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HYDRODYNAMIC CHARACTERISTICS OF A STRUCTURED BED REACTOR SUBJECTED TO RECIRCULATION AND INTERMITTENT AERATION (SBRRIA)

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Abstract - This work aimed at evaluating the effect of recirculation ratio on the degree of mixing in the flow of a structured bed reactor. Stimulus-response assays were carried out with hydraulic retention times of 6 h and 12 h, subjected to recirculation ratios of 0, 1, 2, 3, and 4. The assays were undertaken with dextran blue as a tracer and without aeration. The results fitted a compartment model, allowing the determination of the mixed-flow and plug-flow volumes inside the reactor. The model application showed that recirculation ratios between 1 and 3 do not increase the mixed-flow volume of the reactor. Higher mixed-flow volumes were obtained at recirculation rates higher than 4 for both 6 h and 12 h of hydraulic retention times. The results obtained in this research are important for future applications of this reactor configuration, promoting a mixed flow in the reactor with a minimum recirculation ratio.

Keywords: structured bed reactor, hydrodynamic assay, compartment model, recirculation ratio.

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INTRODUCTION

The simultaneous removal of organic matter and nitrogen in a single reactor has been the subject of several recent researches on wastewater treatment (Liu and Dong, 2011; Moura et al., 2012; Yao et al., 2013). The application of systems based on single-tank reactors presents advantages when compared to conventional ones, as it is unnecessary to install two or more tanks to promote the removal of these pollutants.

The conventional biochemical processes involved in nitrogen removal are denominated nitrification and denitrification. In the nitrification process, ammonium nitrogen (NH_4^+-N) is oxidized to nitrite (NO_2^--N) and nitrate (NO_3^--N) by aerobic chemoautotrophic microorganisms. On the other hand, denitrification occurs in the absence of oxygen, where NO_2^--N and NO_3^--N are reduced to N_2 by heterotrophic microorganisms in the presence of an electron donor (Metcalf and Eddy, 2003; Koops et al., 2006; Canto et al., 2008). For the occurrence of both the processes in a single reactor, the application of systems with intermittent aeration has been the aim of several studies, promoting aerobic conditions for nitrification and anoxic conditions for denitrification.

The application of intermittent aeration for nitrogen removal has been employed mainly in batch reactors (Canto, et al., 2008; Chen et al., 2011; Coma et al., 2012). In this system, there is no variation of effluent characteristics due to intermittent aeration, as the effluent is only discharged after a complete cycle of aeration and non-aeration. Furthermore, the addition of an electron donor during the non-aerated phase is necessary to promote denitrification. However, when intermittent aeration is applied to continuously fed reactors, the variations in the effluent characteristics should be minimized, avoiding peak concentrations of the pollutants in aerated or non-aerated periods. This is possible in continuous fed reactors if the hydrodynamic characteristics are close to those of complete mixing reactors. In this case, therefore, a complete and accurate hydrodynamic characterization of the reactor is mandatory for a safe and economic design of such reactors.

Reactor hydrodynamic characteristics are generally obtained with stimulus-response tracer assays. In these assays, the residence time distribution (RTD) is obtained by adding an impulse of inert tracer to the reactor influent and measuring the time-dependent tracer concentration in the effluent (Stevens et al., 1986). With the help of the RTD curve, it is possible to obtain the mixing degree of the liquid inside the reactor (Levenspiel, 1999). If the RTD curve response is close to Gaussian, the dispersion or tanks in series model is useful for representing the flow inside the vessels. However, in other conditions, compartment models and all types of other models can be evaluated for scaling up or for diagnosing poor flow (Levenspiel, 2012).

The continuous structured bed reactor subjected to recirculation and intermittent aeration (SBRRIA) has been employed successfully to simultaneously incorporate organic matter and nitrogen removal (Moura et al., 2012; Barana et al., 2013; Wosiack et al., 2015; Santos et al., 2016). However, the recirculation ratio, which is an important operational parameter for maintaining low variation of effluent characteristics, was not determined by a rational basis test using hydrodynamic assays. Moura et al., (2012) operated the SBRRIA with the recirculation ratio equal to 5 in order to promote the characteristics of a complete mix in the flow of the reactor. Barana et al., (2013) adopted a recirculation ratio equal to 6 to ensure the SBRRIA would operate as a non-ideal complete mix reactor. Despite the success of these studies, it is important to determine the minimum recirculation ratio that promotes high degree of liquid mixing inside the reactor; this would increase the efficiency of the overall process from an economic standpoint. It is hypothesized that there is a lowest value of the recirculation ratio that ensures efficient mixing conditions at lower energy consumption.

The aim of this study was to evaluate the effect of the effluent recirculation ratio on the degree of liquid mixing of the structured bed reactor. The data obtained would provide information for designing SBRRIA reactors with stable operation and high performance under minimum recirculation ratio.

MATERIALS AND METHODS

The evaluation of the hydrodynamic characteristics of the SBRRIA was carried out by stimulus-response experiments under abiotic conditions.

The assays were performed without aeration as the air bubbles can positively affect the degree of mixing in the reactor. Therefore, the results of this study determine an optimal critical mixing condition inside the reactor, which allows establishing the lowest recirculation ratio to promote a sufficient degree of liquid mixing for a good operation of the system. The assays were performed at room temperature (20-25ºC).

The schematic system is shown in Figure 1. The reactor had a height and internal diameter of 80 cm and 14.5 cm, respectively, with a total volume of 11.6 L (working volume of 6.1 L). Continuous feed and effluent recirculation inlets were located at the bottom of the reactor (represented by A1 and A2, respectively). The recirculation and effluent discharge outlets were at 65 cm (A4) and 70 cm (A3) from the bottom of the reactor, respectively. Thirteen cylindrical polyurethane foam structures (diameter and length of 3 cm and 60 cm, respectively) were employed as a support medium for biomass growth (F-F Cut, Fig. 1) (Moura et al., 2012).

Tracer evaluation experiments were carried out with theoretical hydraulic retention times (HRT_t) of 12 h and 6 h, corresponding to the flow rate of 1 $L.h^{-1}$ and 0.5 $L.h^{-1}$, respectively. For each HRT_t, assays with recirculation ratios (R) of 0 (R0), 1 (R1), 2 (R2), 3 (R3), and 4 (R4) were conducted. The reactor was fed with tap water.

When the steady-state regime flow was obtained, the tracer solution was injected in the effluent stream as a pulse (10 seconds of injection time). The tracer used was dextran blue. This compound is considered to be an adequate tracer because its adsorption or diffusion into the packing media is very low (Jimenez et al., 1988; De Nardi et al., 1999). 50 mL of tracer solution volume was used in each experimental condition with concentration of 8 $g.L^{-1}$ for assays with recirculation ratio equal to zero and 4 $g.L^{-1}$ for the remaining assays. The assays presented a high percent of the tracer recovery, with values higher than 76%.

The samples were collected at the effluent of the reactor and pumped continuously (Gilson Minipuls® pump) for online detection in the spectrophotometer (wavelength of 650 nm), where absorbance data were registered and stored at intervals of 5 minutes. The concentration of dextran blue in the effluent (C) was obtained with a standard calibration curve. After determining the series of concentrations of dextran (C) at time (t), the exit age distribution curve (E) was determined (Equation 1).

$$E = \frac{C(t)}{\int_0^{\infty} C(t)dt} \tag{1}$$

A SBRRIA without effluent recirculation tends to behave as a non-ideal plug-flow reactor. On the other hand, it tends to behave as a non-ideal mixed-flow reactor when it is subjected to high effluent recirculation ratios. Thereby, as the traditional models proposed by Levenspiel (1999) did not fit well the results, an alternative model was tested to simulate the hydraulic behavior in the SBRRIA (Equation 2, Figure 2). Equation 2 is based on the combination of two compartments in series, one complete mix and the other plug flow (regardless of order). The hypothesis of the model is that higher recirculation ratios applied promote higher percentages of mixed volume. The equation was deduced from Levenspiel (2012) considering that the exponential decay after the tracer peak follows a Gaussian curve Levenspiel (1999).

$$E_{adj} = \frac{\nu}{V_m} \times \exp\left[-\frac{\nu}{V_m} \times t + \frac{V_P}{V_m}\right] \tag{2}$$

In this model, it is possible to determine the mixed flow (V_m) and plug flow (V_p) volumes. There are two ways to define V_m and V_p - one is by using the adjusted E-curve (E_{adj}) (Equation 2) and the other by the use of the experimental E-curves (Equation 1) with the determination of the time values at the peak (Equations 3 e 4). After the determination of V_p and V_m, the mixing degree ($\%_{mix}$) (Equation 5) in the reactor was

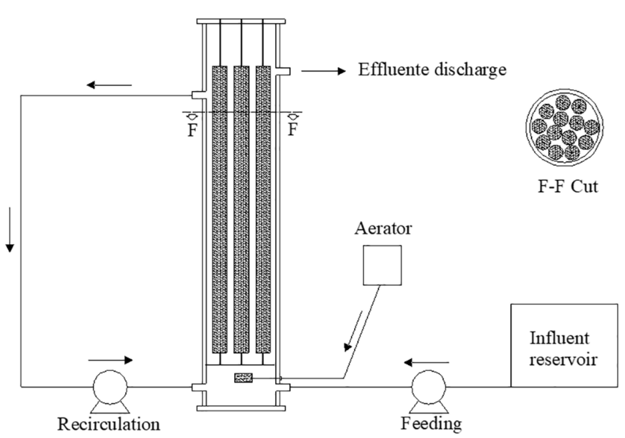

Figure 1. Scheme of structured bed reactor subjected to recirculation and intermittent aeration (Moura et al., 2012).

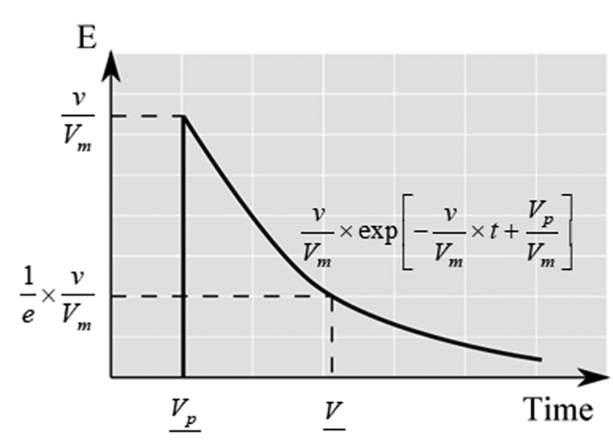

Figure 2. E curve for a combination of CSTR and PFR vessels (adapted from Levenspiel, 2012).

estimated in relation to the experimental total volume (Equation 6).

$$V_m = \frac{\nu}{E - curve\ peak} \quad (3)$$

$$V_P = \nu \times time\ in\ E - curve\ peak \quad (4)$$

$$\%_{mix} = \frac{Vm}{V} \times 100 \quad (5)$$

$$V = V_m + V_P \quad (6)$$

In this study, the values of V_m and V_p were determined by fitting and statistically comparing with the values determined at the peak, using the t paired test. Equation 2 was adjusted by non-linear fitting using the Gauss-Newton algorithm, minimizing the residual sum of squares (Bates and Watts, 1988). The normality of residuals was evaluated by the Shapiro-Wilk test (Royston, 1995).

The significance of the coefficients, V_m and V_p, to describe the experimental data was determined using t-student test. All statistical tests were performed with a 95 % confidence level, using the p-value to facilitate the interpretation of results. The error propagation was performed according to the study by Taylor (1997). The calculations and the statistical analysis were performed with R 3.3.0 (R Core Team, 2016).

RESULTS AND DISCUSSION

The application of flow models is useful to characterize the flow in reactors or even to diagnose their problems. If the flow is close to plug or mixed, the dispersion or tanks in series models and the recirculation models are usually used (Levenspiel, 2012). However, when none of the traditional models represent well the flow in the reactor, the compartment models represent an important alternative to its characterization. The models proposed by Levenspiel (1999) (dispersion or tanks in series models) were tested and did not represent well the flow inside the reactor. These models were then disregarded because they were not statistically significant in the study (data not shown).

The RTD curves obtained experimentally are presented in Figure 3. Table 1 presents the values of V_m and V_p and the corresponding standard errors determined from the adjustment of Equation 1 to experimental data. The values of the experimental total volume (V), the experimental hydraulic retention times (HRT_e), and percentage volumes of the mixed flow ($\%_{mix}$) were determined and are shown in Table 1. The errors of these coefficients were determined by means of an error propagation technique (Taylor, 1997).

Jimenez et al., (1988) and De Nardi et al., (1999) obtained good results using dextran blue as a tracer in the hydrodynamic assay due to the low diffusion

Table 1. Values of the coefficients determined in the adjustment to the compartment model.

HRTt	***R***	V_m±***SE***(L)*	V_p±***SE***(L)*	***V***±***SE***(L)*	***HRTe***±***SE***(h)*	$\%_{mix}$±***SE***(%)*
6 h	R0	4.85±0.32 *p-value = 0*	4.09±0.25 *p-value = 0*	8.94±0.41	8.94±0.41	54.3±4.4
	R1	5.28±0.52 *p-value = 0*	1.97±0.46 *p-value = 0.001*	7.26±0.69	7.26±0.69	72.8±10.0
	R2	3.96±0.20 *p-value = 0*	1.21±0.15 *p-value = 0*	5.17±0.25	5.17±0.25	76.6±5.4
	R3	3.71±0.40 *p-value = 0*	0.96±0.35 *p-value = 0,02*	4.66±0.53	4.66±0.53	79.5±12.4
	R4	3.94±0.47 *p-value = 0*	0.20±0.40 *p-value = 0,63*	4.14±0.62	4.14±0.62	95.1±4.9
12 h	R0	2.80±0.52 *p-value = 0.001*	2.07±0.36 *p-value = 0*	4.87±0.64	9.74±1.27	57.5±3.2
	R1	3.69±0.30 *p-value = 0*	1.85±0.26 *p-value = 0*	5.54±0.40	11.09±0.80	66.5±2.6
	R2	3.33±0.70 *p-value = 0.003*	1.77±0.63 *p-value = 0.03*	5.10±0.94	10.21±1.90	65.2±4.1
	R3	4.60±0.46 *p-value = 0*	1.19±0.42 *p-value = 0.02*	5.79±0.62	11.57±1.25	79.5±3.6
	R4	4.50±0.32 *p-value = 0*	0.64±0.27 *p-value = 0.04*	5.14±0.42	10.28±0.84	87.6±3.4

*Standard Error

Table 2. Paired t-test to evaluate differences between the determination of volumes by the fitting and by the peak.

		HRT_t	
		6h	12h
V_m	p-value	0.23	0.38
	t-computed	1.404	0.98
	t-tabulated	2.776	
V_p	p-value	0.69	0.61
	t-computed	0.431	0.560
	t-tabulated	2.776	
V	p-value	0.31	0.83
	t-computed	1.18	0.453
	t-tabulated	2.776	

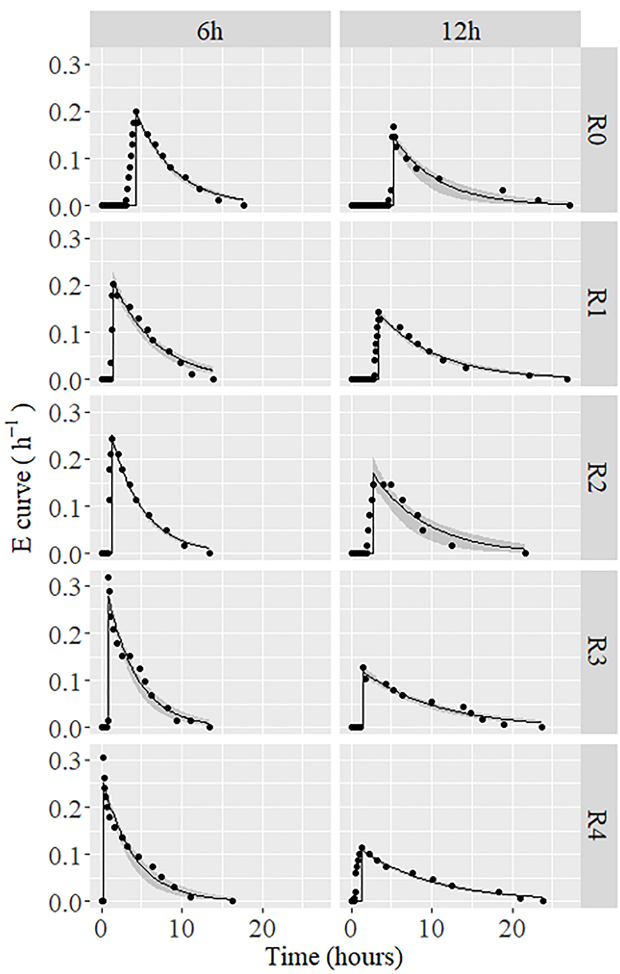

Figure 3. Experimental E-curves in a SBRRIA operating at HRTt of 6 h and 12 h (●) and E-curves of the compartment model (−). The ribbon represents the 95 % confidence interval for the adjusted model.

of this tracer in the support material. However, the authors emphasize that the application of this tracer in reactors with different support materials and with low flow velocity could generate different results of tracer diffusion. The traditional dispersion and the tanks-in-series models, proposed by Levenspiel (1999), are not sufficient to characterize the flow when either plug flow or mixed flow is assumed in a real vessel and when there are large deviations from ideal reactors.

The representation of flow by means of a compartment model allowed quantifying the percentage of mixed and non-mixed volumes within the reactor. The high adherence of the model parameters to the data is demonstrated by the respective p-values (Table 2). The p-value is used in hypothesis testing, notably to evaluate the significance of the null hypothesis in statistical tests. Therefore, the interpretation depends on the null hypothesis used in the test. For the coefficients of Equation 1, the null hypothesis was that the coefficients are not significant to explain the experimental data when used in this equation. Except for the assay with HRT_t of 6 h and recirculation of R4, all the p-values were less than 0.05, indicating that they are important in the mathematical model used. In this assay, such as when the mixing percentage is close to 100 %, the traditional flow models proposed by Levenspiel (1999) may describe the behavior of the reactor, which explains the statistical rejection of the coefficient V_p.

The SBRRIA was designed so that the flow is parallel to the polyurethane foam cylinders (Moura et. al., 2012). Thus, the mass transport of the pollutant inside the cylinders takes place by the concentration gradient between the liquid and the foam surface. For the HRT_t of 6 h and 12 h, this behavior can be observed by the similar values of theoretical and experimental total volumes (Table 1), indicating that the tracer did not penetrate the foam. Trends which reduce the experimental total volumes of the reactor with the increase of the recirculation ratio can also be observed with a HRT of 6 h. This event may have occurred due to the formation of preferred paths inside the reactor, which can affect the efficiency of the reactor negatively. This behavior did not occur with a HRT of 12h.

Another important design variable of the SBRRIA is the mixing percentage in the reactor ($\%_{mix}$) (Moura et al., 2012). From R1 to R3, there is little variation in the mixing percentage in the HRT_t of 6 h. For HRT_t of 12 h, the behavior is similar, showing that the percentage of mixing can be achieved even at lower recirculation ratios. These results are significantly lower than the values of recirculation ratio used by Moura et al. (2012), Barana et al. (2013), and Santos et al. (2016), who adopted recirculation ratios equal to 5, 6, and 5, respectively. High values of the recirculation ratio promote higher pumping energy expenditure, reducing the comparative advantage of this system.

Figure 4 shows the V_m, V_p, and V values determined in the SBRRIA by peak analysis and by adjustment of Equation 1 to the experimental data after the peak. Table 2 shows the results of paired t-test to evaluate differences between the two techniques.

The determination of the volumes by any of the techniques presented similar results. The volumes determined from the peak in most cases were within the standard error of the amount determined by the equation of adjustment (Figure 4). The paired t-test confirmed statistically that the p-values are greater than 0.05 (the null hypothesis was that the difference between the techniques is equal to zero). Although the determination from the peak is slightly easier than the mathematical fit to the experimental data, the latter allows the determination of confidence intervals and other key statistical information for the characterization of reactive systems.

Processes that used reactors with anaerobic and aerobic zones for simultaneous nitrogen and carbon removal promoted an increase in total nitrogen removal with the increase of the recirculation ratio.

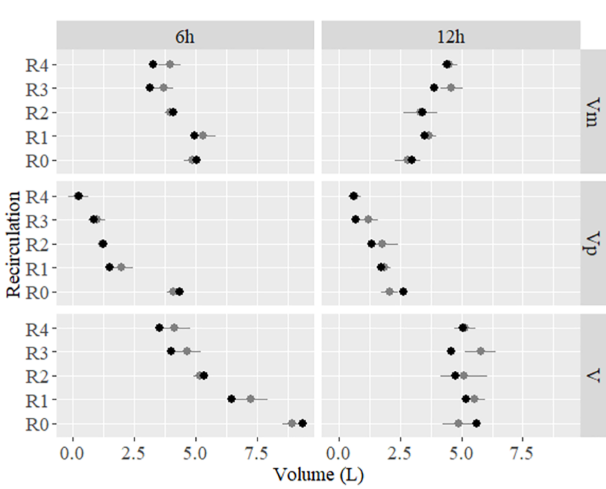

Figure 4. Mixed flow volume (Vm), plug flow volume (Vp) and experimental total volume (V) determined in the SBBIA operating at HRTt of 6 h and 12 h by peak analysis (●) and by adjustment of Equation 1 to the experimental data after the peak (-●-). The continuous lines correspond to the standard error of the parameters with 95 % of confidence.

Netto and Zaiat (2012) operated an anaerobic-aerobic fixed-bed reactor with recirculation of the liquid phase to promote chemical oxygen demand (COD) and total nitrogen removal from sewage. The authors concluded that upon increasing the recirculation ratio from 0.5 to 1.5 (recirculation from aerobic zone to anaerobic zone), the total nitrogen removal efficiency increased from 65 % to 75 %, with COD removal higher than 90 % in both conditions.

Araújo and Zaiat (2009) worked with an up-flow fixed-bed anaerobic-aerobic reactor for removal of organic matter and nitrogen from L-lysine plant wastewater. The authors observed that increasing the liquid recirculation ratio from the aerobic zone to anaerobic zone promoted an increase of total nitrogen removal. The total nitrogen removal efficiency increased from 42 % to 77 % by increasing the recirculation ratio from 0.5 to 3.5, maintaining COD removal efficiencies at an average of 95 %.

In compartmented reactors, the increase of recirculation ratio from the aerated zone to the anaerobic zone provides a larger amount of oxidized nitrogen (nitrite and nitrate) to the anaerobic zone, favoring the denitrification process (region with low dissolved oxygen concentration and with the presence of electron donors). Consequently, the total nitrogen removal increases.

On the other hand, in the SBRRIA, the nitrification and denitrification processes occur simultaneously in the reactor due to both diffusion of oxygen in the carrier and intermittent aeration (Moura et al., 2012). Therefore, the recirculation of the effluent is to ensure the behavior of complete mix flow inside the reactor, avoiding peaks of ammonia concentration during the non-aerated phase and nitrate concentration during the aerated phase in the effluent. Consequently, lower recirculation ratios that ensure the behavior of a complete mixing reactor will result in improved energy efficiency because of the use of a low power pump. Recent data showed efficiencies above 80 % for total nitrogen removal with the recirculating ratio equal to 3 in the SBRRIA reactor operating under intermittent aeration in sewage treatment (unpublished data).

CONCLUSIONS

The results obtained allowed us to conclude that there was no significant increase in the degree of mixing inside the reactor for recirculation ratios between 1 and 3. Consequently, simultaneous removal of nitrogen and organic matter can be obtained with reduced recirculation ratio, thus reducing the energy required for the operation of such reactor configuration. According to the proposed model, the completely mixed volume of the reactor reaches values of approximately 90 % of the experimental total volume for the recirculation ratio of 4. Therefore, the simultaneous nitrification and denitrification processes can be negatively affected due to the higher transport of liquid into the support medium. Finally, the results obtained in this study can significantly contribute to the application of this reactor configuration, as it establishes an important operational parameter for SBRRIA.

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NOMENCLATURES

SBRRIA - Structured bed reactor subjected to recirculation and intermittent aeration
CSTR - Continuous stirred-tank reactor
PFR - Plug flow reactor
COD - Chemical oxygen demand ($mg.L^{-1}$)
NH_4^+-N - Ammoniacal nitrogen ($mg.L^{-1}$)
NO_2^--N - Nitrite nitrogen ($mg.L^{-1}$)
NO_3^--N - Nitrate nitrogen ($mg.L^{-1}$)
N_2 - Nitrogen gas ($mg.L^{-1}$)
RTD - Residence time distribution
HRT_t - Theoretical hydraulic retention times (h)
HRT_e - Experimental hydraulic retention time (h)
R0 - Recirculation ratio equal to 0
R1 - Recirculation ratio equal to 1
R2 - Recirculation ratio equal to 2
R3 - Recirculation ratio equal to 3
R4 - Recirculation ratio equal to 4
C - Concentration of dextran blue in effluent ($mg.L^{-1}$)
t - Time (h)EExit age distribution curve (h^{-1})
E_{adj} - Adjusted E-curve (h^{-1})
M - Instantaneous pulse of tracer ($mg.h.L^{-3}$)
v - Flow rate ($L.h^{-1}$)
Vm - Mixed flow volume (L)
Vp - Plug flow volume (L)
V - Experimental total volume (L)
$\%_{mix}$ - Percentage volumes of the mixed flow

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