

# A tool for predicting the dynamic response of biotrickling filters for VOC removal

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## Abstract

This article presents the development of a MATLAB<sup>®</sup> computer program to simulate the performance of biotrickling filters. Since these filters behave differently during spraying and non-spraying cycles, the presented simulation tool is built on top of a mathematical description of each situation. The resulting variable-structure model is then used as the basis for simulation experiments. The model presented herein represents the first attempt to take into account the variable spraying pattern usually found in industrial installations. Overall, the software is flexible and easy to use, allowing the user to specify the emission concentration pattern, the gas concentration pattern, as well as the spraying cycles period for up to two different emission patterns per day. The model is able to predict experimental data from a biotrickling filter treating isopropanol under intermittent conditions of loading and spraying. Simulation examples are then provided to study the effect of variable inlet

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concentration and gas flow rates.

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## 1 **Introduction**

2 Emission to the atmosphere, from a wide variety of sources, of volatile organic com-  
3 pounds (VOCs) remains one of the most important causes of air pollution. This has  
4 triggered significant research efforts to develop more cost-effective and environmentally  
5 friendly solutions for the treatment of air emissions of VOCs. In particular, there has  
6 been an increasing interest in biofiltration, especially since it has been classified as a best  
7 available technique (BAT) by the European Commission (2003). Among the biofiltration  
8 strategies, biotrickling filters (BTFs) constitute one of the most suitable biotechnologies  
9 for the treatment of VOCs. Biotrickling filters consist of a column filled with an inert  
10 packing material where the biomass attaches to the media and develops a biofilm. In this  
11 configuration, the gas and liquid phases circulate through the column in co- or counter-  
12 current mode. Thus, the pollutant and the oxygen are transferred from the gas phase to the  
13 trickling liquid, and then to the biofilm, where the biodegradation takes place.

14 Biotrickling filtration has been applied successfully to the treatment of VOCs at the  
15 laboratory, pilot, and industrial scales. However, to further improve the performance of  
16 BTFs for the treatment of VOCs it has become necessary to understand the intricacies of

17 the processes involved as well as their rate-limiting steps (Popat and Deshusses, 2010). In  
18 this regard, biotrickling filtration involves a complex set of physico-chemical and biolog-  
19 ical mechanisms and, hence, mathematical models, in conjunction with computer-aided  
20 simulation, appear as fundamental tools to go deeper into the understanding of the in-  
21 volved governing processes.

22 Industrial processes that use solvents are characterized by fluctuating VOC emissions  
23 arising from the specific application and the unit operations dynamics of each particular  
24 industry (Rene et al., 2013). These results in emission levels whose time variations are  
25 related to random fluctuations of the gas velocity and the inlet concentration profile. In  
26 addition, short-time shut-off periods associated with nights, weekends and holiday clo-  
27 sures further contribute to create a variable pattern of VOC emissions at the industrial  
28 scale. This variability may sometimes hinder the performance of field-scale BTFs (Sem-  
29 pere et al., 2010). Also, operating BTFs under cyclic and discontinuous operation has  
30 traditionally produced some problems, as reported in Webster et al. (1999).

31 Intermittent water trickling, in contrast to continuous trickling, is also common prac-  
32 tice in the operation of industrial BTFs. As shown in Sempere et al. (2008), intermittent  
33 trickling may improve the removal efficiency and better control the pressure drop. The  
34 final performance of the BTF is quite dependent on the rate of liquid trickling (Zhu et al.,  
35 1998). An intermittent spraying regime implies that the mobile liquid phase is not al-

36 ways present during the filter operation, making it necessary to distinguish two different  
37 situations, corresponding to *spraying* and *non-spraying* periods. Nevertheless, the mod-  
38 elling and simulation research presented in the literature so far tends to focus only on one  
39 particular case.

40 Several efforts have been made to model biofiltration processes. One of the most used  
41 models for the treatment of organic pollutants in waste gases in a gas–liquid biofilter has  
42 been developed by Ottengraf and Van Den Oever (1983) in steady state conditions. Since  
43 then, there has been increasing interest in the application of dynamic models of biofilters  
44 and BTFs rather than of steady state models. Shareefdeen and Baltzis (1994) published one  
45 of the first attempts to describe the dynamic behaviour of biofilters, including the oxygen  
46 limitation in the biofilm and the adsorption phenomena. Deshusses et al. (1995) proposed  
47 a model for the determination of transient and steady-state conditions degrading MEK and  
48 MIK emissions in biofilters. Zarook et al. (1997) developed a transient biofiltration model  
49 that incorporates oxygen limitation effects, general mixing and adsorption phenomena,  
50 as well as general biodegradation reaction kinetics. Thereafter, many researchers intro-  
51 duced variations of these models by adding new considerations. Métris et al. (2001) used  
52 a simplification of the Zarook et al. (1997) model using  $CO_2$  production to evaluate the  
53 response of the biofilters to starvation and shock loads in the biofiltration of toluene and  
54 xylene. Álvarez Hornos et al. (2009) developed a dynamic model with a Haldane-type

55 kinetic expression that considers oxygen limitation, (cross) inhibition effects due to high  
56 concentration of substrates, and a general axial gradient equation for the biomass density.  
57 Many BTF models derive from biofilter models. Okkerse et al. (1999) presented a de-  
58 tailed dynamic model that includes the growth of methylene chloride degraders and inert  
59 biomass as well as the effect of pH and dissolved oxygen. Kim and Deshusses (2003)  
60 presented a three-phase dynamic model to describe the biotrickling filtration of hydrogen  
61 sulfide with a gas and liquid flowing counter-currently. They assumed that the biofilm was  
62 not completely wetted by the liquid phase and thus, in some parts of the biofilm, the pollu-  
63 tant was transferred directly from the gas phase to the biofilm. In their review of biofilters  
64 and biotrickling modelling, Devinny and Ramesh (2005) pointed out that no single model  
65 has become generally accepted. The complexity behind the operation of BTFs has made  
66 many researchers consider specific situations in their simulation studies (Lee and Heber,  
67 2010; Mannucci et al., 2012).

68 The increase in the number of factors taken into consideration in the mathematical  
69 models has necessitated greater efforts for their mathematical solution. In the case of  
70 models of biotrickling filters, the presence of the liquid phase implies an increase of the  
71 level of complexity and for counter current operation, which is usual found in the industry,  
72 the system of equations obtained can be relatively *stiff* and model instabilities could make  
73 their solution difficult (Deshusses and Shareefdeen, 2005). Even so, it has been recognized

74 that realistic models adapted to the emissions of the industry are needed.

75 The aim of this paper is to present a more flexible tool to simulate the performance  
76 of BTFs. Based on the operational conditions commonly found in industry, the proposed  
77 model allows specifying variable inlet concentration patterns and gas velocities combined  
78 with different spraying patterns. These and other features provide the necessary flexibility  
79 to reproduce typical industrial use cases.

## 80 **Model development**

81 Industrial BTFs operate with intermittent water trickling. This means that the mobile  
82 liquid phase is only present at some times during the day, referred to here as the *spraying*  
83 *periods*. For the rest of the time, referred to as the *non-spraying periods*, the liquid phase  
84 remains as a stagnant phase. Figure 1 illustrates this concept. The modelling step has to  
85 take into account the principal mechanisms of the biofiltration process in each situation.  
86 In this configuration, the pollutant/oxygen is transferred from the gas phase to the liquid  
87 phase and then to the biofilm as is represented in Figure 2. The model has been developed  
88 following the general mass balances of gas phase, liquid phase and biofilm by taking into  
89 account the most important phenomena compiled by Devinny and Ramesh (2005).

90 [Figure 1 about here.]

91 [Figure 2 about here.]

92 For the model derivation, the following general assumptions have been made based on  
93 consolidated models reported (Kim and Deshusses, 2003; Mpanias and Baltzis, 1998) and  
94 adapted to this model.

95 (1) The gas phase flows in a plug flow regime along the filter bed.

96 (2) Axial dispersion is neglected.

97 (3) The adsorption of pollutant in the packing material is negligible.

98 (4) The active biofilm is formed on the external surface of the packing material and no  
99 reaction occurs in the pores. The biofilm covers the surface of the packing material  
100 and its thickness ( $\delta$ ) is much smaller than the size of the solid particles, so a planar  
101 geometry has been assumed.

102 (5) The packing material is completely covered by the biofilm.

103 (6) The diffusion of the biofilm is described by Fick's law.

104 (7) Ideal conditions of nutrients and pH are assumed.

105 (8) The system works under cycling conditions of spraying/non-spraying periods.

106 (9) The status reached at the end of one period determines the initial conditions for the  
107 next period.

108 (10) The biodegradation kinetics is described by a Monod expression, which takes into  
109 account the oxygen limitation.

110 (11) The mass flux at the gas-liquid interface can be expressed by mass transfer coeffi-

111 cients.

112 (12) The mass flux at the liquid-biofilm interface can be expressed by mass transfer co-  
113 efficients.

114 (13) There is no reaction in the liquid phase.

115 (14) The gas-liquid interface is in equilibrium according to Henry's law.

116 Based on the assumptions above, the mass balances for the different phases can be  
117 written as follows:

118 *Spray mode*

*Mass balance in the gas phase.*

$$\theta_G \frac{\partial C_{GP}}{\partial t} = -v_G \frac{\partial C_{GP}}{\partial z} - \alpha_1 K_{LaP} \left( \frac{C_{GP}}{H_P} - C_{LP} \right) \quad (1)$$

$$\theta_G \frac{\partial C_{GO}}{\partial t} = -v_G \frac{\partial C_{GO}}{\partial z} - \alpha_1 K_{LaO} \left( \frac{C_{GO}}{H_O} - C_{LO} \right) \quad (2)$$

119 where, for the pollutant and oxygen, respectively,  $C_{GP}$  and  $C_{GO}$  are the concentration in the  
120 gas phase,  $K_{LaP}$  and  $K_{LaO}$  are the overall mass transfer coefficients,  $\alpha_1$  is the correction  
121 factor of the overall mass transfer coefficients,  $H_P$  and  $H_O$  are the dimensionless Henry's  
122 law constants expressed as concentration of the gas phase/ concentration of the liquid  
123 phase,  $C_{LP}$  and  $C_{LO}$  are the concentration of the liquid phase.  $t$  denotes the time,  $z$  is the  
124 distance from the bottom of the column and  $v_G$  is the superficial air velocity given by

$$v_G = \frac{Q_G}{\frac{\pi D^2}{4}} \quad (3)$$



125 where  $Q_G$  is the volumetric gas flow rate and  $D$  is the column diameter.

126  $\theta_G$  is the porosity of the bioreactor and is given by

$$\theta_G = 1 - (1 - \theta_{pm}) - \theta_L - \theta_B \quad (4)$$

127 where  $\theta_{pm}$  is the void fraction of the packing material,  $\theta_L$  is the fraction occupied by the  
128 liquid film and  $\theta_B$  is the fraction occupied by the biofilm.

129

130 The boundary conditions of Equations 1 and 2 are

$$\begin{aligned} C_{G_P} &= C_{G_P}^{in} \quad \text{at } z = 0 \\ C_{G_O} &= C_{G_O}^{in} \quad \text{at } z = 0 \end{aligned} \quad (5)$$

131 where  $C_{G_P}^{in}$  and  $C_{G_O}^{in}$  are the inlet concentrations in the gas phase of the pollutant and the  
132 oxygen, respectively.

*Mass balance in the liquid phase.*

$$\theta_L \frac{\partial C_{LP}}{\partial t} = v_L \frac{\partial C_{LP}}{\partial z} + \alpha_1 K_{LAP} \left( \frac{C_{G_P}}{H_P} - C_{LP} \right) - \frac{D_{wP} A}{\beta} (C_{LP} - S_{P_1}) \quad (6)$$

$$\theta_L \frac{\partial C_{LO}}{\partial t} = v_L \frac{\partial C_{LO}}{\partial z} + \alpha_1 K_{LAO} \left( \frac{C_{G_O}}{H_O} - C_{LO} \right) - \frac{D_{wO} A}{\beta} (C_{LO} - S_{O_1}) \quad (7)$$

133 where, for the pollutant and oxygen, respectively,  $S_{P_1}$  and  $S_{O_1}$  are the concentration in the  
134 biofilm interface,  $\beta$  is the thickness of the liquid-biofilm interface,  $D_{wP}$  and  $D_{wO}$  are the  
135 diffusion coefficient in water,  $A$  is the specific surface area,  $x$  is the axial position along  
136 the biofilm, and  $v_L$  is the superficial liquid velocity given by

$$v_L = \frac{Q_L}{\frac{\pi D^2}{4}} \quad (8)$$

137 where  $Q_L$  is the volumetric liquid flow rate.

138

139 The boundary conditions of Equations 6 and 7 are

$$\begin{aligned} \frac{\partial C_{LP}}{\partial t} &= \frac{Q_L}{V_T} (C_{LP(z=0)} - C_{LP(z=Z)}) \quad \text{at } z = Z \\ \frac{\partial C_{LO}}{\partial t} &= \frac{Q_L}{V_T} (C_{LO(z=0)} - C_{LO(z=Z)}) \quad \text{at } z = Z \end{aligned} \quad (9)$$

140 The boundary conditions given by Equation 9 correspond to the mass balances in the  
 141 recirculation tank, where  $V_T$  is the water volume in the recirculation tank. It is assumed that  
 142 the liquid inlet concentration in the column is equal to the concentration in the recirculation  
 143 tank, and that the recirculated water depends on the liquid concentration at the bottom of  
 144 the column.

*Mass balance in the biofilm.*

$$\frac{\partial S_P}{\partial t} = D_{PB} \frac{\partial^2 S_P}{\partial x^2} - \frac{\mu_{\max} X_v}{Y_P} \frac{S_P}{S_P + K_P} \frac{S_O}{S_O + K_O} \quad (10)$$

$$\frac{\partial S_O}{\partial t} = D_{OB} \frac{\partial^2 S_O}{\partial x^2} - \frac{\mu_{\max} X_v}{Y_O} \frac{S_P}{S_P + K_P} \frac{S_O}{S_O + K_O} \quad (11)$$

145 where  $S_P$  and  $S_O$  are the concentration in the biofilm. The boundary conditions are given

146 by

$$\frac{\partial S_P}{\partial t} = 0 \quad \text{at } x = \delta \quad (12)$$

$$\frac{\partial S_O}{\partial t} = 0 \quad \text{at } x = \delta$$

147 where  $X_v$  is the concentration of the biomass,  $\mu_{\max}$  is the specific growth rate of the  
148 biomass and, for the pollutant and oxygen, respectively,  $K_{S_P}$  and  $K_{S_O}$  are the half-saturation  
149 constants,  $Y_P$  and  $Y_O$  are the yield coefficients and  $D_{P_B}$  and  $D_{O_B}$  are the effective diffusion  
150 coefficients inside the biofilm corrected with a factor ( $f(X_v)$ ) calculated according to Fan's  
151 equation (Fan et al., 1990):

$$f(X_v) = \left( 1 - \frac{0.43(X_v 10^{-3})^{0.92}}{11.19 + 0.27(X_v 10^{-3})^{0.99}} \right) \quad (13)$$

152 *Non-spray mode*

153 Analogously, the mass balances during non-spraying periods are

*Mass balance in the gas phase.*

$$\theta_G \frac{\partial C_{G_P}}{\partial t} = -v_G \frac{\partial C_{G_P}}{\partial z} - \alpha_2 \alpha_1 K_{L_A P} \left( \frac{C_{G_P}}{H_P} - C_{L_P} \right) \quad (14)$$

$$\theta_G \frac{\partial C_{G_O}}{\partial t} = -v_G \frac{\partial C_{G_O}}{\partial z} - \alpha_2 \alpha_1 K_{L_A O} \left( \frac{C_{G_O}}{H_O} - C_{L_O} \right) \quad (15)$$

154 with the boundary conditions

$$C_{G_P} = C_{G_P}^{in} \quad \text{at } z = 0 \quad (16)$$

$$C_{G_O} = C_{G_O}^{in} \quad \text{at } z = 0$$

155 where  $\alpha_2$  is a switch model parameter (100 indicates that no mass transfer resistance is  
 156 assumed between gas and liquid phase and 1 indicates that there are mass transfer resis-  
 157 tance).

*Mass balance in the liquid phase.*

$$\theta_L \frac{\partial C_{L_P}}{\partial t} = \alpha_2 \alpha_1 K_{L_A P} \left( \frac{C_{G_P}}{H_P} - C_{L_P} \right) - \frac{D_{w_P} A}{\beta} (C_{L_P} - S_{P_1}) \quad (17)$$

$$\theta_L \frac{\partial C_{L_O}}{\partial t} = \alpha_2 \alpha_1 K_{L_A O} \left( \frac{C_{G_O}}{H_O} - C_{L_O} \right) - \frac{D_{w_O} A}{\beta} (C_{L_O} - S_{O_1}) \quad (18)$$

*Mass balance in the biofilm.*

$$\frac{\partial S_P}{\partial t} = D_{P_B} \frac{\partial^2 S_P}{\partial x^2} - \frac{\mu_{\max} X_v}{Y_P} \frac{S_P}{S_P + K_P} \frac{S_O}{S_O + K_O} \quad (19)$$

$$\frac{\partial S_O}{\partial t} = D_{O_B} \frac{\partial^2 S_O}{\partial x^2} - \frac{\mu_{\max} X_v}{Y_O} \frac{S_P}{S_P + K_P} \frac{S_O}{S_O + K_O} \quad (20)$$

158 with the boundary conditions

$$\frac{\partial S_P}{\partial t} = 0 \quad \text{at } x = \delta \quad (21)$$

$$\frac{\partial S_O}{\partial t} = 0 \quad \text{at } x = \delta$$

159 **Numerical solution**

160 The partial differential equations (1), (2), (6), (7), (10), (11) (spray mode), and (14),  
161 (15), (17), (18), (19), and (20) (non-spray mode) constitute two second order nonlin-  
162 ear distributed systems. In order to solve them, the method of lines (MOL) (Schiesser,  
163 1991, 1994; Schiesser and Griffiths, 2009) has been chosen. Although the finite difference  
164 method (FDM) has previously been used in the literature to simulate biofilter and biotrick-  
165 ling filters (Ikemoto et al., 2006; Álvarez Hornos et al., 2009) in different ways, the MOL  
166 has some advantages that make it more suitable here. Apart from its simplicity, it allows  
167 taking advantage of the available ODE solvers. Note, in addition, that the overall MOL  
168 process can be regarded as an FDM procedure where the discretization in  $t$  is independent  
169 of that in  $x, z$ , which provides extra flexibility. Since the resulting systems have been found  
170 to be *stiff*, as is normally the case when applying the MOL (Schiesser, 1994), the ODE23t  
171 solver from the MATLAB<sup>®</sup> has been selected for solving the corresponding equations.  
172 The ODE23t is based on an implicit integration method and it is quite concerned with the  
173 stability issue. Other ODE solvers were tested, but the reported ODE gave the best results  
174 in practice. The MOL method is applied here following the steps:

- 175 • Generate a uniform grid in the space dimensions, i.e.  $(x_i, z_j)_{i,j}$ , where it is going  
176 to find an approximate solution.  $Z$ , the height of the column (the  $z$  axis), is divided  
177 into  $N$  sections. Similarly, the biofilm thickness  $\delta$  is divided into  $M$  sections with

178  $M + 1$  points. Values of  $N = 20, M = 40$  are used for the spatial discretization in  
179 each mode.

- 180 • For each node in the grid, substitute the partial derivatives in the model equations  
181 with finite difference approximations.
- 182 • Solve the resulting system of ordinary differential equations (ODE) using standard  
183 numerical methods; note that the time variable  $t$  was left continuous in the first step.

#### 184 **Developed software tool**

185 The main objective of this paper is to introduce a tool for the simulation of biotrickling  
186 filters using the mathematical models and numerical procedures described in the previous  
187 sections. This section describes the basic features implemented in the presented tool,  
188 focusing on its usability. The software has been developed in MATLAB<sup>®</sup>. It can be used  
189 with the basic MATLAB<sup>®</sup> package and it is available as a MATLAB package as well as  
190 a compiled standalone application. The graphical user interface (GUI) has been created  
191 using the GUIDE–MATLAB<sup>®</sup> toolbox. A screenshot of the GUI is shown in Figure 3.  
192 In the present example, the option *two emissions pattern (per day)* allows specifying two  
193 different patterns of inlet concentration, gas velocity, and spraying, over a period of 86,400  
194 seconds (i.e., one day). When this option is marked, the user indicates the duration of the  
195 first pattern ( $< 86,400$  seconds). The duration of the second pattern is then calculated

196 automatically (86,400 seconds – time pattern 1). The resulting global daily pattern is the  
197 combination of the two specified patterns in series, and the total simulation time in this  
198 case equals the number of days specified by the user.

199 [Figure 3 about here.]

200 The emission pattern and the spraying pattern are defined by the user by

201 ● VOC inlet concentrations. For the inlet VOC concentration ( $C_{Gp}^{in}$ ) pattern, a drop-  
202 down list presents the user with the following options for the input profile:

- 203 – Constant. The inlet concentration is assumed constant.
- 204 – Ramp+Constant. A constant concentration is considered as before, but pre-  
205 ceded by a ramp profile until the final value is reached.
- 206 – Pulse train. The inlet concentration oscillates between two values, describing  
207 a pulse train input signal.
- 208 – Piecewise constant. The inlet concentration consists of a step (or staircase)  
209 function, i.e., it is piecewise constant having only finitely many pieces.

210 After making a choice, a dialog window allows introducing the defining parameters  
211 for each case. For instance, for the Ramp+Constant profile, Figure 4 shows the  
212 resulting dialog.

213

[Figure 4 about here.]

214

In contrast with the inlet VOC concentration, the inlet oxygen concentration ( $C_{G_O}^{in}$ ) is assumed constant throughout the whole simulation ( $276 \text{ g m}^{-3}$ ).

215

216

- Inlet gas flow pattern. This consists of a step (or staircase) function, i.e., it is piecewise constant having only finitely many pieces.

217

218

- Spray settings. The spraying pattern will consist of an ON/OFF signal. As an example, the scheme of the spraying pattern for the option *two emissions pattern (per day)* is illustrated in Figure 5.

219

220

221

[Figure 5 about here.]

222

The spraying panel includes the following information.

223

- Number of spray cycles ( $n$ ). Defines how many times to spray during each emission pattern.

224

225

- Spraying time ( $T_s$ ). Duration of spraying, i.e., the duration of the ON part of one spray cycle.

226

227

Simulation requires the user's specifying the initial conditions:



228 • Initial conditions for the simulation experiment. This includes the VOC concen-  
229 tration in the liquid phase, the VOC concentration in the water tank, and the VOC  
230 concentration inside the biofilm and the oxygen concentration in the liquid concen-  
231 tration and inside the biofilm.

232 Input related to the BTF configuration, the pollutant and packing material data, and the  
233 model parameters are defined by the user:

234 • BTF set-up. This part defines the characteristics of the BTF system, such as the  
235 column diameter, column height, and the volume of the water tank. This panel  
236 provides automatically the column volume of the reactors.

237 • Physical properties. The physical properties panel includes the selection of the pol-  
238 lutant and the selection of the packing material. The selection of the pollutant uses  
239 a pop-up menu where it is possible to choose from among some predefined VOCs,  
240 whose information includes the diffusion coefficient in water ( $D_{P_w}$ ), the Henry's law  
241 constant ( $H_P$ ) at 25°C, and the chemical formula. Alternatively, the user can select  
242 a user defined pollutant by specifying its diffusion coefficient in water, its Henry's  
243 law constant, and its chemical formula. The selection of the packing material uses  
244 another pop-up menu in which there are some predefined packing materials for each  
245 of which there are provided its specific surface area ( $A$ ), porosity ( $\theta_{pm}$ ), and specific  
246 coefficients to calculate the overall mass transfer coefficients ( $K_{LaP}$ ,  $K_{LaO}$ ) using

247 the correlations proposed by San-Valero et al. (2014). Alternatively, it is possible to  
248 define other packing material by specifying its specific surface area, porosity, and  
249 overall mass transfer coefficients.

250 ● Hydrodynamic conditions such as liquid flow rate and fraction occupied by the liq-  
251 uid film.

252 ● Biofilm properties. In this panel there should be indicated its biomass density ( $X_V$ ),  
253 the thickness of its biofilm ( $\delta$ ), and its fraction occupied by the biofilm ( $\theta_B$ ).

254 ● Kinetics data. In this panel the user indicates the kinetical parameters regarding  
255 to the pollutant degradation ( $\mu_{max}$ ,  $K_S$  and  $Y_P$ ). Regarding the oxygen parameters,  
256  $K_O$  has been predefined from the literature as  $0.26 \text{ g m}^{-3}$  and  $Y_O$  is calculated by  
257 stoichiometry balance.

258 ● Advanced options. This button opens a dialog where other properties related with  
259 the mass transfer can be defined.

260 After all the input data and parameters have been defined, the simulation is run by pressing  
261 the Start button. When it concludes, the results are presented to the user in a new window,  
262 shown in Figure 6. The main items are described next.

263 [Figure 6 about here.]

264 • A graph showing both the inlet and the outlet VOC concentrations in the gas phase  
265 (details about the information plotted in this graph will be given in Section 5).

266 • A graph showing the evolution of the VOC concentration in the liquid tank (details  
267 about the information plotted in this graph will be given in Section 5).

268 • Some relevant averages, over the whole simulation time, are displayed by this panel:

269 - Inlet/Outlet VOC concentration,

270 - Inlet load (IL) defined as

$$IL\left(\frac{g-C}{m^3h^1}\right) = \frac{\overline{C_G^{in}}\overline{Q_G}}{V_R 3600} \quad (22)$$

271 where  $\overline{C_G^{in}}$  is the average inlet concentration and  $\overline{Q_G}$  is the average of the gas  
272 flow rate.

273 - Removal efficiency (RE):

$$RE(\%) = \frac{\overline{C_G^{in}} - \overline{C_G^{out}}}{\overline{C_G^{in}}} 100 \quad (23)$$

274 where  $\overline{C_G^{out}}$  is the average outlet concentration

275 - Elimination Capacity (EC):

$$EC\left(\frac{g-C}{m^3h^1}\right) = \frac{RE}{100} IL \quad (24)$$

- 276 • Panel with averages for the emission pattern 1 only.
- 277 • Panel with averages for the emission pattern 2 only.
- 278 • This button allows exporting the simulation results to a *Comma-Separated Values*  
279 (CSV) file.
- 280 • Closes the results window.

## 281 **Model Calibration and Validation**

282 The model was calibrated and validated by using the experimental data corresponding  
283 to the dynamic response of a biotrickling filter treating isopropanol obtained by San-Valero  
284 et al. (2013). In this data, the BTF was operated under intermittent loading conditions  
285 and intermittent spraying frequency. These ones are typically found in the operation of  
286 industrial BTFs. During these experiments it was observed that the discontinuous regime  
287 of spraying of the bed resulted in outlet emissions of isopropanol during spraying periods.  
288 Based on this observation, the effect of the spraying pattern was evaluated and it was  
289 pointed out that the spraying frequency is a critical parameter to achieve low emissions.  
290 The BTF was operated by using an IL of  $32 \text{ g-Cm}^{-3}\text{h}^{-1}$  and empty bed residence time  
291 (EBRT) of 30 s. The EBRT is defined as:

$$EBRT(s) = \frac{V_R}{Q_G} \quad (25)$$

292 These VOC feeding conditions were applied for a total time of 57600 s (16 h) from  
293 6:00 to 22:00h. The rest of the day, the biotrickling filter remained without VOC supply  
294 and without spraying. The parameters used in the modelling of the BTF behaviour are  
295 summarized in Table 1. The experimental parameters were taken from the literature or  
296 experimentally determined. The calibrated parameters were determined to fit the transient  
297 response data of the biotrickling filter. An independent experiment with a spraying pattern  
298 of 15 min every 1.5 h was used in the calibration step. Thus, time durations of 900 and  
299 4500 s for the spraying and non-spraying periods, respectively, were set. In this experi-  
300 ment, it was assumed no mass transfer resistance at the gas-liquid interface ( $\alpha_2=100$ ). The  
301 comparison of experimental results and model predictions are shown in Figure 7. Figure  
302 7(a) displays the evolution of the inlet and outlet VOC concentrations while Figure 7(b)  
303 displays the evolution of the concentration of carbon dissolved in the water tank.

304 [Table 1 about here.]

305 [Figure 7 about here.]

306 As it is shown in Figure 7(a), maximum concentrations of the pollutant are reached during  
307 the spraying periods, whereas during the non-spraying periods, nearly complete biodegra-  
308 dation of the pollutant is obtained. In addition, the peaks increase as the system gets filled  
309 with pollutant, reaching a stationary value for an outlet VOC concentration of around 0.2

310  $\text{g-Cm}^{-3}$  after the third cycle. An EC of  $27.2 \text{ g-Cm}^{-3}\text{h}^{-1}$  is obtained for an IL of 32  
311  $\text{g-Cm}^{-3}\text{h}^{-1}$  during VOC feeding periods. The model successfully predicts the behaviour  
312 obtained, achieving maximum outlet concentrations during spraying periods at the last cy-  
313 cles of the day. The experimental data fits with the model prediction with a relative error  
314 less 3 % in the EC (EC of the model  $28.0 \text{ g-Cm}^{-3}\text{h}^{-1}$ ). Also, the model prediction for  
315 the carbon dissolved in the water tank is in good agreement with the measured carbon in  
316 the water tank.

317 The validation of the model was carried out by using data from two experiments. The  
318 first experiment was carried out with low spraying frequency of 15 min every 3h and mod-  
319 erate IL= $32 \text{ g-Cm}^{-3}\text{h}^{-1}$ . The second experiment was carried out with double spraying  
320 frequency (15 min every 1.5h) and double IL ( $65 \text{ g-Cm}^{-3}\text{h}^{-1}$ ). The experimental data  
321 and the model prediction are shown in Figure 8. Figure 8(a) displays the evolution of the  
322 inlet and outlet VOC concentrations for the first experiment while Figure 8(b) displays the  
323 evolution of the inlet and outlet VOC concentrations for the second experiment.

324 [Figure 8 about here.]

325 For the experiments carried out with a spraying regime of 15 min every 3 hours and  
326 IL of  $32 \text{ g-Cm}^{-3}\text{h}^{-1}$ , the relative error between experimental and simulated EC is 3.2 %  
327 (experimental EC of  $28.8 \text{ g-Cm}^{-3}\text{h}^{-1}$  and modelled EC of  $29.7 \text{ g-Cm}^{-3}\text{h}^{-1}$ ). For the  
328 experiments carried out with a spraying regime of 15 min every 1.5 hours and an IL of 65

329  $\text{g-Cm}^{-3}\text{h}^{-1}$ , the error between the experimental and simulated EC is 4.0% (experimental  
330 EC of  $50.3 \text{ g-Cm}^{-3}\text{h}^{-1}$  and modelled EC of  $52.3 \text{ g-Cm}^{-3}\text{h}^{-1}$ ). The concentration of  
331 the dissolved carbon in the tank is in agreement with the measured values. As example,  
332 for the serie with a spraying regime of 15 min every 3 hours and IL of  $32 \text{ g-Cm}^{-3}\text{h}^{-1}$ ,  
333 the measured dissolved carbon was  $357 \text{ g-Cm}^{-3}$  and the model predicted a value of  $365$   
334  $\text{g-Cm}^{-3}$ , with a relative error of 2.2 %.

335 So, the model has been proven suitable in describing the complex phenomena observed  
336 in the transient response of the biotrickling filter to variations of the spraying pattern.

### 337 **Study of the dynamic response of the BTF to variable inlet concentrations and gas** 338 **flow rates**

#### 339 *Effect in the dynamic response of the BTF to oscillating inlet concentration*

340 The effect in the dynamic response of the BTF to oscillating inlet VOC concentration  
341 is investigated by using a periodic pulse train concentration pattern. The pulse train profile  
342 is used here to study the influence in the performance of high shock loads during regular  
343 changes in the operation. In particular, the selected inlet concentration takes on two alter-  
344 nating values:  $C_{Gp}^{in} = 0.7 \text{ g-Cm}^{-3}$  (for 7200 s) and  $C_{Gp}^{in} = 0.2 \text{ g-Cm}^{-3}$  (for 3600 s). A  
345 linear transition with a duration of 15 minutes is used to connect the two different values.  
346 A constant EBRT of 60 s is applied. Also, durations of 0.25 and 1 hours are specified  
347 for the spraying and non-spraying periods, respectively, and the pattern is applied for  $T$

348 = 59400 s. The simulation results are presented in Figure 9(a) for the gas phase and in  
349 Figure 9(b) for the liquid phase. Figure 9(a) shows that the concentration peaks not only  
350 depend on the spraying cycles but also on the pattern of the inlet concentration. An EC  
351 of  $30 \text{ g-Cm}^{-3}\text{h}^{-1}$  is obtained for an IL of  $32 \text{ g-Cm}^{-3}\text{h}^{-1}$ . The evolution of the VOC  
352 in the tank is presented in Figure 9(b). To observe the accumulation of dissolved carbon  
353 in the water tank, in this example the concentration of dissolved carbon in the tank was  
354 set to  $0 \text{ g-Cm}^{-3}$ . In this example, two different phenomena can be observed: absorption  
355 and desorption processes. These processes are markedly dependent on the equilibrium  
356 between the gas and liquid phases. As can be observed, when the inlet concentration in-  
357 creases during the spraying periods, a desorption of pollutant from the liquid phase to the  
358 gas phase is produced, and the opposite occurs when the inlet concentration increases. At  
359 the end of the period, the water contains  $200 \text{ g-Cm}^{-3}$  of dissolved carbon.

360 [Figure 9 about here.]

361 *Effect in the dynamic response of the BTF to oscillating inlet concentration combined with*  
362 *spraying times during non-VOC feeding periods*

363 [Figure 10 about here.]

364 The effect in the dynamic response of the BTF to oscillating inlet concentration com-  
365 bined with spraying times during non-VOC feeding periods is investigated. An oscillating



366 emission pattern has been applied for a total of 59400 s per day. The inlet VOC concen-  
367 tration is exactly as the pulse train profile used in the previous example. A period without  
368 VOC feeding has been applied for 27000 s with a Ramp+Constant profile of  $C_{Gp} = 0.01$   
369  $\text{g-Cm}^{-3}$  and a spraying time of 1 hour every 4 h. The results for the gas and liquid  
370 phases, respectively, are shown in Figures 10(a) and 10(b). The combination of differ-  
371 ent input profiles leads to some remarkable observations of the behaviour of the system.  
372 Namely, the presence of dissolved VOCs in the water recirculation tank, combined with  
373 the spraying cycles during the shut-off periods, produces peaks of pollutant even in the  
374 absence of VOCs in the inlet stream. Also desorption is present during these periods. The  
375 decrease of these peaks during the shut-off periods are related to the transfer of VOCs to  
376 the column, where they get degraded.

377 *Effect in the dynamic response of the BTF to oscillating gas flow rates*

378 The effect of the gas flow rate on the BTF is carried out. A constant concentration of  
379  $C_{Gp} = 0.53 \text{ g-Cm}^{-3}$  is selected. The gas flow rate takes on two alternating values:  $4.8$   
380  $10^{-4} \text{ m}^3 \text{ s}^{-1}$  and  $6.8 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}$  applied each one for periods of 14400 s. Note that  
381 the average value of the EBRT is 60 seconds, as in the previously considered examples.  
382 The simulation results are shown in Figure 11. Figure 11(a) displays the evolution of the  
383 inlet and outlet VOC concentrations in the gas phase, Figure 11(b) displays the evolution  
384 of the concentration of carbon dissolved in the water tank, and Figure 11(b) represents

385 the oscillating EBRT pattern. From Figure 11(a), the evolution of the peaks of the outlet  
386 gas concentration are different than those obtained in the previous examples. The gas  
387 velocity is directly related to the mass transfer of the pollutant between the gas and liquid  
388 phases, obtaining a greater mass transfer at large gas velocities, and thus, smaller EBRTs.  
389 The peaks obtained at the outlet VOC concentration pattern do oscillate according to the  
390 oscillating EBRT pattern. This contrasts with Figure 7(a), where the peaks increase until  
391 reaching the stationary state. These VOC emissions are related to an increase of the IL  
392 generated by an increase in the gas velocity and thus a decrease in the EBRT. As for the  
393 liquid phase, in Figure 11(b) it is possible to observe the influence of the gas velocity and  
394 EBRT on the absorption and desorption processes. In this situation, the increase in the  
395 amount of carbon dissolved in the water tank is combined with the desorption processes,  
396 producing oscillations as in the case of the outlet concentration.

397 [Figure 11 about here.]

## 398 **Conclusions**

399 Industrial biotrickling filters (BTFs) usually employ alternating spraying and non-  
400 spraying periods. A software tool to simulate the behaviour of BTFs under this and other  
401 typical conditions found in industrial facilities has been presented. The partial differential  
402 equations of the BTF model have been solved numerically using the method of lines. In

403 particular, the software also allows simulating the treatment of volatile organic compound  
404 (VOC) air emissions under variable inlet concentrations and gas velocities. The model was  
405 calibrated and validated by using data from a biotrickling filter treating isopropanol under  
406 intermittent conditions of loading and spraying. The capability of the model to reproduce  
407 the complex phenomena involved in the dynamic response of the treatment of hydrophilic  
408 compounds by biotrickling filters have been proven. Several examples demonstrate that  
409 the pattern of the outlet emissions depends on the pattern of the gas velocity and inlet  
410 concentration, showing the utility of the tool to assist in the design and operation of BTFs.  
411 The software tool presented herein will be a basis for implement new features. For exam-  
412 ple, it would be interesting to allow multi-component mixtures in order to go deeper into  
413 the interaction between pollutants. This and other extensions are left for future research.

#### 414 **Nomenclature**

415 [Table 2 about here.]

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514 322.

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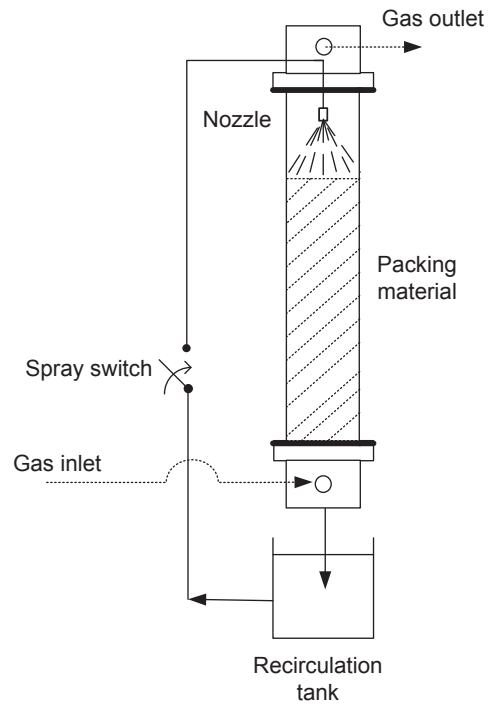


Figure 1: Diagram of a BTF. Liquid recirculation only happens during spraying periods.

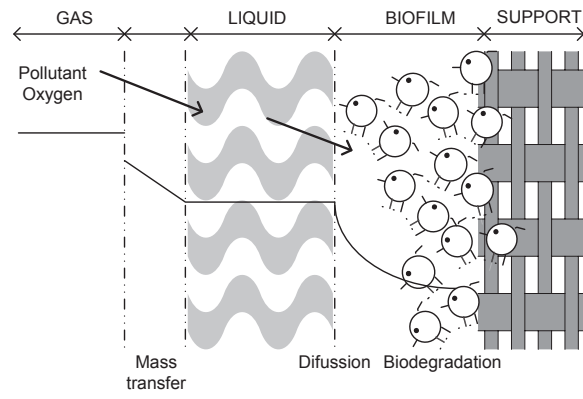


Figure 2: Mechanisms involved in the process of BTF

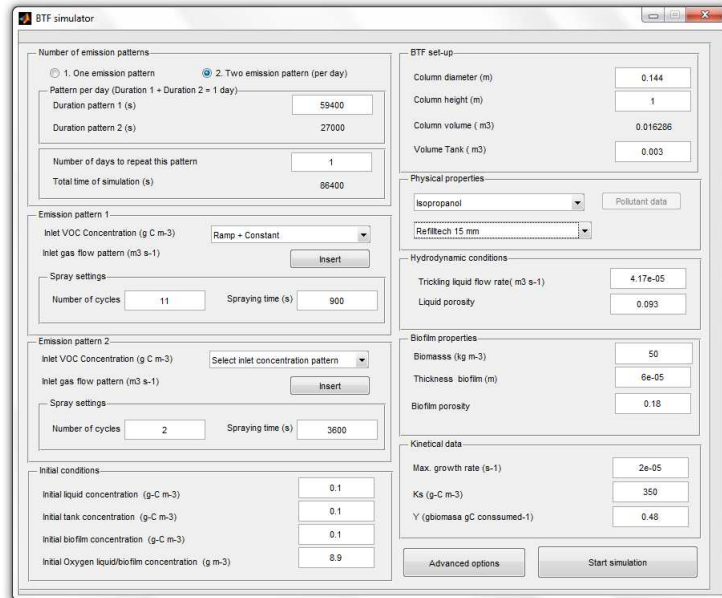


Figure 3: Main window of the GUI: Two emission patterns (per day)

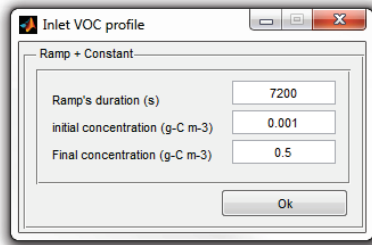


Figure 4: GUI of the MATLAB<sup>®</sup> tool. Dialog for the Ramp+Constant inlet VOC concentration profile.

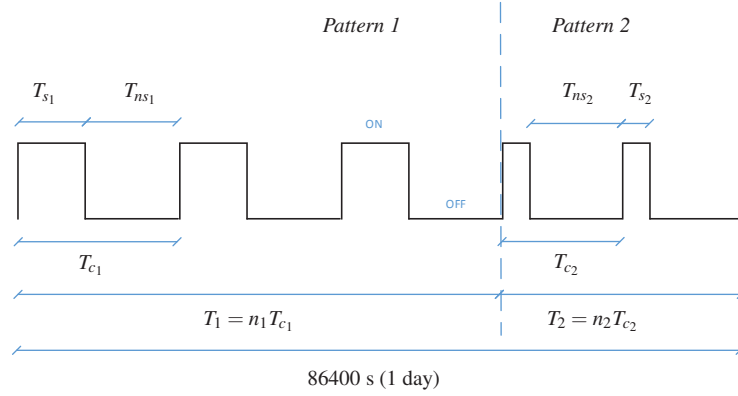


Figure 5: Spray cycle patterns for one day. Emission pattern 1 has three cycles ( $n_1=3$ ), and emission pattern 2 has two ( $n_2=2$ ) spray cycles. The user specifies  $T_{s1}$ ,  $n_1$ ,  $T_1$ ,  $T_{s2}$ , and  $n_2$ . *Non-spray times* are given by  $T_{ns1} = (T_1 - n_1 T_{s1})/n_1$  and  $T_{ns2} = (T_2 - n_2 T_{s2})/n_2$  for emission pattern 1 and emission pattern 2, respectively.

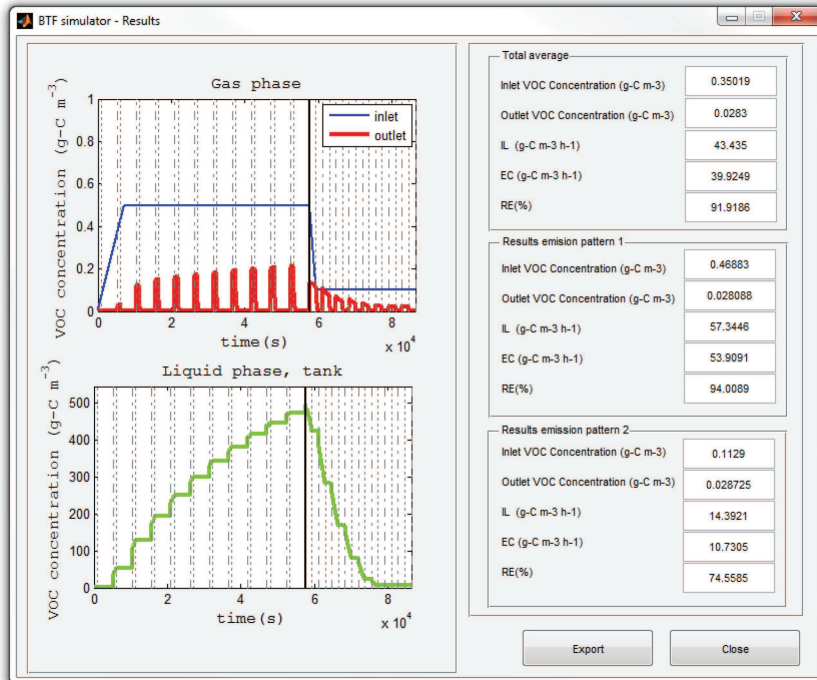
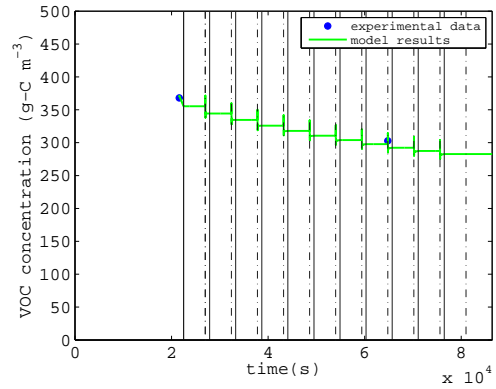
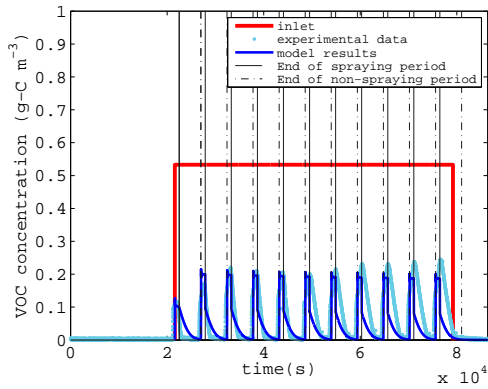


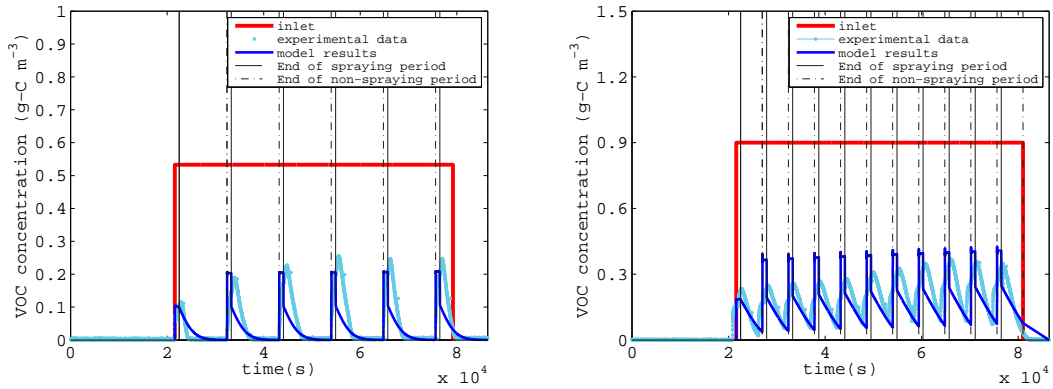
Figure 6: GUI of the MATLAB<sup>®</sup> tool (results window).





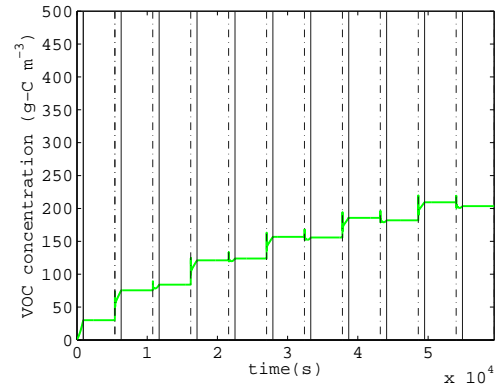
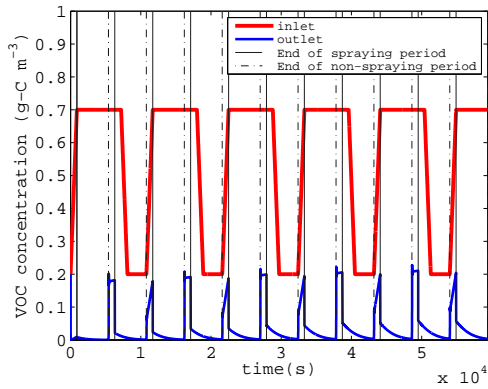
(a) Evolution of the concentration in the gas phase (b) Evolution of the dissolved organic carbon in the recirculation tank

Figure 7: Model Calibration with experimental data from San-Valero et al. (2013)



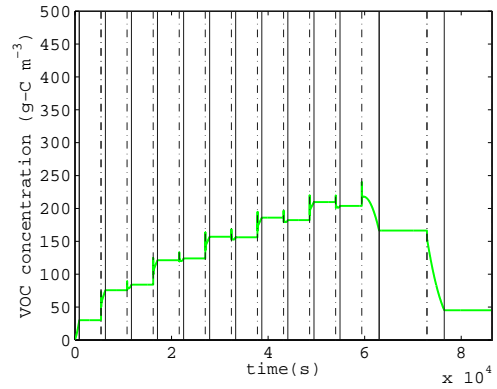
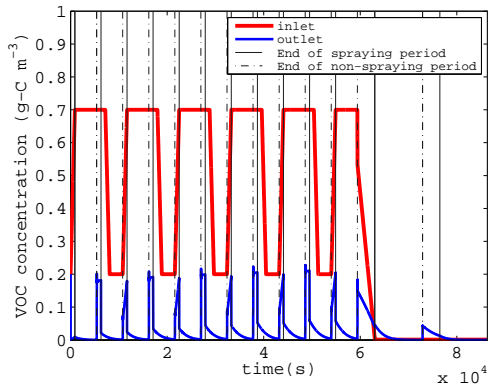
(a) Spraying regime 15 min every 3h and IL 32 g-C m<sup>-3</sup> h<sup>-1</sup> (b) Spraying regime 15 min every 1.5 h and IL 65 g-C m<sup>-3</sup> h<sup>-1</sup>

Figure 8: Model Validation with experimental data from San-Valero et al. (2013)



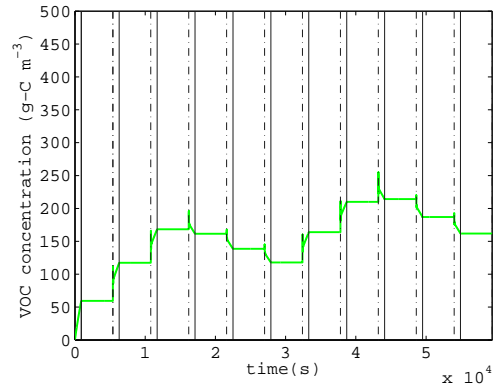
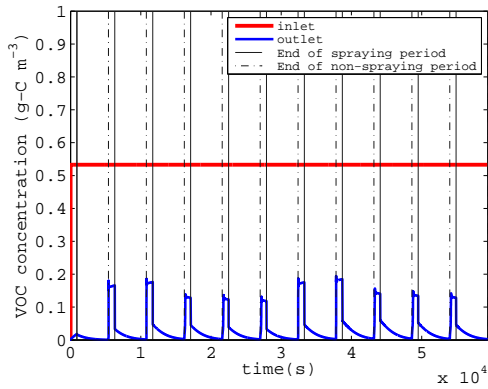
(a) Evolution of the concentration in the gas phase (b) Evolution of the dissolved organic carbon in the recirculation tank

Figure 9: Effect in the dynamic response of the BTF to oscillating inlet concentration

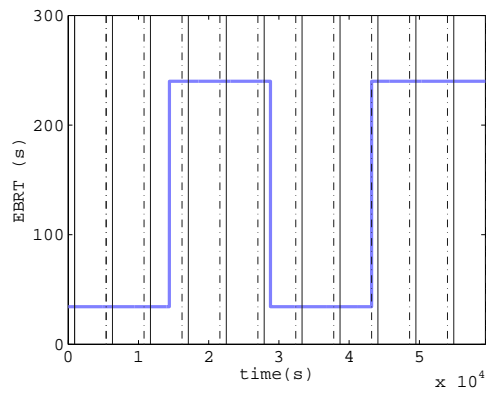


(a) Evolution of the concentration in the gas phase (b) Evolution of the dissolved organic carbon in the recirculation tank

Figure 10: Effect in the dynamic response of the BTF to oscillating inlet concentration combined with spraying times during non-VOC feeding periods



(a) Evolution of the concentration in the gas phase (b) Evolution of the dissolved organic carbon in the recirculation tank



(c) Evolution of the EBRT

Figure 11: Effect in the dynamic response of the BTF to oscillating gas flow rates

534 **List of Tables**

535     1     Model parameters used in the mathematical model . . . . . 46

Variable	Specific Value	Units	Reference
<b>Experimental parameters</b>			
$A_v$	348	$m^{-1}$	San-Valero et al. (2013)
$D$	0.144	m	San-Valero et al. (2013)
$D_{P_w}$	$1.13 \times 10^{-9}$	$m^2 s^{-1}$	Tucker and Nelken (1982)
$D_{O_w}$	$2 \times 10^{-9}$	$m^2 s^{-1}$	Reid et al. (1987)
$H_P$	$2.8 \times 10^{-4}$		San-Valero et al. (2014)
$H_O$	31.4		Sander (2005)
$K_{LAP}$	$\frac{H_P}{3600} \left( 11.59 (v_G 3600)^{0.85} \right)$	$s^{-1}$	San-Valero et al. (2014)
$K_{La0}$	$1.15 \times 10^{-2}$	$s^{-1}$	San-Valero et al. (2014)
$Q_L$	$41.7 \times 10^{-6}$	$m^3 s^{-1}$	San-Valero et al. (2013)
$V_R$	0.0163	$m^3$	San-Valero et al. (2013)
$V_T$	0.003	$m^3$	San-Valero et al. (2013)
$Y_P$	0.48	$\frac{\text{g biomass}}{\text{g consumed}}$	Lu et al. (2004)
$Y_O$	0.14	$\frac{\text{g biomass}}{\text{g consumed}}$	Stoichiometric balance
$Z$	1	m	San-Valero et al. (2013)
$\theta_B$	0.18		This work
$\theta_L$	0.093		This work
<b>Calibration parameters</b>			
$K_{sP}$	350	$g-C m^{-3}$	
$X_v$	$50 \times 10^3$	$g m^{-3}$	
$\alpha_1$	0.23 (except for cycle 1 that takes $\alpha_1 = 1$ )		
$\beta$	$6.4 \times 10^{-6}$	m	
$\delta$	$60 \times 10^{-6}$	m	
$\mu_{max}$	$2 \times 10^{-5}$	$s^{-1}$	

Table 1: Model parameters used in the mathematical model

Nomenclature	
$A$	specific surface area of the packing material ( $\text{m}^{-1}$ )
$C$	concentration ( $\text{g m}^{-3}$ )
$D$	diffusion coefficient of substrates ( $\text{m}^2 \text{s}^{-1}$ )
$f(X_v)$	correction factor of diffusivity in biofilm according to Equation 13
$H$	Henry constant of the substrates
$K_s$	half saturation rate constants of substrate ( $\text{g-C m}^{-3}$ )
$K_{La}$	overall mass transfer coefficients of the substrates ( $\text{s}^{-1}$ )
$M$	number of divisions along the column
$N$	number of divisions along the biofilm
$Q$	flow rate ( $\text{m}^3 \text{s}^{-1}$ )
$S$	concentration in the biofilm ( $\text{g m}^{-3}$ )
$t$	time (s)
$v$	superficial velocity ( $\text{m s}^{-1}$ )
$V$	volume ( $\text{m}^3$ )
$x$	coordinate for the depth in the biofilm, perpendicular to the biofilm surface
$X_v$	biomass concentration in the biofilm ( $\text{g m}^{-3}$ )
$Y$	yield coefficient (g of dry biomass synthesized per g consumed)
$z$	axial coordinate in the reactor
$Z$	height of the reactor (m)
$C_{G_P}^{in}$	inlet VOC concentration ( $\text{g-C m}^{-3}$ )
$C_{G_O}^{in}$	inlet oxygen concentration ( $\text{g m}^{-3}$ )
Greek letters	
$\delta$	active biofilm thickness (m)
$\theta_B$	fraction occupied by the biofilm
$\theta_G$	porosity of the bioreactor
$\theta_L$	fraction occupied by the liquid film
$\theta_{pm}$	void fraction of the packing material
$\mu_{max}$	maximum specific growth rate of the substratum ( $\text{s}^{-1}$ )
Subscripts	
$G$	gas
$L$	liquid
$B$	biofilm
$P$	pollutant
$O$	oxygen
$R$	reactor
$T$	tank
$w$	water