary settler tank is considered in the case-study B  
 336 since the usage of two secondary settlers would give infeasible solutions.

## 337 5. Results and discussion

### 338 5.1. Generation of alternatives

339 Nutrient needs to be dosed in the influent of viscose and cellulose wastewater since this  
 340 type of wastewater do not contain them. Correct balance of nutrient dosage is crucial  
 341 considering that: i) *nutrient overdosing* can produce effluents with high nitrogen and  
 342 phosphorus discharges into the sewer (Malmqvist et al. 2007) or unwanted nitrifying

343 bacteria, which consume oxygen and generates additional liquid-solid separation problem  
344 due to denitrification in secondary settler tanks (Henze 2008), ii) severe deficiency of  
345 nutrient results in a loss of COD removal efficiency and bulking (van Haandel and van  
346 der Lubbe 2015; Welander et al. 2002) and, iii) *nutrient dosage* also influences the sludge  
347 production and reduction on dosage can result in growth limitation in the MBBR reactors  
348 and a consequent reduction of the waste sludge production (Welander et al. 2002).

349 Nutrient dosage is incorporated in the mathematical model in order to generate a wide  
350 number of alternatives. The dosage used in the regular operation condition is considered  
351 the starting point for the generation of alternatives (100:2.14:0.28 for case study A and  
352 100:1.13:0.24 for case-study B); and the alternatives are generated by decreasing nitrogen  
353 and phosphorous dosage by 5% until the efficiency of the process (Tn de COD removed  
354 by day) decreases more than 2.0% in relation to the regular operation conditions. In this  
355 way, in case-study A, 17 different alternatives of nutrient dosage are generated until  
356 nutrient dosage value of 100:0.34:0.06 and in case study B, 15 different alternatives are  
357 generated until nutrient dosage value of 100:0.34:0.07. Alternatives generated are enough  
358 for this study but higher number of alternatives can be easily considered if necessary when  
359 the characteristics of the processes are particularly sensitive.

## 360 **5.2. Simulation of the MBBR reactors until steady-state**

361 The MBBR reactors of the treatment plant under-study are simulated using Aspen Custom  
362 Modeler software to describe the dynamic and longitudinal behaviour of the reactors. The  
363 generated alternatives with different nutrient dosage are simulated during 30 days until  
364 steady-state is reached. Once steady-state is attained, the simulated results obtained at the  
365 outlet stream of the second MBBR reactor are sent to optimization software to obtain the  
366 optimal conditions of the remaining BAS process under-study.

### 367 **5.3. Multicriteria optimization**

368 The BAS process is optimized for each alternative minimizing total cost index, which  
369 include operation cost and wastewater quality criteria. As one optimal solution is obtained  
370 for each alternative, 15 and 17 optimal solutions are obtained for case-study A and B  
371 respectively. In general, it is observed that the decrease of nutrient dosage has a big impact  
372 on objective index. The TCI lowest score alternative occurs for the lowest nutrient  
373 dosage: 100:0.34:0.06 and 100:0.34:0.07 for case-study A and B respectively (TCI=0.208  
374 €/m<sup>3</sup>, TCI=0.244 €/m<sup>3</sup>).

375 Table 2 shows the optimal values of selected variables for the lowest score alternative  
376 using TCI as objective function; the key characteristic of BAS process is the low sludge  
377 yield (Tn TSS/ Tn COD<sub>f</sub> removed) without compromising treatment efficiency, and  
378 therefore the sludge yield and efficiency appears as key variables. First of all, it is  
379 observed that the comparison between the key variables in Table 1 (regular operational  
380 conditions) and Table 2 (optimal values using TCI as objective function) shows that the  
381 optimal conditions decrease the sludge yield to the half respect to the regular operational  
382 conditions (from 0.207 to 0.108 Tn TSS/Tn COD removed) and a little decrease of the  
383 efficiency is also observed (from 35.4 to 34.5 Tn COD removed /day) for case-study A.  
384 For case study B, similar decreases are observed for the best alternative: sludge yield from  
385 0.155 to 0.082 and efficiency decrease from 28.3 to 27.9Tn COD removed /day.

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388

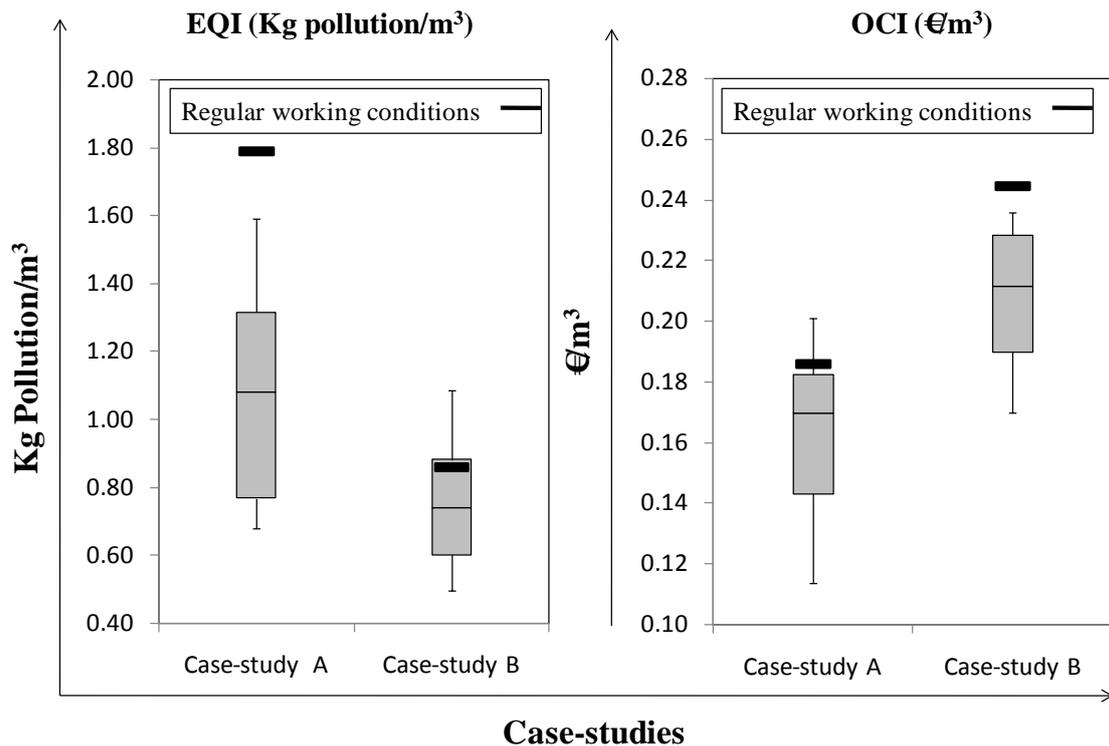
389

390 Table 2. Optimal values of the operation conditions for the best alternative minimizing  
 391 total cost index (TCI) *before* and *after* the decision making process.

Variables and Index  Optimal solutions	Optimal values <i>before</i> decision making process		Optimal values <i>after</i> decision making process	
	Case study A	Case study B	Case study A	Case study B
<b>Manipulated Variables</b>				
COD <sub>r</sub> :N:P	100:0.34:0.06	100:0.34:0.07	100:0.54:0.07	100:0.45:0.10
Q <sub>w</sub> (m <sup>3</sup> /day)	461	313	1.218	655
R (%)	109	82	109	82
<b>Objective Indexes</b>				
TCI (€m <sup>3</sup> )	0.208	0.244	0.223	0.270
EQI (Kg pollution/m <sup>3</sup> )	0.684	0.541	0.680	0.593
OCI (€m <sup>3</sup> )	0.114	0.170	0.130	0.189
<b>Key variables</b>				
Sludge yield (Tn TSS/Tn COD removed)	0.108	0.082	0.155	0.108
Efficiency (Tn COD removed/day)	34.5	27.9	35.4	28.3
<b>Technical decision making variables</b>				
SRT (days)	39	<b>51</b>	17	28
NO (g/m <sup>3</sup> )	5.4	1.8	7	3.8
TN (g/m <sup>3</sup> )	5.9	<b>1.9</b>	6.5	4
P (g/m <sup>3</sup> )	<b>0.1</b>	<b>0.2</b>	0.3	0.6

392 **Value:** Values that not fulfil the refine operation bounds.

393 Figure 4 shows in a box plot fashion the summary of effluent quality and operating cost  
 394 indexes for all studied alternatives when total cost index is minimized. The lowest values  
 395 of EQI and OCI correspond with the lowest nutrient dosage. Figure 4 also compares the  
 396 optimal results with the results of the regular operational conditions. It is observed that  
 397 for nearly all of the alternatives the operating cost and the effluent quality index values  
 398 are lower than the regular operational conditions in both case studies.



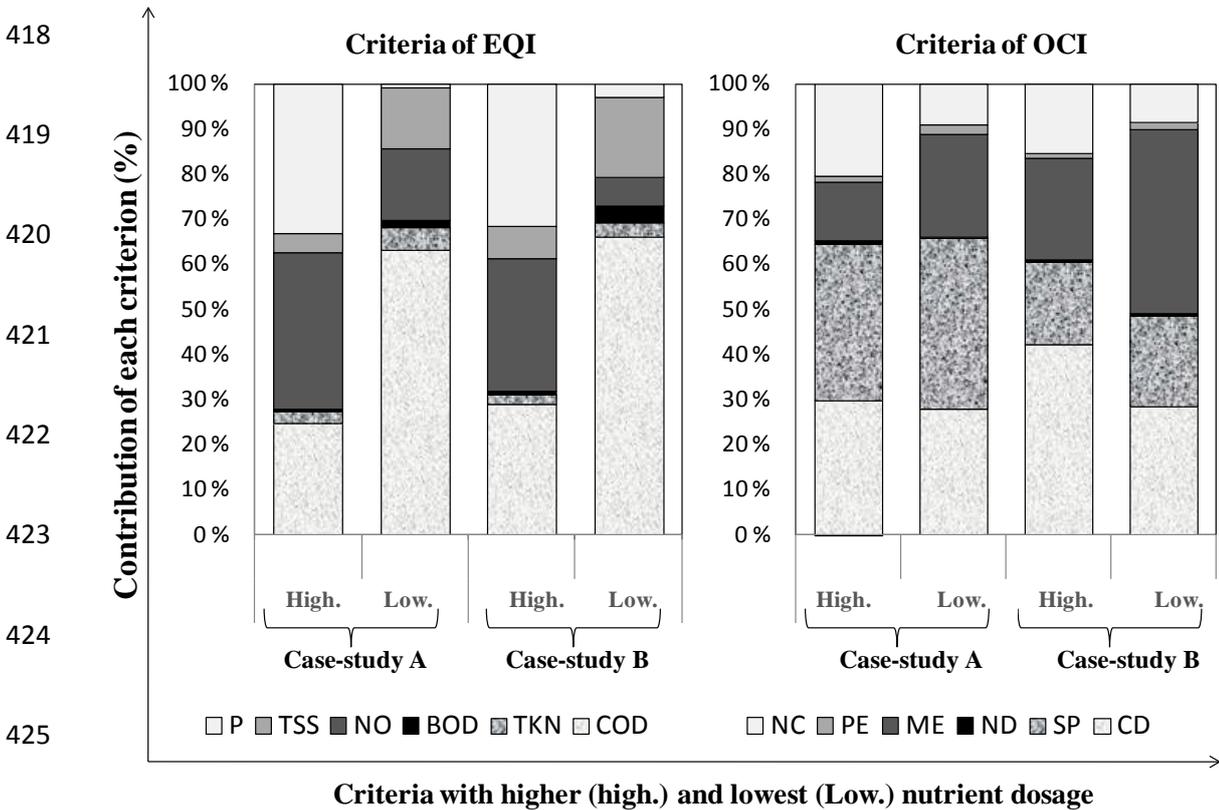
399

400 Figure 4. Box plots for the distribution of EQI and OCI indexes in case-study A (17  
 401 alternatives) and case-study B (15 alternatives) when TCI is minimized, and the  
 402 comparison with the results at the regular working conditions.

403

404 Figure 5 show the contribution of each particular criterion on the effluent quality (EQI)  
 405 and operating cost (OCI) indexes for the lowest and the highest values of nutrient dosage  
 406 when TCI in minimized. In general, the lower the nutrient dosage in the influent, the  
 407 lower  $Q_w$  is purged and higher SRT is obtained and consequently: i) less nitrogen and  
 408 phosphorous are discharged in the effluent and less nitrogen is available in the AS reactor  
 409 resulting in a decrease of nitrification rate and, therefore, less nitrogen is oxidized to  
 410 nitrate (NO) by autotrophic microorganisms and, ii) more TSS is discharged in effluent  
 411 resulting in increase of COD and BOD. For these reasons, when the nutrient dosage is the  
 412 lowest, COD contributes up to 63% and 66 % of the pollution discharge in case-study A  
 413 and B respectively and NO contributes 7-16%. When the nutrient dosage is the highest,

414 the COD contributes 25-30%, NO contributes 30-35% and P contributes 31-33%. In  
 415 relation to OCI there is a lower influence of the nutrient concentration; in both case-study,  
 416 it is remarkable that when the nutrient dosage is the lowest, CD is reduced 2-14% due to  
 417 airflow supplied in AS reactor is lower than in MBBR reactor.



426

427 Figure 5. Contribution of each criterion on the effluent quality (EQI) and operating cost  
 428 (OCI) indexes in each case-study for the highest (High.) and the lowest (Low.) values of  
 429 nutrient dosage when TCI is minimized.

430

#### 431 5.4. Decision-making process.

432 Decision making process is the last step of the methodology and includes the  
 433 determination of some discrimination variables and their limits. The operational  
 434 constraints and bounds included in the step 1 of the methodology are essential for the  
 435 adequate running of an activated sludge process; however, there are other variables whose

436 limits are recommendable but that nonetheless under diverse circumstances do not need  
437 to be considered. At present there are more than 600 municipal or industrial Veolia  
438 MBBR treatment plants in operation or under construction in more than 50 countries (van  
439 Haandel and van der Lubbe 2015); some of these installations are used as pre-treatment  
440 of activated sludge since MBBR is an upgrade to existing AS systems with little  
441 disturbance but high efficiency. The diverse circumstances that can take places in each  
442 installation support the idea of classifying the variables into essential and refinement.

443 The consideration of the refinement conditions in the decision making process of the  
444 optimization methodology will help, a) to facilitate the trade-off among effluent quality  
445 index, operating cost index and technical arrangement (Hakanen et al. 2013) and, b) to  
446 obtain BAS process adapted to different operation circumstances, for example: low pH  
447 wastewater, oversize installations, wastewater with different origin, low  
448 Food/Microorganisms (F/M) ratio for AS process and excess of greases and oils among  
449 others.

450 In the activated sludge process, the operational problems with biological origin are among  
451 the most serious and most difficult matter to solve in wastewater treatment plants. Even  
452 though the optimal conditions shown in Table 2 fulfil the appropriated operational  
453 conditions for BAS process of Figure 2, in the refinement conditions three biological  
454 aspects are pointed related with the growth of microorganism: i) “bulking” since too low  
455 nutrient dosage can bring excessive growth of filamentous bacteria (van Haandel and van  
456 der Lubbe 2015; Welander et al. 2002) that leads to solid-liquid separation problems  
457 (Flores-Alsina et al. 2009), ii) “pin-point floc” due to the formation of the old and  
458 overoxidised sludge produce by endogenous metabolism (Comas et al. 2003) and iii)  
459 rising sludge due to denitrification in the secondary settler where nitrates are converted

460 to nitrogen gas (Flores-Alsina et al. 2010). The refinement operation bounds included in  
461 the decision making variables are divided into two categories:

462 *Bulking and decrease of COD removal efficiency*

463 Until now, the nutrient dosage has been considered *in the influent*; however, very low  
464 concentrations of the TN and P *in the effluent* can mean too severe decrease of  
465 nutrient dosage resulting in sludge settleability (bulking) or even in a loss of COD  
466 removal efficiency (van Haandel and van der Lubbe 2015). For this reason, in this  
467 work has taken into account lower limits in the *effluent* for the concentration of TN  
468 of 4 g/m<sup>3</sup> and 0.3 g/m<sup>3</sup> for P as proposed by Welander et al. 2002.

469 It is observed at Table 2 (Bold figures) that the optimal solutions before decision  
470 making process do not fulfil some of the P or/and TN values in the *effluent* and  
471 therefore the undesirable bulking can occur.

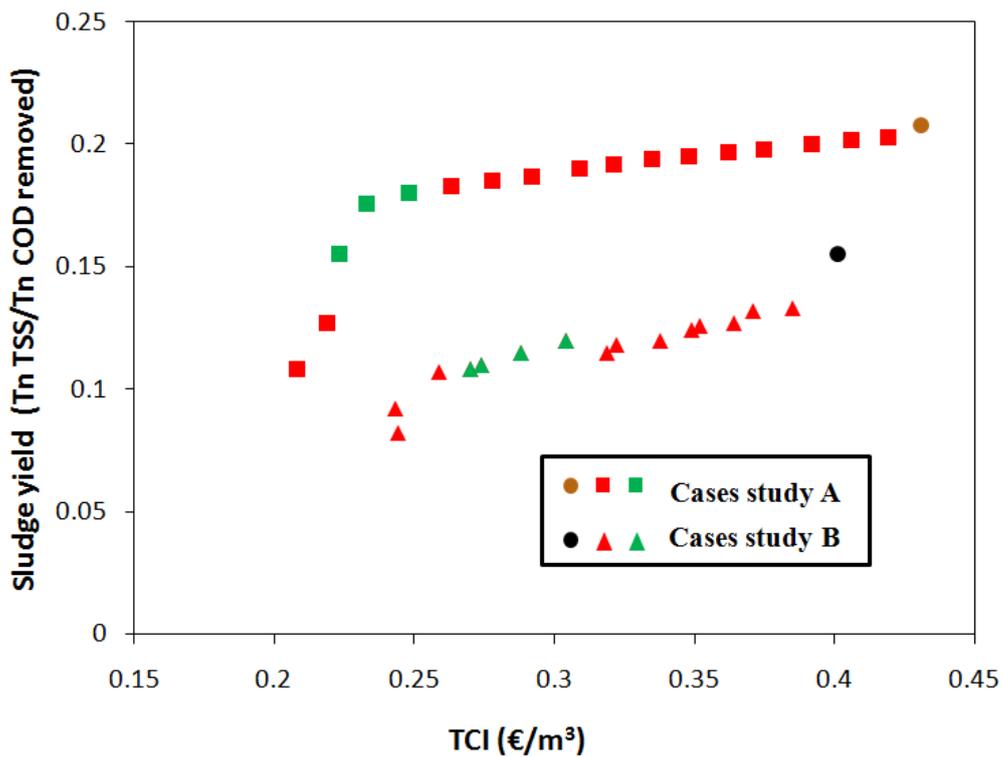
472 *Risk of solid-liquid separation problems due to “pin-point-floc” and rising sludge*

473 In addition to the TN and P bounds described above, two new technical requirements  
474 are now adding (Figure 2): i) concentration of nitrate (NO) and, ii) SRT in the AS  
475 reactor of the BAS process. Nitrate concentration in the secondary settler higher than  
476 8 g/m<sup>3</sup> (Henze et al. 1993) is not recommended since the bottom layer of the  
477 secondary settler (where nitrogen bubble formation through biological  
478 denitrification) can break into small flocs and part of the sludge would flow out into  
479 the effluent; furthermore, an increase of COD, BOD, TKN and P in the effluent  
480 would occurs (Flores-Alsina et al. 2010).

481 High values of SRT produce an old and overoxidised sludge with large amount of  
482 inert matter called “pin-point floc” (Comas et al. 2003). 40 days is selected in this  
483 work as upper limit of SRT because the hydraulic retention time (HRT) in AS reactor

484 is greater than 30 hours and it is considered an extended aeration tank which optimal  
 485 SRT value lower or equal to 40 days (Tchobanoglous et al. 2003).

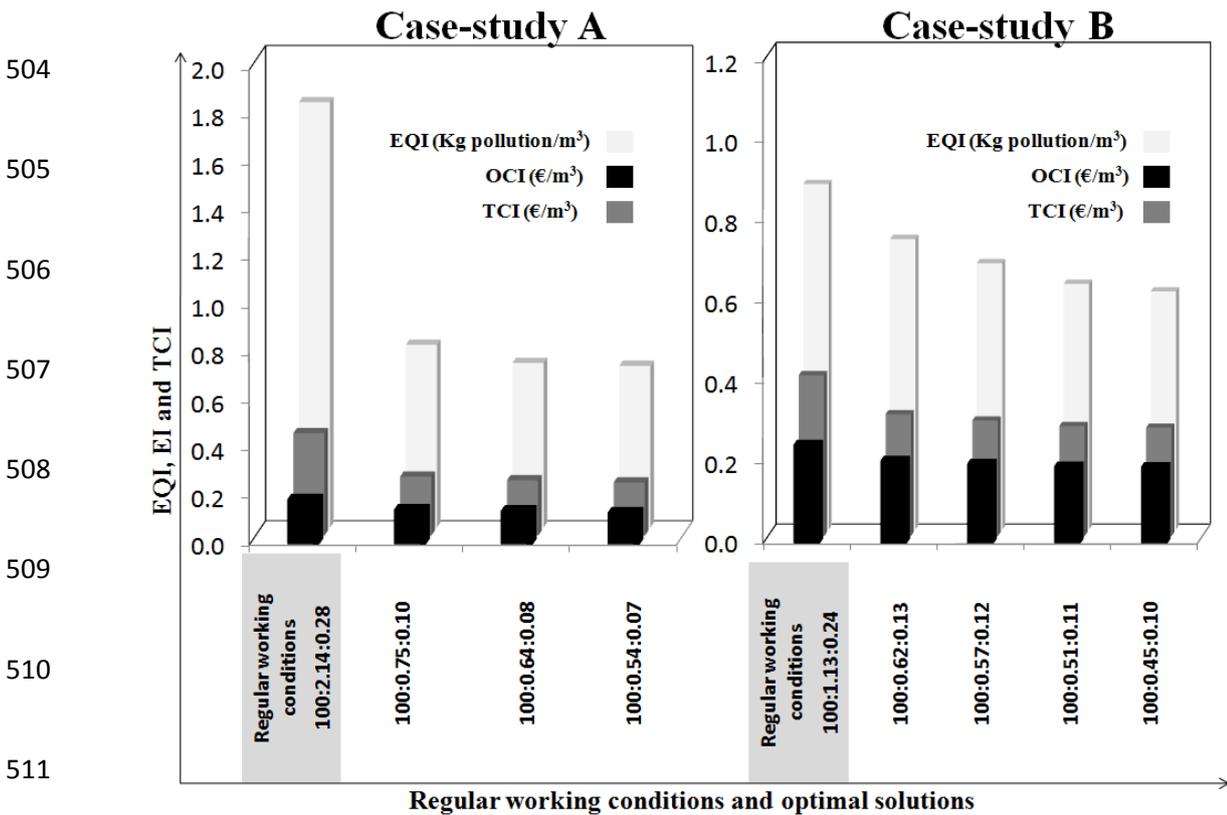
486 Above described operational bounds of the effluent are included in step 4 of the  
 487 optimization methodology shown in Figure 2. Figure 6 shows the pareto graph between  
 488 the sludge yield and TCI index values for all alternatives before (red symbols) and after  
 489 the decision making process (green symbols) as well as the regular operational conditions  
 490 (black and brown symbols). It is observed that in both case-studies the minimum values  
 491 of TCI and sludge yield agree but only few alternatives (green symbols) fulfil the  
 492 refinement bounds.



493

494 Figure 6. Pareto graph between sludge yield and TCI index values before decision making  
 495 process in Case Study A (■) and B (▲) and after the decision making process in Case  
 496 Study A (■) and B (▲). The regular operational conditions are represented in Case Study  
 497 A (●) and Case Study B (●).

498 Figure 7 shows the 3 (case-study A) and 4 (case-study B) alternatives that fulfil the values  
 499 of refinement operation bounds simultaneously and compares the results of these  
 500 alternatives respect to the regular operational conditions. It is observed in Figure 7  
 501 reductions of TCI up to 45% and 25% for case-study A and B respectively in comparison  
 502 with the regular operational conditions, mainly due to the high reductions of EQI (up to  
 503 60% in A and up to 31% in B).



512 Figure 7. Optimal values of the feasible alternatives for the effluent quality index (EQI),  
 513 operating cost index (OCI) and total cost (TCI) index after of the decision making and  
 514 the comparison with the regular operational conditions.

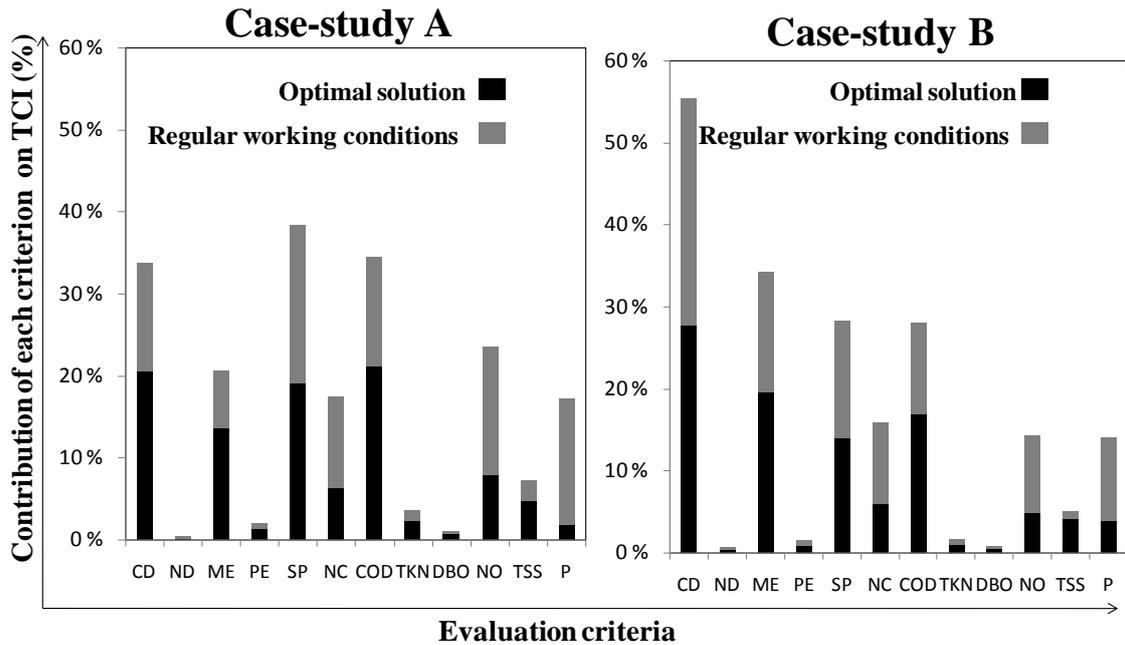
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516 Finally, the right column in Table 2 shows the optimal values of variables for the best  
 517 alternative for case-study A and B after decision making selection. It is observed that the  
 518 nutrient dosage and sludge yield are higher than before to decision making process.

519 Besides, the sludge yields in Table 2 are inside the range of others studies as such as  
520 Malmqvist et al. 2007 and Rankin et al. 2007.

521 The application of the proposed methodology allows a reduction of the operating cost of  
522 treatment ( $\text{€m}^3$ ) up to 30% whereas the efficiency (Tn COD removed per day) do not  
523 decrease respect to the regular operational conditions used in the full-scale wastewater  
524 plant. Furthermore, the quality of the wastewater is also much better since the EQI (Kg  
525 pollution/ $\text{m}^3$ ) is reduced up to 62%.

526 In Figure 8, the different contributions of each evaluation criterion for the best alternative  
527 are shown. The major contribution to effluent quality index (EQI) is the COD (21 % in  
528 case-study A and 17 % in case-study B), since the contribution of NO and P are reduced  
529 in comparison to regular operational conditions due to low nutrient dosage. Respect to  
530 the major contribution to operation cost index (OCI) after decision making is the energy  
531 consumption for carbonaceous demand (CD) (21% in case-study A and 28 % in case-  
532 study B) due to the BAS process in this work was designed to the high removal of COD  
533 in aerobic conditions (Revilla et al. 2016a); moreover, a large decrease in the nutrient cost  
534 (NC) is observed compared to regular operational conditions.



535

536 Figure 8. Contribution of evaluation criteria on the total cost index (TCI) of the optimal  
 537 solution compared to the regular operational conditions.

## 538 5. Conclusions

539 This paper presents a methodology that allows obtaining the optimal operational  
 540 conditions for an industrial BAS plant treating highly COD wastewater under limited  
 541 nutrient conditions. The methodology formulates BAS process as a mathematical  
 542 optimization problem combining economic and environmental criteria as objective  
 543 function and overcomes some challenges such as high number of (non-linear) equations  
 544 and variables and the dynamic and spatial distribution behaviour of components into the  
 545 biofilm. The optimization methodology is divided into four consecutive steps: i)  
 546 generation of alternatives, ii) simulation of the MBBR reactors (biofilm) until steady-  
 547 state, iii) multicriteria optimization and iv) decision making process. The optimization  
 548 methodology establishes priority levels of several technical specifications (essential and  
 549 refinement variables) related with the activated sludge tank and secondary clarifiers.

550 The application of this methodology at two industrial-scale case studies from viscose and  
551 cellulose wastewaters allows a reduction of quantity of pollutants per m<sup>3</sup> wastewater  
552 treated up to 60% and a reduction of the operating costs (€m<sup>3</sup>) up to 30% in comparison  
553 with the regular operational conditions used in the industrial BAS wastewater plant.

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557

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