

## CO2 mass transfer and conversion to biomass in a horizontal gas–liquid photobioreactor

P. Valiorgue, H. Ben Hadid, M. El Hajem, L. Rimbaud, A. Muller-Feuga, J.Y.

Champagne

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Author: P. Valiorgue H. Ben Hadid M. El Hajem L. Rimbaud A. Muller-Feuga J.Y. Champagne

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#### **Research Highlights**

The  $CO_2$  conversion to biomass efficiency is modelled.

The influence of gas stripping is shown to be not negligible.

Effects of design and operating parameters are investigated.

# CO<sub>2</sub> mass transfer and conversion to biomass in a horizontal gas-liquid photobioreactor

P. Valiorgue<sup>a,\*</sup>, H. Ben Hadid<sup>a</sup>, M. El Hajem<sup>a</sup>, L. Rimbaud<sup>b</sup>,

A. Muller-Feuga<sup>b</sup>, J.Y. Champagne<sup>a</sup>

<sup>a</sup>LMFA, UMR CNRS 5509, Université de Lyon, Ecole Centrale de Lyon,
 Université Lyon 1, INSA de Lyon, ECL, 20, avenue Albert Einstein - 69621
 Villeurbanne Cedex - tel: +33472436436, fax: +33472438718.

<sup>b</sup>Microphyt, 713, Route de Mudaison 34670 Baillargues – France, http://www.microphyt.eu/, Cell: +33 6 14 79 68 92

#### Abstract

This study deals with  $CO_2$  mass transfers and biomass conversion in an industrial horizontal tubular photobioreactor. An analytical approach is used to determine an expression modeling the influence of  $CO_2$  mass transfers on the overall biomass conversion efficiency for a given culture broth, heat and light conditions. Fluid mechanics and mass transfer are predicted with a classical two-phase flow approach [1] combined with a dissolution correlation developed and tested in the laboratory [2]. The influence of the stripping gas, removing the excess of oxygen in the liquid, on the conversion to biomass efficiency is shown to be not negligible. The expression is used to evaluate how the photobioreactor's design and process parameters can be tuned in order to improve biomass conversion efficiency. The biomass conversion efficiency evolution with the photobioreactor's length was found to behave asymptotically and it was explained by the relative orders of magnitude of gas dis-

solution and gas stripping. It has been shown that the gas flow rate for stripping and therefore the oxygen removal will be limited when further increasing the industrial photobioreactor's length for a given objective of  $CO_2$  conversion to biomass efficiency.

*Key words:* mass transfer, gas-liquid, CO2 biomass conversion, photobioreactor, microalgae.

#### Nomenclature

#### Greek symbols

η	Mass Transfer Efficiency (MTE),
ν	Kinematic viscosity, $\left[\frac{m^2}{s}\right]$
Φ	Pipe diameter, $[m]$
$\Phi_h$	Hydraulic diameter of the duct, $[m]$
$\phi$	Molecular gas flux, $\left[\frac{mol}{s \times m^2}\right]$
ρ	Density, $\left[\frac{kg}{m^3}\right]$

#### Latin symbols

A Section area,  $[m^2]$ 

\* Corresponding author. Email address: pierre.valiorgue@gmail.com (P. Valiorgue).

С	Constant in the friction factor correlation
C	Carbon dioxide molar concentration, $\left[\frac{mol}{m^3}\right]$
D	Diffusion constant, $\left[\frac{m^2}{s}\right]$
g	Gravity, $\left[\frac{m}{s^2}\right]$
h	Height, [m]
k	Mass transfer coefficient, $\left[\frac{m}{s}\right]$ ,
$k_{La}$	Volumetric mass transfer coefficient, $\left[\frac{1}{s}\right]$ ,
$L_{tubes}$	Total tube length of the photobioreactor, $[m]$ ,
М	Molar mass, $\left[\frac{kg}{mol}\right]$
m	Carbon dioxide mass, $[kg]$
p	Pressure, $[Pa]$
R	Ideal gas constant, $\left[\frac{J}{mol.K}\right]$
Re	Reynolds number, $Re = \frac{U \cdot \Phi_h}{\nu}$
S	Perimeter, $[m]$
Sc	Schmidt number, $Sc = \frac{\nu}{D}$
Sh	Sherwood number, $Sh = \frac{k_L \cdot \Phi_h}{D}$
Т	Temperature, $[K]$

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t	Time, $[s]$	
U	Mean velocity over the duct section area, $\left[\frac{m}{s}\right]$	
v	Exponent in equation (8), see Taitel in [1]	
w	Exponent in equation (8), see Taitel in [1]	
x	Coordinate in the downstream direction along the duct, $\left[m\right]$	
X	Lockhart and Martinelli parameter.	
Subscripts and superscripts		

## Subscripts and superscripts

biomassconv	$CO_2$ contained in the output dry microalgae,
diss	$CO_2$ dissolved,
eq	Equivalent height,
G	Gas,
i	Interface,
injected	$CO_2$ injected into the photobioreactor,
L	Liquid,
undiss	$\mathrm{CO}_2$ undissolved and directly rejected to the atmosphere,
prod	Production of the microalgae mass harvested,
residual	CO <sub>2</sub> dissolved in the liquid and remaining dissolved,

S Superficial, for single fluid flow,

sat Saturation,

stripped CO<sub>2</sub> driven to the atmosphere by the stripping air,

Dimensionless variable.

#### 1 1 Introduction

Microalgae cultures have been recently regarded as a means to bioremediate 2 industrial  $CO_2$  waste. However, the efficiency of the conversion of the  $CO_2$ 3 into biomass has been seldom documented, and a significant part of the gas introduced in microalgae production systems is suspected of being released in 5 the atmosphere. Improving the modelling of such biotechnological processes 6 will help increasing the biomass conversion efficiency of industrial waste gas 7 containing 10 to 20% of CO<sub>2</sub> [3–5]. High-density photoautotrophic microalgal 8 growth in enclosed photobioreactors necessitates gas, light, momentum and 9 heat exchanges [3,6]. Mass transfer modelling is a first step toward under-10 standing the coupled physics-biology in the photobioreactor and improving 11 the  $CO_2$  conversion to biomass. 12

This study deals with the experimental assessment and modelling of  $CO_2$ biomass conversion in a horizontal co-current gas-liquid photobioreactor converting  $CO_2$  into value-added microalgae. The windy, wavy and wiped tubular photobioreactor investigated has been designed for slow growing and fragile species with the ambition of improving accessibility to the huge biodiversity

of microalgae. For the purpose of reducing mechanical stress and avoiding cell 18 wall disruption, the pump device is operated under low pressure and bubbling 19 has been reduced by removing the direct airlift achieving mass transfer usu-20 ally placed in the culture loop [7,8]. Carbon dioxide and oxygen mass transfers 21 are achieved through co-current circulation of gas and liquid within the pho-22 tobioreactor along with photosynthesis. Carbon is provided to the cells via 23 punctual injections of gas that contains up to 10% of  $CO_2$  and are monitored 24 by a feedback loop maintaining a constant pH. Excess of oxygen inhibiting 25 microalgae growth is removed from the culture broth by continuously injected 26 stripping air [8,9]. 27

A few mass transfer models and measurements applied to photobioreactors can 28 be found in the literature of the two last decades. As highlighted in [10,11], CO<sub>2</sub> 29 biomass conversion efficiency depends on  $CO_2$  concentrations both in the air-30  $CO_2$  mixture injected in the photobioreactor and in the algal suspension [11]. 31 Therefore, mass transfer efficiencies (MTE) measurements should be reported 32 for a given operating injected gas conditions as done in [11], for an outdoor 33 culture of *Chlorella spirulina*. Very few data concerning the produced biomass 34 and the injected mass of  $CO_2$  are available in the literature as can be found 35 in [11]. 36

<sup>37</sup> Mass transfer models of photobioreactors reported in the literature enable to <sup>38</sup> determine overall mass transfer coefficients from correlations of the parame-<sup>39</sup> ters of the process [12–15]. To our knowledge, there is no analytical study in <sup>40</sup> the literature concerning  $CO_2$  mass transfer and biomass conversion in a hor-<sup>41</sup> izontal gas-liquid photobioreactor. Such an analytical study would determine <sup>42</sup> where the  $CO_2$  which has not been converted to biomass has been lost and <sup>43</sup> how process parameters can be tuned in order to improve biomass conversion

<sup>44</sup> efficiency for a given culture condition.

This paper is structured in three sections. In the first section, a CO<sub>2</sub> mass balance over an horizontal photobioreactor will allow to express mass transfer efficiencies as a function of operating parameters. Experimental measurements will then be explained in the second section and finally, results will be presented and discussed.

#### 50 2 Mass transfer modeling

#### 51 2.1 Conservation of mass

As depicted on figure (1,a), the carbon dioxide mass injected in the photo-52 bioreactor,  $m_{injected}$ , is partially dissolved into the liquid phase  $(m_{diss})$  and the 53 remaining part is directly rejected to the atmosphere, as written in equation 54 (1). The continuously injected air represented in figure (1,b) is also driving a 55 mass  $(m_{stripped})$  of carbon dioxide dissolved into the liquid to the atmosphere. 56 The carbon dioxide dissolved in the liquid phase will either be transformed into 57 biomass  $(m_{biomassconv})$  or remain as a dissolved form in the liquid  $(m_{residual})$ , 58 as described by the mass balance written in equation (2). 59

$$m_{injected} = m_{undiss} + m_{diss} \tag{1}$$

$$m_{diss} = m_{stripped} + m_{residual} + m_{biomassconv} \tag{2}$$

 $_{60}$  Dividing equation (2) by the carbon dioxide mass injected, equation (3), the

<sup>61</sup> photobioreactor's MTE is obtained. The photobioreactor's CO<sub>2</sub> conversion to <sup>62</sup> biomass MTE,  $\eta_{biomassconv}$ , is defined as the ratio of the carbon dioxide mass <sup>63</sup> contained in the output microalgae dry mass ( $m_{biomassconv}$ ) over the carbon <sup>64</sup> dioxide mass injected ( $m_{injected}$ ). Similarly,  $\eta_{diss}$ ,  $\eta_{stripped}$  and  $\eta_{residual}$  are de-<sup>65</sup> fined as the ratios of respectively  $m_{diss}$ ,  $m_{stripped}$  and  $m_{residual}$  over  $m_{injected}$ .

# $\eta_{biomassconv} = \eta_{diss} - \eta_{stripped} - \eta_{residual} \tag{3}$

In the next sections, dissolution and stripping phases are modelled as functions
 of process parameters .

#### 68 2.2 Model for $\eta_{diss}$

As described in [7,8], the flow regime in the photobioreactor is mainly constituted of long slugs that are more or less wavy. From [2], a model for dissolution efficiency ( $\eta_{diss}$ ) of an elongated gas bubble into a turbulent flow has been developed and is expressed in equation (4).

$$\eta_{diss} = 1 - e^{-k_L(Re) \times (C_{sat} - C) \times \frac{R \times T}{p \times h_G} \times t}$$
(4)

<sup>73</sup> In this model, the interface is considered as a flat plane and the equivalent <sup>74</sup> bubble height  $h_G$ , is defined as the ratio of the cross section area occupied by <sup>75</sup> the gas,  $A_G$ , over the interfacial contact width  $S_i$ , see figure (2).

The contact time t between the bubble and the liquid is estimated from the gas section area  $A_G$ , the photobioreactor's total length  $L_{tubes}$  and the gas flow rate  $Q_G$ , as expressed in equation (5). Replacing  $h_G$  and contact time into equation

<sup>79</sup> (4) leads to the expression of  $\eta_{diss}$  as a function of operating parameters in <sup>80</sup> equation (6).

$$t = \frac{A_G \times L_{tubes}}{Q_G}$$

$$\eta_{diss} = 1 - e^{-k_L(Re) \times (C_{sat} - C) \times \frac{R \times T}{p} \times \frac{S_i}{Q_G} \times L_{tubes}}$$
(5)
(6)

Estimations of the mass transfer coefficient and of the equivalent bubble height are required in order to evaluate  $\eta_{diss}$  from equation (6). The mass transfer coefficient  $k_L$ , has been evaluated using a correlation of the Sherwood number  $Sh = \frac{k_L \cdot \Phi_h}{D}$ , the Schmidt number,  $Sc = \frac{\nu}{D}$ , and the Reynolds number,  $Re = \frac{U \cdot \Phi_h}{\nu}$ , proposed in Lamourelle [16] and expressed in equation (7) which has been tested successfully for an immobilised elongated bubble in [2].

$$Sh_L = 1.76 \times 10^{-5} \times Re^{1.506} \times Sc^{0.5}$$
 (7)

In order to calculate the hydraulic diameter  $\Phi_h$ , the equivalent bubble height,  $h_G$ , was estimated by solving the momentum balance equation (8) for horizontal stratified flow. In this dimensionless equation, X is the Lockhart and Martinelli's parameter defined in equation (9). The other dimensionless parameters in equation (8) are explicitly defined in [17] as a function of the dimensionless liquid height,  $\tilde{h}_L$ .

$$X^{2}\left[ (\tilde{U}_{L}\tilde{\Phi}_{L})^{-v} \tilde{U}_{L}^{2} \frac{\tilde{S}_{L}}{\tilde{A}_{L}} \right] - \left[ (\tilde{U}_{G} \tilde{\Phi}_{G})^{-w} \tilde{U}_{G}^{2} \left( \frac{\tilde{S}_{G}}{\tilde{A}_{G}} + \frac{\tilde{S}_{i}}{\tilde{A}_{L}} + \frac{\tilde{S}_{i}}{\tilde{A}_{G}} \right) \right] = 0 \qquad (8)$$

$$X^{2} = \frac{\left| \left(\frac{dp}{dx}\right)_{S,L} \right|}{\left| \left(\frac{dp}{dx}\right)_{S,G} \right|} = \frac{\frac{4c_{L}}{\Phi} \left(\frac{U_{S,L}\Phi}{\nu_{L}}\right)^{-\nu} \frac{\rho_{L}(U_{S,L})^{2}}{2}}{\frac{4c_{G}}{\Phi} \left(\frac{U_{S,G}\Phi}{\nu_{G}}\right)^{-w} \frac{\rho_{G}(U_{S,G})^{2}}{2}}$$
(9)

A relationship between liquid flow rate, gas flow rate and liquid height can be expressed replacing equation (9) into equation (8). The obtained equation is solved for  $\tilde{h}_L$  using dichotomy knowing the gas and liquid velocities for the guessed liquid height.

A Scilab algorithm calculates the mass transfer coefficient and the mass transfer efficiency for each rectilinear section of the photobioreactor using respectively Lamourelle's correlation (7) and equation (6).

## 100 2.3 Model for $\eta_{stripped}$

As the stripping air is injected continuously, the interface is ditributed along the entire length of the photobioreactor. The interface for the gas-liquid flow is approximated to  $S_i \times L_{tubes}$ . The interface width  $S_i$  is considered as a constant along the photobioreactor's length since the mass transferred is small compared to the mass injected.

As noted by Boettcher in [18], the molecular gas flux through a gas-liquid interface can be expressed as a function of the local molar concentration Cand the molar concentration at saturation of the stripping gas  $C_{sat \ stripped}$  by equation (10).

$$\frac{dn_{stripped}}{dt} = -(S_i \times L_{tubes}) \times \phi = -k_L \times (S_i \times L_{tubes}) \times (C_{sat \ stripped} - C)$$
(10)

<sup>110</sup> Integrating equation (10) with respect to time, the stripping MTE can be <sup>111</sup> expressed as in equation (11).

$$\eta_{stripped} = -k_L \times (S_i \times L_{tubes}) \times (C_{sat \ stripped} - C) \times M_{CO2} \times \frac{t_{prod}}{m_{injected}}$$
(11)

In equation (11), the coupled effects of cell density, light, momentum or temperature influencing the culture growth are affecting the stripping phase and therefore the CO<sub>2</sub> conversion to biomass process. They are implicitly taken into account through  $t_{prod}$ , the production time.

In order to evaluate mass transfer efficiencies, mass transfer measurements
in an industrial photobioreactor have been realised and their acquisition is
described in the following section.

#### <sup>119</sup> 3 Measurements of Mass Transfer Efficiencies

During data acquisitions, the microalgae specie cultivated in the photobioreactor is chlorophyte *Neochloris oleoabundans*. The aim of the measurements in
the photobioreactor is to determine the orders of magnitudes for dissolution,
stripping and photobioreactor's MTEs.

#### 124 3.1 Measurement of $\eta_{diss}$

A Gastec  $CO_2$  dosage kit has been used for the measurements. These kits 125 are given for carbon dioxide concentrations from 0.5 to 20 % of the volume. 126 During data acquisition, injection flow rates of air and  $CO_2$  have been set 127 with an injected gas containing up to 10% of CO<sub>2</sub>. Measurements of CO<sub>2</sub> 128 concentration at the injection point and the exit of the photobioreactor show 129 a dissolution MTE,  $\eta_{diss}$ , of 72%. The coefficient of variation of 5% given 130 for this probe is considered as the relative error for each measurement. The 131 relative error on  $\eta_{diss}$  is then of 10%. 132

#### 133 3.2 Measurement of $\eta_{biomassconv}$

From its definition,  $\eta_{biomassconv}$  evaluation requires the measurement of the harvested microalgae's dry mass and of the consumed mass of CO<sub>2</sub> to produce it, considering that  $\eta_{residual}$  is considered to be negligible. The residual mass of dissolved CO<sub>2</sub> in the culture broth photobioreactor has been measured with a metler toledo inPro 5000 and is evaluated to be less than 20 mg/L.

During the photobioreactor's development phase, food grade carbon dioxide
gas bottles of 37.4 kg were used to feed microalgae. The injected mass of CO<sub>2</sub>
have been estimated from the gas bottles' weight.

From June 16<sup>th</sup> to July 7<sup>th</sup>, 18.54 kilograms of dry biomass were harvested. It should be noted that, from the photosynthesis' stoichiometry for *Neochloris oleoabundans* given in [19], 1.88 grams of CO<sub>2</sub> are converted to biomass when 1 gram of dry microalgae is produced. This coefficient is then applied in order

to find the equivalent gas mass of CO<sub>2</sub> contained into the produced mass ofmicroalgae.

As can be seen in figure (3), microalgae concentration evolution with time 148 was maintained in a small range over the 22 days harvesting period. The final 149 cell density was measured around  $1.6 \times 10^8$  cells per millilitre, higher than 150 the starting cell density around  $1 \times 10^8$  cells per millilitre. Given that on 151 average 1.7 g of dry biomass were harvested from a litre of culture broth for 152 an average cell density of  $1.3 \times 10^8$  cells per millilitre, the equivalent of 3.69 153 kg dry biomass have been produced without being harvested. Using Pruvost 154 [19], 34.85 kg of CO<sub>2</sub> were converted into the harvested dry biomass which has 155 been produced using 105.4 kg of  $CO_2$  gas. The minimum biomass conversion 156 efficiency is then estimated to 33% considering only the carbon fraction in 157 the harvested biomass. Considering that 3.69 kg of dry biomass that have 158 been estimated to remain in the photobioreactor without being harvested, the 159 maximum biomass conversion efficiency is estimated to 40%. 160

## 161 3.3 Evaluation of $\eta_{stripped}$

 $\eta_{stripped}$  is evaluated from equation (3). The error is the sum of errors on  $\eta_{diss}$ 162 and  $\eta_{biomassconv}$ . For the considered culture conditions, orders of magnitude 163 of  $\eta_{stripped}$  and  $\eta_{biomassconv}$  are similar. Considering that from equation (11), 164  $\eta_{stripped}$  is proportional to production time,  $t_{prod}$ , if the time of production 165 is reduced, the photobioreactor's MTE will increase by the same proportion. 166 As a consequence, research efforts on optimum light, momentum and heat 167 conditions for microalgae's growth rate will have a strong impact on the global 168 photobioreactor's  $CO_2$  conversion to biomass efficiency. 169

#### 170 4 Results and discussion

#### 171 4.1 Dissolution model

The dissolution model presented in section 2.2 is plotted against the dissolu-172 tion measurement on figure (4). Asterisk and plus marks in the figure represent 173 the abscissa at the end of a linear section of the photobioreactor along which 174 the liquid height,  $CO_2$  concentration, pressure and mass flow rates are consid-175 ered as constants. For the considered conditions, carbon dioxide is dissolved 176 regularly along the photobioreactor. A good agreement was found between 177 dissolution measurements on the photobioreactor and the dissolution model. 178 The order of magnitude of the influence of the injected gas carbon dioxide 179 concentration can also be evaluated from figure (4). 180

Another result from the dissolution model concerns optimum flow conditions 181 for dissolution. As expressed in equation (6), for a fixed photobioreactor's 182 length, dissolution is improved when the ratio  $\frac{k_L(Re) \times S_i}{Q_G}$  is as large as possible. 183 This ratio is a function of three variables  $Q_G$ ,  $Q_L$  and  $h_L$  that are related by the 184 momentum balance equation (8). For industrial parameters  $5 < Q_G \left[ \frac{Nl}{min} \right] <$ 185 100 and 20 <  $Q_L\left[\frac{l}{min}\right]$  < 450,  $\tilde{h}_L$  and  $S_i$  can be considered as constants 186 since the standard deviations are evaluated as respectively 6% and 8.5% of 187 the averaged values, for the considered flow rates. As a consequence, effects of 188  $Q_L$  and  $Q_G$  on  $\frac{k_L(Re) \times S_i}{Q_G}$  are decoupled. Practically, as can be seen on figure 189 (5) for the range of considered flow rates, dissolution will be improved by 190 increasing the liquid flow rate and therefore turbulence or by decreasing the 191 gas flow rates and thus enhancing contact time, as long as the flow rates are 192 compatible with culture conditions. 193

# 4.2 Influence of dissolution and stripping on the photobioreactor's MTE and optimum length

<sup>196</sup> Combining equations (3), (6) and (11), an expression of  $\eta_{biomassconv}$  as a func-<sup>197</sup> tion of the photobioreactor's length,  $L_{tubes}$ , is obtained. In order to assess the <sup>198</sup> interest of scaling, it is possible to derive an semi-empirical relation for the <sup>199</sup> optimum length of the photobioreactor considering an equivalent constant con-<sup>200</sup> centration gradient along the photobioreactor as expressed in equation (12).

$$L_{opti} = \frac{L_{tubes}}{1 - \eta_{diss}(L_{tubes})} \times ln \left( \frac{\eta_{diss}(L_{tubes}) - \eta_{biomassconv}(L_{tubes})}{ln\left(\frac{1}{1 - \eta_{diss}(L_{tubes})}\right)} \right)$$
(12)

For the considered conditions, the optimum length and relative influence of dissolution and stripping on scaling for the photobioreactor's MTE are plotted in figure (6). It appears that the photobioreactor's length is close to the optimum length for carbon dioxide  $CO_2$  conversion to biomass efficiency for the operated conditions.

#### 206 5 Conclusion

An analysis of mass transfer in a horizontal photobioreactor based on a mass balance has been presented. The influence of the  $CO_2$  dissolved in the liquid and remaining dissolved have been shown to be negligible compared to the injected  $CO_2$  mass during the measurements. Under such conditions, stripping effects have been determined using dissolution and conversion to biomass measurements. They were found to be not negligible. A good agreement was

found between dissolution measurements and results predicted by the theo-213 retical prediction from the dissolution model for elongated bubbles adapted 214 for two phase flow. The influence of injected CO<sub>2</sub> concentration and flow rates 215 on the dissolution efficiency as predicted by the dissolution model have been 216 emphasized. A numerical investigation of the dissolution model highlighted 217 the effects of gas and liquid flow rates on the dissolution efficiency. Effects of 218 dissolution and stripping on the asymptotic behaviour of biomass conversion 219 efficiency evolution with the photobioreactor's length for given culture con-220 ditions were identified. It is expected that stripping will have to be reduced 221 when further increasing the photobioreactor's length for a given  $CO_2$  conver-222 sion to biomass efficiency objective. Research efforts on optimum growth rate 223 conditions and growth rate with oxygen will contribute to diminish the lim-224 iting effects of stripping and improve the global photobioreactor's conversion 225 to biomass prediction and efficiency. 226

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#### 283 Captions for tables and figures

Fig. 1. Diagram of the carbon dioxide punctual dissolution phase (a) and continuous air stripping phase (b).

Fig. 2. Summary of the notations used in this article are defined to be congruent to Taitel [1].

Fig. 3. Cell density evolution with time and details for the measurement period.

Fig. 4. Dissolution MTE,  $\eta_{diss}$ , as a function of the total tube length of the photobioreactor,  $L_{tubes}$ , and injected gas concentration as predicted by the one-dimensional model presented in section 2.2. The liquid and gas flow rates used,  $Q_L$  and  $Q_G$ , were corresponding to the experimental measurement operating flow rates on Microphyt's photobioreactor, respectively 130 l/min and 35 N.l/min.

Fig. 5. The ratio  $\frac{k_L(Re) \times S_i}{Q_G}$  is a function of  $Q_L$  and  $Q_G$ . Flow rates maximising this ratio are the optimum flow rates for dissolution.

Fig. 6. Measurements and extrapolation of the evolution of  $\eta_{biomassconv}$ ,  $\eta_{diss}$  and  $\eta_{stripped}$  as a function of the total tube length of the photobioreactor,  $L_{tubes}$ , for carbon dioxide concentration in the injected gas up to 10% and non-optimised flow rates.







time





