K_La Determination Using the Effectiveness-NTU Method: Application to Countercurrent Absorbers in Operation Using Viscous Solvents for VOCs Mass Transfer

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Abstract: In this study, the Effectiveness-NTU method, which is usually applied to heat exchanger design, was adapted to gas–liquid countercurrent absorbers to determine the overall mass transfer coefficient, K_La, of the apparatus in operation. It was demonstrated that the ε-NTU method could be used to determine the K_La using the Henry coefficient of the solute to be transferred (H_{VOC}), the gas flow-rate (Q_G), the liquid flow-rate (Q_L), the scrubber volume (V), and the effectiveness of the absorber (ε). These measures are calculated from the gaseous concentrations of the solute measured at the absorber inlet (C_{Gin}) and outlet (C_{Gout}), respectively. The ε-NTU method was validated from literature dedicated to the absorption of volatile organic compounds (VOCs) by heavy solvents. Therefore, this method could be a simple, robust, and reliable tool for the K_La determination of gas–liquid contactors in operation, despite the type of liquid used, i.e., water or viscous solvents.

Keywords: absorption; mass transfer; heat exchanger; scrubber; effectiveness-NTU method

1. Introduction

The removal of volatile organic compounds (VOCs) from the air can be achieved using gas–liquid reactors. However, some VOCs are poorly soluble in water leading to mass transfer limitations. In response to the low solubility of hydrophobic pollutants, water can be replaced by heavy viscous organic solvents (silicone oils, alkanes, phthalates, [1]) to improve the mass transfer [2]. Basically, the absorption step is crucial, since VOCs have to be solubilized for further treatments. Usually, the mass transfer between the gas phase and the liquid phase (water) is performed in countercurrent gas–liquid packed absorbers, where the design procedure is clearly described in the extant literature [3–5]. Basically, the design of absorbers implies the use of hydrodynamic studies to determine the gas–liquid two-phase flows in the column and mass transfer studies, so as to calculate the absorption efficiency according to the height of the column. The overall volumetric mass transfer coefficient K_La, which depends on the local volumetric mass transfer coefficients (k_La, k_Ga) and on the Henry coefficient, is a key parameter in the calculation of the absorption efficiency, and subsequently in the reactor design. However, for viscous solvents, the physico–chemical properties of the couple COV/solvent (mainly the Henry coefficient and the liquid diffusivity) are not sufficiently known to enable an accurate K_La prediction from empirical correlations. Additionally, in the case of VOCs absorption by mixtures of two immiscible liquids (i.e., multiphasic gas–liquid–liquid systems), the K_La cannot be predicted. Several techniques for experimental K_La determination exist; however, considerable problems concerning the accuracy of the measurements in relation to the high viscosity of the solvents have been encountered [6,7]. Moreover, these techniques cannot be applied to multiphasic gas–liquid–liquid systems. Therefore, the aim of this study was to develop a generally applicable method for experimentally determining

the overall volumetric mass transfer coefficient in the liquid phase, K_La, regardless of the solute to be transferred, the solvent and the packing material used, the operating conditions applied, and the hydrodynamic conditions encountered in the gas–liquid absorber. This method, based on the "effectiveness-NTU" method used for the design of heat exchangers [8], could then be considered as a complementary tool for K_La determination to the empirical correlations usually applied.

2. Gas–Liquid Absorber Analysis

Although the countercurrent flow in a gas–liquid absorber occurs in vertical devices due to the significant density difference between gas and liquid, the mass balance between the phases was presented horizontally in this study. This was done for convenience, and to facilitate an easier analogy with the heat exchanger (Figures 1 and 2). For dilute concentrations, the gas side mass balance can be written as:

$$\frac{Q_G}{V} C_G = d\phi + \frac{Q_G}{V} (C_G + dC_G) \tag{1}$$

Then, the amount of the solute transferred from the gas phase is:

$$d\phi = -\frac{Q_G}{V} dC_G \tag{2}$$

Similarly for the liquid side:

$$\frac{Q_L}{V} (C_L + dC_L) + d\phi = \frac{Q_L}{V} C_L \tag{3}$$

$$d\phi = -\frac{Q_L}{V} dC_L \tag{4}$$

Consequently:

$$dC_L = \frac{Q_G}{Q_L} dC_G \tag{5}$$

This equation can be rearranged as:

$$H_{VOC} \, dC_L = \frac{H_{VOC} \, Q_G}{Q_L} dC_G = \frac{1}{A} dC_G \tag{6}$$

The flow–rate ratio ($H_{VOC} \, Q_G/Q_L$) corresponds to the reciprocal of the absorption factor that is usually applied to the design of gas–liquid absorbers. Since the Henry coefficient can be expressed using different definitions, it is necessary to apply the appropriate units. For the purposes of this work, the parameter H_{VOC} was defined as the dimensionless air-to-water concentration ratio ($H_{VOC} = C_G/C_L$), which is the most convenient measure for mass distribution calculations. The alternative forms of the Henry coefficient and corresponding conversion factors are given in Reference [9].

Based on the assumption of dilute systems, i.e., the gas flow-rate and liquid flow-rate are constant along the apparatus (Figure 2), where the molar flux ϕ of the transferred solute is:

$$\phi = \frac{Q_L}{V}(C_{Lout} - C_{Lin}) = \frac{Q_G}{V}(C_{Gin} - C_{Gout}) \tag{7}$$

Consequently:

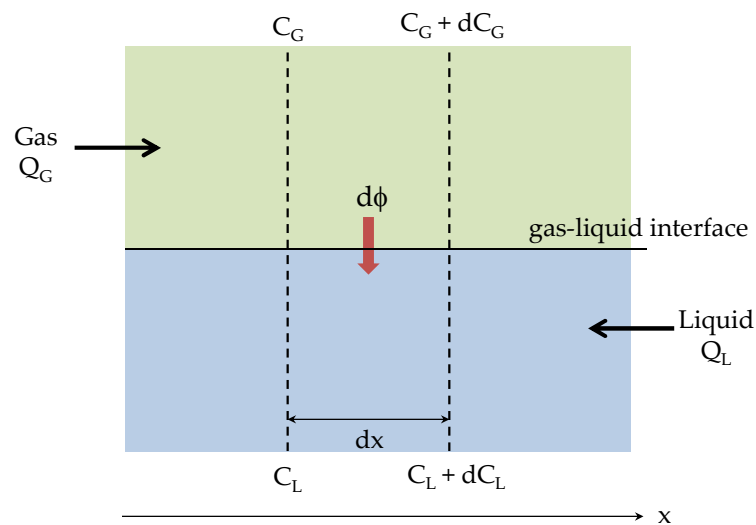$$C_{Lout} = C_{Lin} + \frac{Q_G}{Q_L}(C_{Gin} - C_{Gout}) \tag{8}$$

Figure 1. Nomenclature for the material balances in a countercurrent gas–liquid absorber.
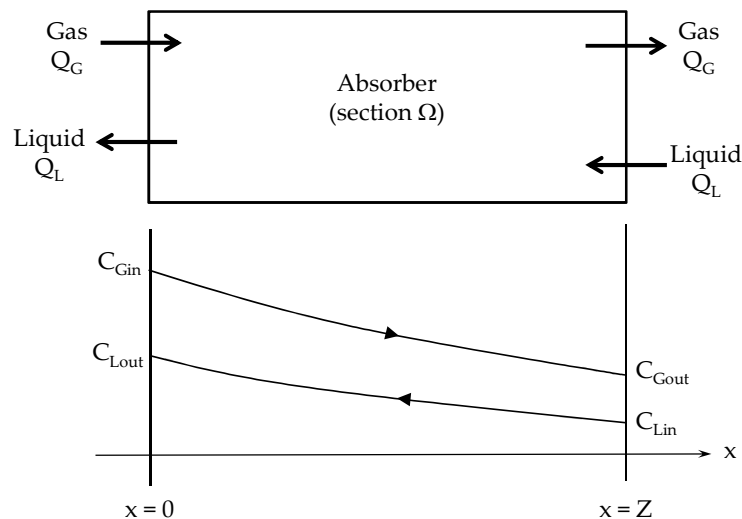

Figure 2. Concentration profile throughout the length of the gas–liquid countercurrent absorber.

Change in the solute concentration throughout the length of the absorber can be expressed as:

$$Q_G \frac{dC_G}{dx} = -\Omega K_L a \left(\frac{C_G}{H_{VOC}} - C_L \right) \tag{9}$$

To adequately integrate this equation, it is useful to make a change in the variables. Thus, using Equation (6):

$$d(C_G - H_{VOC} C_L) = dC_G - H_{VOC} dC_L = dC_G - \frac{1}{A} dC_G = \left(1 - \frac{1}{A}\right) dC_G \tag{10}$$

Replacing Equation (10) in Equation (9):

$$\frac{Q_G}{\left(1 - \frac{1}{A}\right)} d(C_G - H_{VOC} C_L) = \frac{-\Omega K_L a}{H_{VOC}} (C_G - H_{VOC} C_L) dx \tag{11}$$

Considering that concentrations of the values C_G and C_L are given at positions $x = 0$ and $x = Z$ in Figure 2, the solution to this equation, integrated between $x = 0$ and $x = Z$, is:

$$\ln\left[\frac{(C_{Gout} - H_{VOC}\,C_{Lin})}{(C_{Gin} - H_{VOC}\,C_{Lout})}\right] = \frac{-\Omega\,Z\,K_L a}{H_{VOC}\,Q_G}\left(1 - \frac{1}{A}\right) \tag{12}$$

As (ΩZ) is the volume of the apparatus, Equation (12) can be rearranged under the following form:

$$\frac{(C_{Gout} - H_{VOC}\,C_{Lin})}{(C_{Gin} - H_{VOC}\,C_{Lout})} = \exp\left(-NTU\left(1 - \frac{1}{A}\right)\right) \tag{13}$$

With:

$$NTU = \frac{K_L a}{H_{VOC}}\frac{V}{Q_G} \tag{14}$$

In Equation (14), the dimensionless number of transfer units (NTU) corresponds to the ratio of the gas residence time in the device (V/Q_G) to the time needed for the mass transfer of the solute from the gas phase to the liquid phase ($H_{VOC}/K_L a$). This NTU parameter can be directly compared to the NTU parameter used in the heat exchanger design (Table 1). Moreover, the reciprocal of the absorption factor ($H_{VOC}\,Q_G/Q_L$) is also directly comparable to the relative magnitudes of the hot and cold fluid heat capacity rates that are used in the heat exchanger design. Consequently, the analogy with the heat exchanger design implies the definition of the maximum possible mass transfer rate ϕ_{max} between the gas phase and the liquid phase. Assuming a countercurrent absorber of infinite length, the solute concentrations in the gas phase and in the liquid phase are at an equilibrium. Therefore, C_{Gout} tends to ($H_{VOC}\,C_{Lin}$) at $x \to \infty$ and:

$$\phi_{max} = \frac{Q_G}{V}(C_{Gin} - H_{VOC}\,C_{Lin}) \tag{15}$$

The effectiveness of the absorber, ε, can be defined as the ratio of the actual mass transfer rate ϕ (Equation (7)) to the maximum possible mass transfer rate ϕ_{max}:

$$\varepsilon = \frac{\phi}{\phi_{max}} = \frac{(C_{Gin} - C_{Gout})}{(C_{Gin} - H_{VOC}\,C_{Lin})} \tag{16}$$

According to Equation (16), the solute gas concentration at the outlet of the absorber C_{Gout} would tend toward zero only for a countercurrent absorber of infinite length using a solute-free liquid, i.e., $C_{Lin} = 0$. It should be noted that the inlet liquid concentration is usually equal to zero, and consequently $\varepsilon = (C_{Gin} - C_{Gout})/C_{Gin}$, which corresponds to the equation that is generally applied to determine the absorber efficiency. Combining Equations (8), (13), and (16), the effectiveness of the absorber can be expressed using the following form (calculation details are given in Appendix A):

$$\varepsilon = \frac{1 - \exp\left(-NTU\left(\frac{A-1}{A}\right)\right)}{1 - \frac{1}{A}\exp\left(-NTU\left(\frac{A-1}{A}\right)\right)} \tag{17}$$

Equation (17) becomes identical to the expression used for the countercurrent heat exchanger. The effectiveness versus the NTU is plotted in Figure 3, for various A values. This analogy between countercurrent devices, absorbers, and heat exchangers, can be extended to a gas–liquid stirred tank reactor (STR), as well as to a single stream heat exchanger (typically, a boiler or a condenser). In a gas–liquid STR, the gas flow-rate is dispersed through a known volume of a well-mixed liquid phase, and consequently $Q_L = 0$. In this case, the effectiveness of the absorber is presented as (calculation details are given in Appendix B):

$$\varepsilon = 1 - \exp(-NTU) \tag{18}$$

In the case of a single stream heat exchanger, A tends to ∞, and consequently, the heat exchanger behavior of the apparatus becomes independent of the flow arrangement. As a result, when $A \to \infty$,

Equation (17) is reduced to Equation (18). The analogies between the heat and mass transfer given in the current paper are summarized in Table 1.
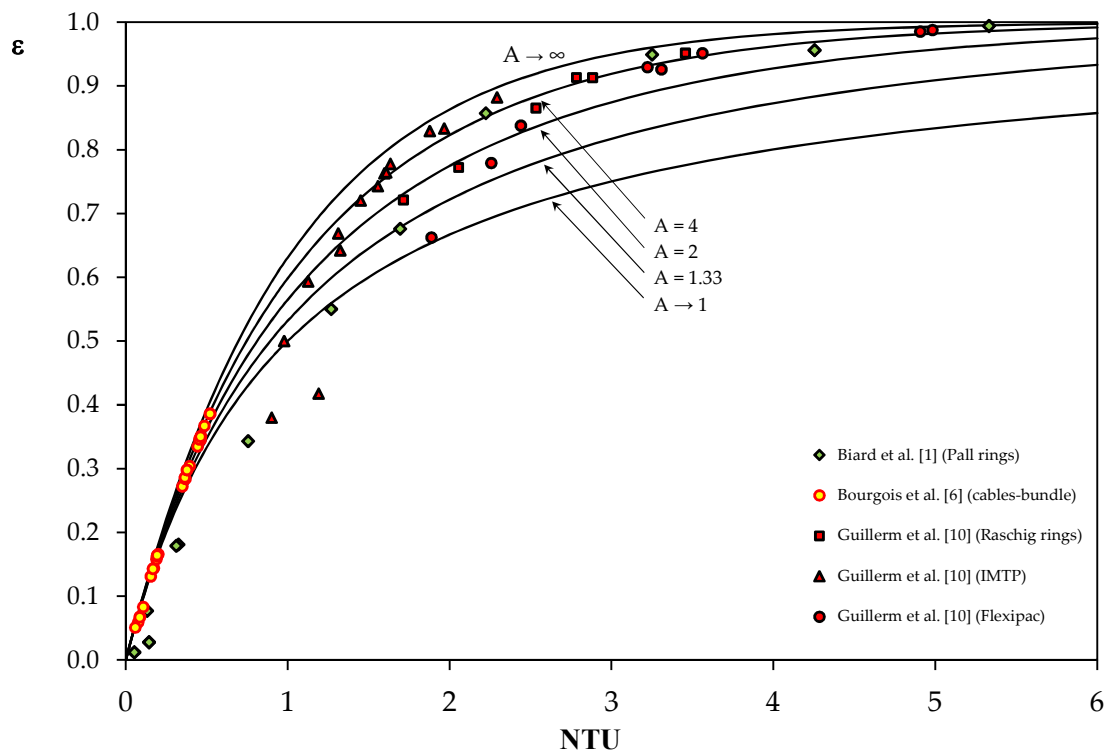

Figure 3. Effectiveness (ε) of a countercurrent gas–liquid absorber versus the number transfer units (NTU).

Table 1. The effectiveness-NTU method for the countercurrent devices. Analogy between the gas–liquid mass absorber and the heat exchanger (see nomenclature for details).

	Mass Absorber	Heat Exchanger
Physical parameter	Concentration C (mol m^{-3})	Temperature T (K)
Transfer coefficient	K_L (m s^{-1})	U (W m^{-2} K^{-1})
Transfer surface area	a V (m^2)	A (m^2)
Transfer rate	$\phi = \frac{Q_L}{V}(C_{Lout} - C_{Lin})$ $\phi = \frac{Q_G}{V}(C_{Gin} - C_{Gout})$ $\phi = K_L a\, \Delta C_{ML}$ (mol m^{-3} s^{-1})	$\phi = M_c C_{pc} (T_{cout} - T_{cin})$ $\phi = M_h C_{ph} (T_{hin} - T_{hout})$ $\phi = U A\, \Delta T_{ML}$ (W)
Maximum possible transfer rate	$\phi_{max} = \frac{Q_G}{V}(C_{Gin} - H_{VOC}C_{Lin})$ (mol m^{-3} s^{-1})	$\phi_{max} = C_{min}(T_{hin} - T_{cin})$ (W)
Effectiveness (dimensionless) $\varepsilon = \frac{\phi}{\phi_{max}}$	$\varepsilon = \frac{(C_{Gin} - C_{Gout})}{(C_{Gin} - H_{VOC}\,C_{Lin})}$ $\varepsilon = \frac{1 - \exp\left(- NTU\left(1-\frac{1}{A}\right)\right)}{1 - \frac{1}{A}\exp\left(- NTU\left(1-\frac{1}{A}\right)\right)}$ $\frac{1}{A} = \frac{H_{VOC}\,Q_G}{Q_L}$	$\varepsilon = \frac{\max((T_{cout} - T_{cin}),(T_{hin} - T_{hout}))}{(T_{hin} - T_{cin})}$ $\varepsilon = \frac{1 - \exp(- NTU\,(1-R))}{1 - R\,\exp(- NTU\,(1-R))}$ $R = \frac{C_{min}}{C_{max}}$
NTU (dimensionless)	$\frac{K_L a}{H_{VOC}}\frac{V}{Q_G}$	$\frac{U A}{C_{min}}$

3. Validation of the ε-NTU Method from the Data Reported in the Literature

The aim of this section is to demonstrate that the use of the ε-NTU method permits the calculation of the overall mass transfer coefficient, $K_L a$, of an operating absorber when parameters Q_L, Q_G, V, H_{VOC}, and ε are known. The validation is based on data from three studies [1,6,10], as detailed in Table 2. As observed, the experimental data are diversified in terms of solvent used, solutes to be transferred (i.e., various H_{VOC} values), and operating conditions. From the absorption efficiency values reported in these studies, the NTU and $K_L a$ values were calculated using Equations (19) and (14), respectively.

$$\text{NTU} = \frac{A}{1-A} \ln\left(\frac{\varepsilon - 1}{\varepsilon/A - 1}\right) \tag{19}$$

All experimental results are reported in Figure 3.

Table 2. Literature data used for the $K_L a$ determination using the ε-NTU method.

Solvent	Solute	H_{VOC}	Operating Conditions	Ref
Water 1 mPa s	Toluene Dichloromethane Propanol Acetone	2.09×10^{-1} 9.03×10^{-2} 1.27×10^{-4} 9.36×10^{-4}	V = 2.36 m³ Packed column (Pall rings) $Q_L = 3.06 \times 10^{-3}$ m³/s Q_G: 1.20 m³/s (gas EBRT: 2 s) 20 °C	Study n°1 [1]
DEHA (bis(di-2-ethylhexyl)adipate) 12.5 mPa s	Toluene Dichloromethane Propanol Acetone	3.12×10^{-4} 1.94×10^{-3} 2.70×10^{-3} 5.21×10^{-3}		
Silicone oil 50 mPa s	Toluene Dichloromethane Propanol Acetone	5.58×10^{-4} 9.04×10^{-3} 8.79×10^{-3} 2.17×10^{-2}		
DEHP (di-2-ethylhexyl phthalate) 76 mPa s	Hexane Toluene Octane Methylcyclohexane	4.74×10^{-3} 3.19×10^{-4} 4.40×10^{-4} 9.79×10^{-4}	V = 1.46×10^{-2} m³ Cables-bundle contactor $Q_L = 1.00 \times 10^{-5}$; 1.25×10^{-5}; 1.50×10^{-5} m³/s Q_G: 1.39×10^{-2} m³/s (gas EBRT: 1 s) 20 °C	Study n°2 [6]
Silicone oil 50 mPa s	Toluene	1.17×10^{-3}	V = 1.13×10^{-2} m³ Packed column (Raschig rings) $Q_L = 1.53 \times 10^{-5}$–$4.58 \times 10^{-5}$ m³/s Q_G: 7.15×10^{-3} m³/s (gas EBRT: 1.6 s) 25 °C	Study n°3 [10]
Silicone oil 50 mPa s	Toluene	1.17×10^{-3}	V = 1.13 10^{-2} m³ Packed column (IMTP®) $Q_L = 6.62 \times 10^{-6}$–$1.16 \times 10^{-4}$ m³/s Q_G: 1.10×10^{-2} m³/s (gas EBRT: 1 s) 25 °C	Study n°3 [10]
Silicone oil 50 mPa s Silicone oil 5 mPa s	Toluene Toluene	1.17×10^{-3} 1.09×10^{-3}	V = 1.13×10^{-2} m³ Packed column (Flexipac®) $Q_L = 1.36 \times 10^{-5}$–$9.28 \times 10^{-5}$ m³/s Q_G: 1.10×10^{-2} m³/s (gas EBRT: 1 s) 25 °C	Study n°3 [10]

3.1. Data from Study n°1

Biard et al. [1] proposed a simulation of the absorption of hydrophilic and hydrophobic compounds (toluene, dichloromethane, propanol, and acetone) in pure water and pure organic solvents (silicone oil and DEHA, i.e., bis(di-2-ethylhexyl)adipate). A countercurrent column packed with metal Pall rings was used for this purpose. The authors calculated the $K_L a$ values according to the Billet–Schultes

correlation coupled with the Piché et al. correlation [1]. From the removal efficiencies reported in this study, the ε-NTU method was applied to the 12 cases considered. Results are detailed in Table 3. The $K_L a$ values calculated using the ε-NTU method for heavy solvents, i.e., silicone oil and DEHA, were compared to the values determined by the authors from the coupling of the Billet–Schultes and Piché theories (Figure 4). Logically, the results are in good agreement due to the similarity between the definitions used to express the absorber efficiency (i.e., Equation (2) given in Reference [1] and Equation (17) presented in the present paper). Consequently, it could be argued that the ε-NTU method could be a simple, robust, and reliable method to determine the $K_L a$ values of operating absorbers. Moreover, the calculation procedure highlights the fact that the $K_L a$ mainly depends on the H_{VOC} (Figure 5). Such an expected result was consistent with the mathematical expression of the $K_L a$ given in Equation (14).
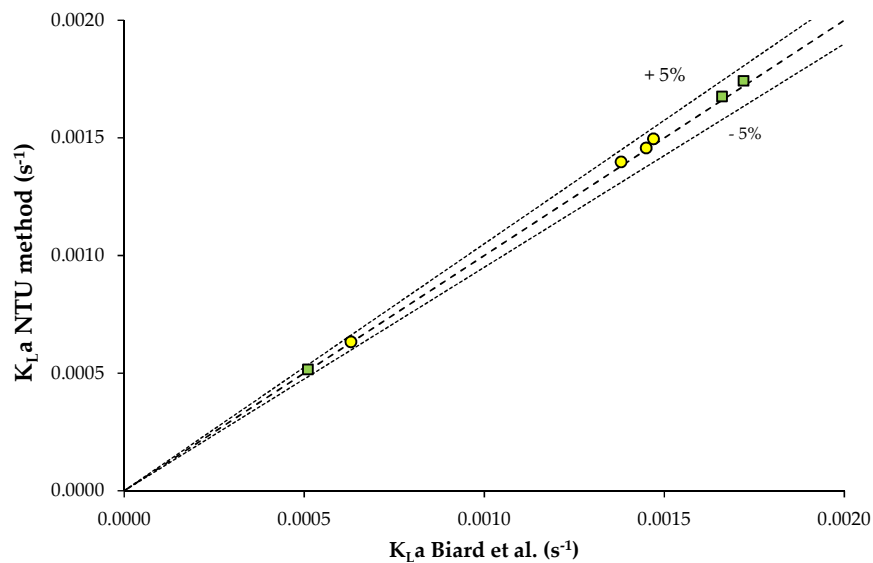

Figure 4. $K_L a$ parity plot obtained from the data analysis reported in Biard et al. [1] (circles: silicone oil; squares: DEHA; water data not provided by the authors).
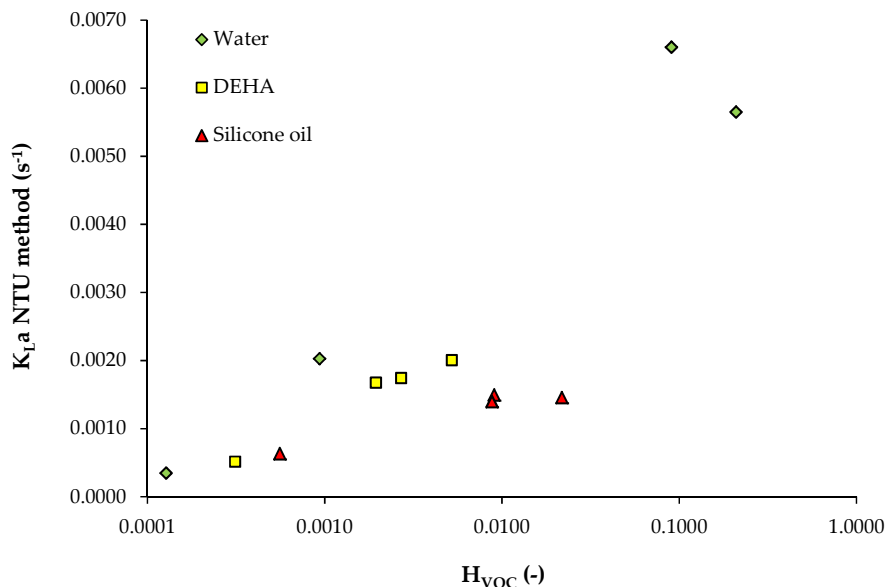

Figure 5. $K_L a$ versus the Henry coefficient (H_{VOC}) from the data analysis reported in Biard et al. [1]. For all the simulations: gas flow-rate = 1.20 m^3/s (gas EBRT = 2 s); liquid flow-rate = 3.06 × 10^{-3} m^3/s; and V = 2.36 m^3 (T = 20 °C).

Table 3. K_La determination using the data reported in Biard et al. [1]. For all the simulations: gas flow-rate = 1.20 m³/s (gas EBRT = 2 s); liquid flow-rate = 3.06 × 10⁻³ m³/s; and V = 2.36 m³ (T = 20 °C).

| Solute		H_{VOC} (-)	A (-)	ε (-)	NTU (-)	K_La (s⁻¹)
Toluene	Water	0.20936	0.012	0.012	0.053	5.63 × 10⁻³
	DEHA	0.00031	8.162	0.949	3.251	5.17 × 10⁻⁴
	PDMS50	0.00056	4.561	0.857	2.225	6.33 × 10⁻⁴
Dichloromethane	Water	0.09031	0.028	0.028	0.144	6.60 × 10⁻³
	DEHA	0.00194	1.311	0.676	1.695	1.68 × 10⁻³
	PDMS50	0.00904	0.282	0.181	0.325	1.50 × 10⁻³
Propanol	Water	0.00013	20.009	0.994	5.331	3.46 × 10⁻⁴
	DEHA	0.00270	0.944	0.550	1.269	1.74 × 10⁻³
	PDMS50	0.00879	0.290	0.179	0.312	1.40 × 10⁻³
Acetone	Water	0.00094	2.721	0.956	4.254	2.03 × 10⁻³
	DEHA	0.00521	0.488	0.343	0.756	2.01 × 10⁻³
	PDMS50	0.02174	0.117	0.077	0.132	1.46 × 10⁻³

3.2. Data from Study n°2

The main objective of this study was to investigate the absorption of four hydrophobic compounds in pure DEHP (Table 2) using a cables-bundle contactor. Experiments were performed at ambient temperature for three liquid flow-rates. The K_La results obtained using the ε-NTU method are summarized in Table 4. It was clear that regardless of the liquid flow-rate, the same order of magnitude of the K_La was observed for all the investigated compounds, even though the results were slightly higher for hexane (Figure 6). For this solute, the operating conditions led to very low removal efficiencies relative to the low absorption factors. For other compounds (toluene, octane, and methylcyclohexane), the absorption factors ranged from 0.7 to 3.4. Nonetheless, moderated absorption efficiencies were recorded (Figure 3), and these were potentially linked to the viscosity of the DEHP, which explained the low K_La values. According to Figure 6, a slight increase in the K_La relative to the liquid flow-rate was suggested by the authors. The weak influence of the liquid flow-rate in the case of viscous fluid was also reported in the literature [11]. Although the K_La values calculated using the ε-NTU method could not be directly validated in the present case, from the K_La values given by the authors, we observed that these results were still in agreement with the results of Biard et al. [1]. For instance, the influence of the Henry coefficient on the K_La value was confirmed (Figure 7). Moreover, from the solute diffusivities in DEHP given by the authors (toluene: 5.51×10^{-11} m² s⁻¹; hexane: 1.16×10^{-10} m² s⁻¹; octane: 6.23×10^{-11} m² s⁻¹; methylcyclohexane: 4.24×10^{-11} m² s⁻¹), and knowing that the specific interfacial area determined by the authors was 129 m⁻¹, it was possible to calculate the thickness of the liquid film around the cables available for mass transfer (using the two-film theory) in each experiment. For the four solutes, all the calculated results were in good agreement. The thickness δ of the liquid film around the cables could be estimated from 30 to 56 μm (Table 4).

Table 4. K_La determination using the data reported in Bourgois et al. [6]. For all the experiments, solvent: DEHP (di-2-ethylhexyl phtalate); gas flow-rate = 1.39 × 10⁻² m³/s (gas EBRT = 1 s; V = 1.46 × 10⁻² m³).

Solute	H_{VOC} (-)	Q_L (m³/s)	A (-)	ε (-)	NTU (-)	K_La (s⁻¹)	δ (m)
Hexane	4.7 × 10⁻³	1.00 × 10⁻⁵	0.152	0.059	0.077	3.48 × 10⁻⁴	4.30 × 10⁻⁵
		1.25 × 10⁻⁵	0.190	0.068	0.087	3.94 × 10⁻⁴	3.80 × 10⁻⁵
		1.25 × 10⁻⁵	0.190	0.067	0.086	3.87 × 10⁻⁴	3.87 × 10⁻⁵
		1.50 × 10⁻⁵	0.228	0.051	0.059	2.67 × 10⁻⁴	5.59 × 10⁻⁵
		1.50 × 10⁻⁵	0.228	0.083	0.108	4.87 × 10⁻⁴	3.07 × 10⁻⁵

Table 4. *Cont.*

Solute	H_{VOC} (-)	Q_L (m³/s)	A (-)	ε (-)	NTU (-)	$K_L a$ (s⁻¹)	δ (m)
Toluene	3.2×10^{-4}	1.00×10^{-5}	2.257	0.345	0.462	1.40×10^{-4}	5.07×10^{-5}
		1.00×10^{-5}	2.257	0.335	0.444	1.35×10^{-4}	5.27×10^{-5}
		1.25×10^{-5}	2.821	0.346	0.455	1.38×10^{-4}	5.15×10^{-5}
		1.25×10^{-5}	2.821	0.351	0.464	1.41×10^{-4}	5.05×10^{-5}
		1.25×10^{-5}	2.821	0.35	0.462	1.40×10^{-4}	5.07×10^{-5}
		1.50×10^{-5}	3.386	0.367	0.486	1.48×10^{-4}	4.82×10^{-5}
		1.50×10^{-5}	3.386	0.386	0.520	1.58×10^{-4}	4.50×10^{-5}
		1.50×10^{-5}	3.386	0.386	0.520	1.58×10^{-4}	4.50×10^{-5}
Octane	4.4×10^{-4}	1.00×10^{-5}	1.636	0.272	0.349	1.46×10^{-4}	5.50×10^{-5}
		1.00×10^{-5}	1.636	0.284	0.369	1.54×10^{-4}	5.21×10^{-5}
		1.25×10^{-5}	2.045	0.304	0.394	1.65×10^{-4}	4.87×10^{-5}
		1.25×10^{-5}	2.045	0.286	0.364	1.53×10^{-4}	5.27×10^{-5}
		1.50×10^{-5}	2.455	0.298	0.379	1.58×10^{-4}	5.07×10^{-5}
		1.50×10^{-5}	2.455	0.298	0.379	1.58×10^{-4}	5.07×10^{-5}
Methylcyclo-hexane	9.8×10^{-4}	1.00×10^{-5}	0.735	0.144	0.174	1.62×10^{-4}	3.38×10^{-5}
		1.00×10^{-5}	0.735	0.131	0.155	1.44×10^{-4}	3.79×10^{-5}
		1.25×10^{-5}	0.919	0.158	0.189	1.76×10^{-4}	3.10×10^{-5}
		1.25×10^{-5}	0.919	0.143	0.168	1.57×10^{-4}	3.49×10^{-5}
		$1.25 \ 10^{-5}$	0.919	0.166	0.201	1.87×10^{-4}	2.92×10^{-5}
		1.50×10^{-5}	1.103	0.163	0.193	1.80×10^{-4}	3.04×10^{-5}
		1.50×10^{-5}	1.103	0.164	0.194	1.81×10^{-4}	3.02×10^{-5}
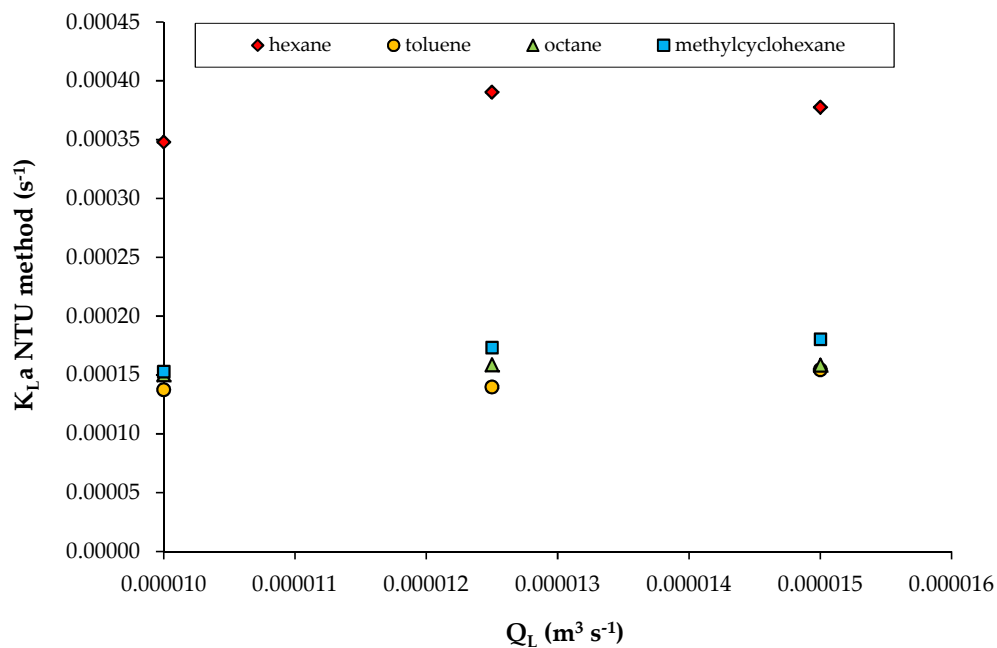
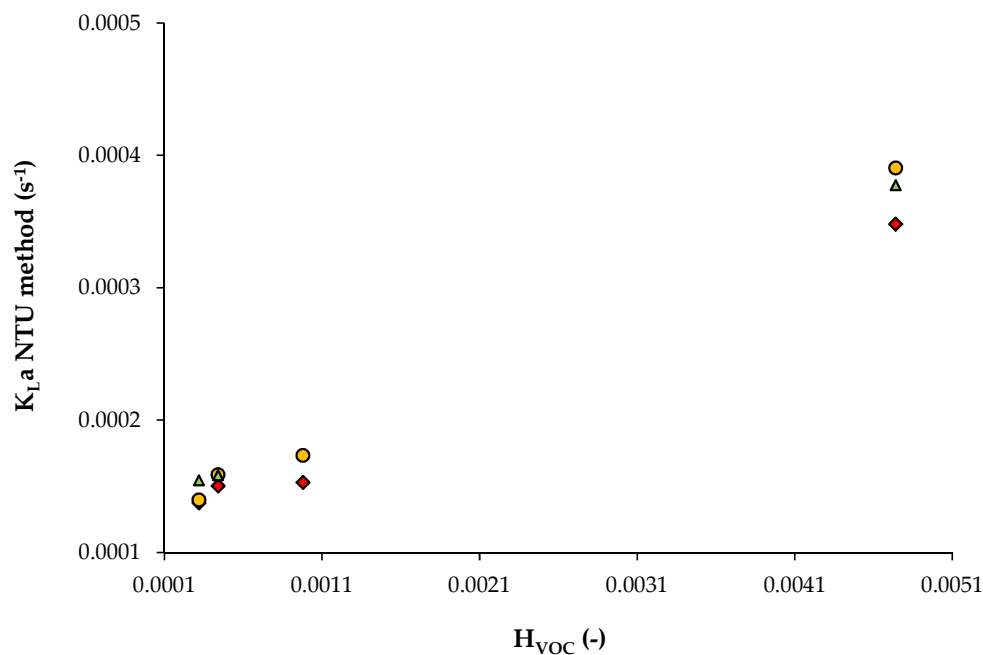**Figure 6.** $K_L a$ versus liquid flow-rate (from the data analysis reported in Reference [6]).

Figure 7. K_La versus Henry coefficient (from the data analysis reported in Reference [6]). Diamond: gas flow-rate $= 1.0 \times 10^{-5}$ m³/s; circle: gas flow-rate $= 1.25 \times 10^{-5}$ m³/s; square: gas flow-rate $= 1.50 \times 10^{-5}$ m³/s.

3.3. Data from Study n°3

The purpose of this study was to compare the toluene absorption using two silicone oils with different dynamic viscosities (5 mPa s and 50 mPa s, respectively) in a countercurrent gas-liquid absorber filled with three types of packing material (Raschig rings, IMTP®, and Flexipac®). The K_La values calculated using the ε-NTU method were compared to the values determined by the authors using the following equation reported in Heymes et al. [11].

$$K_La = \frac{Q_G}{V} \frac{(C_{Gin} - C_{Gout})}{\frac{(C_{Gin}/H_{VOC} - C_{Lout}) - (C_{Gout}/H_{VOC} - C_{Lin})}{\ln\left[\frac{(C_{Gin}/H_{VOC} - C_{Lout})}{(C_{Gout}/H_{VOC} - C_{Lin})}\right]}} \tag{20}$$

From Figure 8, it appears that the K_La parity plot is validated for the solvent with low dynamic viscosity (5 mPa s). Conversely, for the solvent with the high dynamic viscosity (50 mPa s), the K_La determined using the ε-NTU method was usually 30% higher than the values calculated using Equation (20). According to this equation, the K_La determination required the measurement of solute concentrations in the gas phase and in the liquid phase. As the concentration measurement of a solute in a viscous fluid could be relatively inaccurate, the K_La determination using Equation (20) should be considered cautiously. Consequently, the K_La determination using the ε-NTU method should be preferred, since only the inlet and outlet gaseous concentrations are needed. Results from this study also confirmed that the viscosity of the liquid, as well as the type of packing material filling the column, has a considerable impact on the volumetric mass transfer coefficient of the operating absorber (Figure 9).
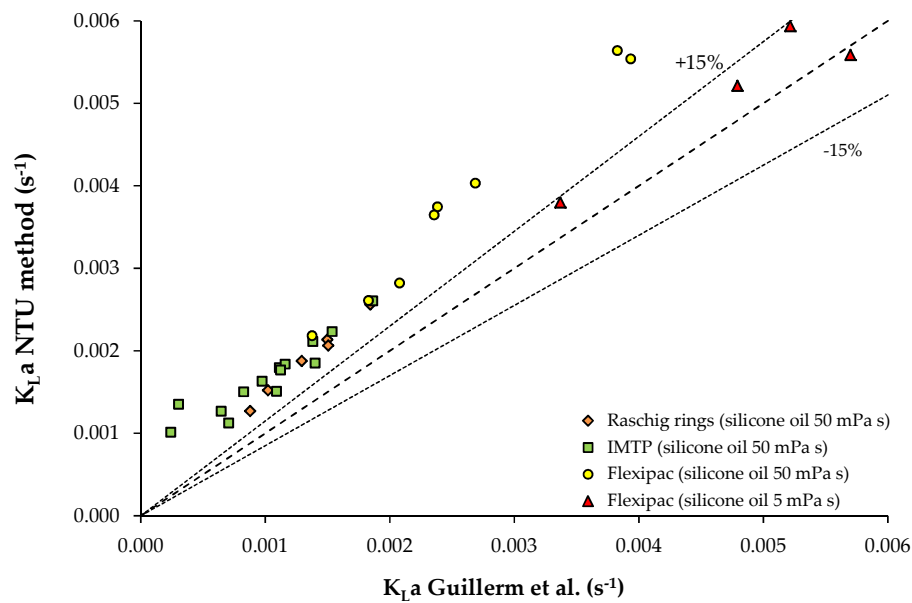
Figure 8. $K_L a$ parity plot obtained from the data analysis reported in Reference [10], ($V = 1.13 \times 10^{-2}$ m^3; $T = 25$ °C).
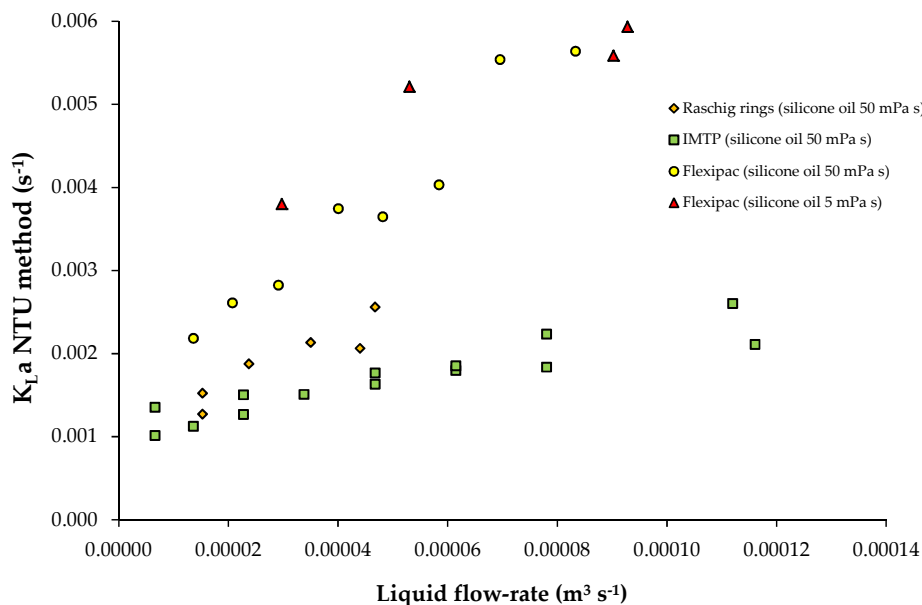

Figure 9. $K_L a$ versus the liquid flow-rate (calculated from Reference [10], ($V = 1.13 \times 10^{-2}$ m^3; $T = 25$ °C).

4. Conclusions

We adapted the ε-NTU method, which is usually applied to heat exchanger design, to gas–liquid countercurrent absorbers. We demonstrated that this method could be successfully adapted to determine the overall mass transfer coefficient of the absorber in operation, the $K_L a$, using advance knowledge of the physical parameters: H_{VOC}, Q_G, Q_L, V, C_{Gin}, and C_{Gout}. Therefore, the ε-NTU method may be a robust and reliable tool for the $K_L a$ determination of gas–liquid contactors used in environmental applications for treating the removal of air pollutants. Consequently, new empirical correlations for the prediction of $K_L a$ for heavy viscous solvents may be developed. Moreover, since this method only requires the measurement of solute concentrations in the gas phase, it should be efficient in determining the $K_L a$ values of complex systems, such as Two-Phase Partitioning bioreactors (TPPB) used to remove hydrophobic pollutants using mixtures of immiscible liquids. In such a case,

the main difficulty lies in the knowledge of the H_{VOC}. As a result, the ε-NTU method must be combined with the "equivalent absorption capacity" concept [12,13], thereby enabling calculation of the physical properties of the mixture [14].

Nomenclature

absorber

a	specific interfacial area (m^{-1})
A	absorption factor
C	concentration (mol m^{-3})
EBRT	Empty Bed Residence Time (s)
H	Henry coefficient
k_G, k_L	local mass transfer coefficients (m s^{-1})
$K_G a$, $K_L a$	overall mass transfer coefficients (s^{-1})
NTU	Number of Transfer Units
Q	flow-rate (m^3 s^{-1})
V	packing volume (m^3)
Z	packing height (m)

Greek letters

δ	film thickness (m)
ε	effectiveness
Ω	cross sectional area (m^2)
ϕ	molar flux (mol m^{-3} s^{-1})

Subscripts

G	gas
in	inlet
L	liquid
Max	maximum
out	outlet
VOC	Volatile Organic Compound

heat exchanger, Table 1

A	heat-transfer area (m^2)
C	heat capacity rate of the fluid = M C_p (W K^{-1})
C_p	specific heat capacity (J kg^{-1} K^{-1})
M	flow rate (kg s^{-1})
NTU	Number of Transfer Units
T	temperature (K)
U	heat transfer coefficient (W m^{-2} K^{-1})

Greek letters

ΔT_{ML}	log mean temperature difference (K)
ε	effectiveness
ϕ	power, heat transferred per unit time (W)

Subscripts

c	cold
h	hot
in	inlet
Max	maximum
Min	minimum
out	outlet

Appendix A   ε-NTU Relationship for a Gas-Liquid Absorber

Equations (8), (13), and (16) are rewritten:

$$C_{Lout} = C_{Lin} + \frac{Q_G}{Q_L}(C_{Gin} - C_{Gout}) \tag{A1}$$

$$\frac{(C_{Gout} - H_{VOC} C_{Lin})}{(C_{Gin} - H_{VOC} C_{Lout})} = \exp\left(-NTU\left(1 - \frac{1}{A}\right)\right) = \beta \tag{A2}$$

$$\varepsilon = \frac{\phi}{\phi_{max}} = \frac{(C_{Gin} - C_{Gout})}{(C_{Gin} - H_{VOC} C_{Lin})} \tag{A3}$$

Multiplying Equation (A1) by $(-H_{VOC})$ gives:

$$-H_{VOC} C_{Lout} = -H_{VOC} C_{Lin} - \frac{H_{VOC} Q_G}{Q_L}(C_{Gin} - C_{Gout}) \tag{A4}$$

$$-H_{VOC} C_{Lout} = -H_{VOC} C_{Lin} - \frac{1}{A}(C_{Gin} - C_{Gout}) \tag{A5}$$

Introducing Equation (A5) in Equation (A2):

$$\frac{(C_{Gout} - H_{VOC} C_{Lin})}{\left(C_{Gin} - H_{VOC} C_{Lin} - \frac{1}{A}(C_{Gin} - C_{Gout})\right)} = \beta \tag{A6}$$

Rearranging Equation (A6):

$$H_{VOC} C_{Lin} = C_{Gout}\left(\frac{\beta/A - 1}{\beta - 1}\right) + C_{Gin}\beta\left(\frac{1 - 1/A}{\beta - 1}\right) \tag{A7}$$

Introducing Equation (A7) in Equation (A3):

$$\varepsilon = \frac{(C_{Gin} - C_{Gout})(\beta - 1)}{(\beta - 1)C_{Gin} - [C_{Gout}(\beta/A - 1) + C_{Gin}\beta(1 - 1/A)]} \tag{A8}$$

Rearranging Equation (A8):

$$\varepsilon = \frac{(1 - \beta)(C_{Gout} - C_{Gin})}{(1 - \beta/A)(C_{Gout} - C_{Gin})} \tag{A9}$$

As $\beta = \exp(-NTU(1-1/A))$, Equation (A9) can be rewritten under the final form:

$$\varepsilon = \frac{1 - \exp\left(-NTU\left(\frac{A-1}{A}\right)\right)}{1 - \frac{1}{A}\exp\left(-NTU\left(\frac{A-1}{A}\right)\right)} \tag{A10}$$

Appendix B   Gas-Liquid Stirred Tank Reactor (STR)

In a gas–liquid stirred tank reactor (STR), a gas flow-rate, Q_L, continuously bubbles through a known volume, V, of the well-mixed liquid phase. The solute mass transfer from the gas to the liquid phase is expressed as:

$$\frac{dC_G}{dt} = -K_L a\left(\frac{C_G}{H_{VOC}} - C_L\right) \tag{A11}$$

During the mean residence time of the gas in the liquid phase, $t = V/Q_G$, the solute concentration in the rising gas bubbles changes from C_{Gin} to C_{Gout}, and consequently:

$$H_{VOC} \int_{C_{Gin}}^{C_{Gout}} \frac{dC_G}{(C_G - H_{VOC} C_L)} = -K_L a \int_{t=0}^{t=V/Q_G} dt \tag{A12}$$

$$\ln\left[\frac{(C_{Gout} - H_{VOC} C_L)}{(C_{Gin} - H_{VOC} C_L)}\right] = -\frac{K_L a}{H_{VOC}} \frac{V}{Q_G} \tag{A13}$$

Defining NTU = $(K_L a\, V)/(H_{VOC}\, Q_G)$, Equation (A13) becomes:

$$C_{Gout} \;=\; H_{VOC}\, C_L\, (1 - \exp(-NTU)) \;+\; C_{Gin}\, \exp(-NTU) \tag{A14}$$

Moreover, the molar flux ϕ of the transferred solute is:

$$\phi = \frac{Q_G}{V}(C_{Gin} - C_{Gout}) \tag{A15}$$

Introducing the expression of C_{Gout} (Equation (A14)) in Equation (A15) gives:

$$\phi = \frac{Q_G}{V}(1 - \exp(-NTU))(C_{Gin} - H_{VOC}\, C_L) \tag{A16}$$

where the term $(1-\exp(-NTU))$ is the solute fraction that can be transferred from the gas phase to the liquid phase. Basically, for industrial applications, the initial concentration of the liquid phase is $C_L = 0$. Consequently:

$$\phi = \frac{Q_G}{V}(1 - \exp(-NTU))\, C_{Gin} \tag{A17}$$

According to the NTU expression, the maximum solute fraction that can be transferred is obtained for a long residence time of the gas phase in the liquid phase (i.e., using a large liquid volume and a small gas flow-rate). In that case, the NTU is significantly higher than 1 and $\exp(-NTU)$ tends to 0. As a result:

$$\phi_{max} = \frac{Q_G}{V}\, C_{Gin} \tag{A18}$$

The effectiveness of the STR is then:

$$\varepsilon \;=\; \frac{\phi}{\phi_{max}} \;=\; 1 \,-\, \exp(-\,NTU\,) \tag{A19}$$

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