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## Mass transfer coefficient of ammonia in the air stripping process for municipal wastewater: An experimental study

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### ABSTRACT

This study evaluated the effects of different operating conditions and the air-to-water ratio (G/L) on the kinetics and the mass transfer coefficient of ammonia ( $K_L$ ) in the air stripping method for removing ammonium ions ( $\text{NH}_4^+$ ) from wastewater with low concentrations in municipal wastewater treatment plants (WWTPs). The impact of operating conditions including the temperature, initial ammonium ion concentration, pH, and air-to-water ratio (G/L) of <2000:1 (60:1, 70:1, and 80:1) on  $K_L$  in the air stripping method was investigated using artificial wastewater at laboratory scale. The  $\text{NH}_4^+$  concentrations in the wastewater samples were determined with the Nesslerization method (the standard method for the examination of water and wastewater). According to the results, the minimum ( $0.0528 \text{ h}^{-1}$ ) and maximum ( $0.64825 \text{ h}^{-1}$ ) of  $K_L$  were obtained within 1 to 4 h in the operating status that included an initial ammonium ion concentration of 33.63-52.81 mg/l, a temperature of 34-45.7 °C, a pH of 9.48-12.2, and an air-to-water ratio of 60:1-80:1. A comparison of the results of three regression models showed that the air-to-water ratio was the most effective factor on  $K_L$ . Furthermore, in Model 3 (multivariate linear regression model/comparing four parameters), the effects of the air-to-water ratio, pH, and temperature increased, leading to the acceleration and conversion of ammonium ions ( $\text{NH}_4^+$ ) to a gaseous form ( $\text{NH}_3$ ). Also, the initial  $\text{NH}_4^+$  concentration and pH in Model 4 (multivariate linear regression model by subgroup) at a low (60:1) and high (80:1) G/L ratio were the most influential factors on  $K_L$ , respectively. The results of this study revealed that the air-to-water ratio (60:1, 70:1, and 80:1) could be used successfully for the elimination of ammonium ions from municipal WWTPs, leading to lower energy costs for the required aeration in the air stripping method.

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## 1. Introduction

Ammonium ions ( $\text{NH}_4^+$ ) is an important pollutant in the wastewater of commercial fertilizers, chemical industrial plants' landfill leachate, agriculture, swine farms, and domestic wastewater [1]. Significant amounts of  $\text{NH}_4^+$  released into surface waters may cause oxygen depletion, eutrophication phenomenon, and toxicity of aquatic biota and human beings [2,3]. The US Environmental Protection Agency (EPA) has set the maximum permissible amount of ammonium released from municipal WWTPs into surface water to be a monthly average of 4.9 mg/L, as the total ammonia nitrogen (TAN) [4]. Many methods, such as biological processing, have been extensively utilized for nitrogen removal from sewage wastewaters [5-8]. Owing to the adverse impact of elevated levels of  $\text{NH}_4^+$  on the action of microorganisms and the high production of sludge in biological processes, the use of alternative processes such as the physico-chemical process should be considered [9]. The air stripping method is a physico-chemical process that has been successfully used for ammonium ions elimination [9-12]. It is a simple and effective physico-chemical method that does not produce extra sludge [13,14]. Currently, many studies have been performed for the elimination of ammonium ions in ammonia-rich wastewater using the air stripping method, including landfill leachate [15,16], anaerobic digestion of wastewater [17,18], and industrial wastewaters [19]. More than 75% of nitrogen compounds in municipal and sanitary wastewater are in the form of ammonium ions; conventional wastewater treatment in the secondary treatment process can remove up to 30% of nitrogen from wastewater. Nitrogen compounds need to be significantly removed from sewage to discharge the municipal treatment effluent into the receiving waters and reuse the effluent for various applications. The advanced wastewater treatment method uses the air stripping method as a physico-chemical process that can remove >90% of  $\text{NH}_4^+$  from wastewater [20]. The principal variables affecting the elimination efficiency and the mass transfer coefficient of ammonia ( $K_L$ ) in the air stripping method are pH, initial ammonium ion concentration, temperature, and air-to-water ratio (G/L) [21,22]. To increase the elimination

efficiency in the air stripping method, a counter-current packed tower is usually employed to increase the contact surface and the exposure of air and polluted water to promote mass transfer from the aqueous to the gaseous phase [23,24]. Numerous studies have advised an air-to-water ratio (G/L) of more than 2000:1 to eliminate  $\text{NH}_4^+$  from wastewater via the air stripping method [25,20,26], which can lead to increased energy consumption. The results of a study on the air stripping method showed that the maximum removal efficiency of ammonium ions with the air-to-water ratio of 3360 was 75% [27]. Bui et al. showed that at the air-to-water ratio of 2925 and a pH of 11, the removal efficiency and mass transfer coefficient of ammonia were more than 90% and 0.0125 l/s, respectively [28]. Due to the limited studies conducted regarding the effect of air-to-water ratios of less than 2000:1 in wastewater with low concentrations of ammonium ions in municipal WWTPs, this study evaluated the effects of different operating conditions and an air-to-water ratio (G/L) of <2000:1 (60:1, 70:1 and 80:1) on the kinetics and  $K_L$  in the air stripping method for removing ammonium ions from wastewater with low concentration in these plants. For this purpose, the impact of operating status (temperature, initial ammonium ion ( $\text{NH}_4^+$ ) concentration, pH, and the air-to-water ratio (G/L) (60:1, 70:1 and 80:1) on  $K_L$  was investigated by an artificial solution in a range of  $\text{NH}_4^+$  concentrations, which were the same as the concentrations of municipal WWTPs. To this end, besides comparing the impact of the operating status on  $K_L$ , the effects of each operating parameter in the air-to-water ratio (G/L) (60:1, 70:1 and 80:1) on  $K_L$  and for the elimination of ammonium ion by the air stripping method in WWTPs were investigated separately.

## 2. Materials and methods

### 2.1. Design of the tower strippers

In the stripper experiments, a Plexiglas column with a height and inner diameter of 1m×10 cm was employed. Figure 1 illustrates the pilot schematic specifications of the experimental scale. The features of the packed tower stripper and the pilot experiment layout for the air stripping of ammonium ions are presented in Table 1. This packing media provides a high surface area for

water-liquid contact and mass transfer coefficient of ammonia from the aqueous ( $\text{NH}_4^+$ ) to the gaseous phase ( $\text{NH}_3$ ). The choice of packing media type is based on the results of other studies that show that plastic packing has the highest removal efficiency for ammonium ions attributable to enhance in the surface area of liquid-air contact [23]. This study used a temperature control module (Model XH-W3001) based on the desired range to adjust the temperature. Moreover, in order to supply the air and water flow at the rate of 45 and 0.56 liters per minute (air-to-water ratio 80:1) an air pump (HAILEA Aq. Air pump, the maximum flow rate 45 L/minute, 22 w), and a water pump (the maximum flow rate 40 L/minute, 0.37 Kw) were used.

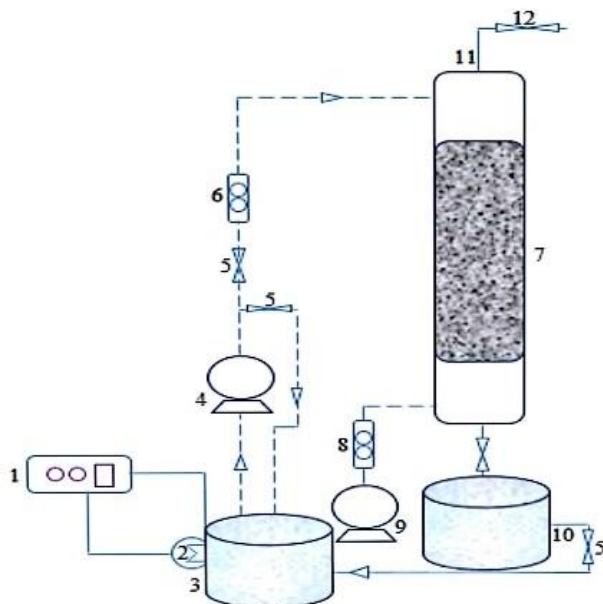


Fig. 1. A Schematic diagram of ammonia stripping pilot.

① temperature controller system; ② heating pack; ③ feed reservoir; ④ water pump; ⑤ water faucet; ⑥ water rotameter; ⑦ air stripping tower; ⑧ air rotameter; ⑨ air pump; ⑩ water recycle reservoir; ⑪ gas exhaust; ⑫ gas exhaust faucet.

Table 1. Specifications of the packed tower stripper and the pilot experiment layout for the ammonia stripping.

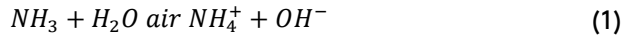
Parameter	Set A	Set B	Set C
Liquid flow- rate (L/min)	0.42	0.5	0.56
Air flow- rate (L/min)	25	35	45
Air-to-water ratio (G/L)	60	70	80
Packing media volume	4.71 x10 <sup>-3</sup> m <sup>3</sup>		
Packing media area	725 m <sup>2</sup> /m <sup>3</sup>		
Column diameter	0.1 m		
Height of packing	0.6 m		
Packing media type	Kaldness 3		

## 2.2. Material and process

In this study, to assess the mass transfer coefficient of ammonia in the air stripping method, the impacts of variables consisting of temperature, initial ammonia ions concentration, pH, and the air-to-water ratio (G/L) were investigated. The experiments were carried out in sets A, B, and C in operating times of 1-14 h(hours) for 48 days. To evaluate the efficacy of the initial ammonia ion concentration on mass transfer coefficient in the stripping method for municipal WWTPs, ammonium chloride salt ( $\text{NH}_4\text{Cl}$ ) and distilled water were used to produce artificial wastewater with a range of  $\text{NH}_4^+$  concentration similar to that of municipal WWTP. The examination was carried out at various temperature ranges ( $34.25 \pm 0.44$ ,  $38.57 \pm 3.4$ ,  $40.5 \pm 7.68$  °C) and pH values ( $9.7 \pm 0.26$ ,  $10.93 \pm 0.16$ ,  $11.94 \pm 0.32$ ). To adjust the pH, sodium hydroxide 6 and 25 Molar ( $\text{NaOH}$ ; Merck) were used. Sampling with a volume of 10 ml was performed at regular intervals to measure the ammonium ions and pH. The  $\text{NH}_4^+$  concentrations in the wastewater samples were determined via the Nesslerization method (the standard method for the examination of water and wastewater) with a spectrophotometer (HACH, DR 5000, Germany) at a 425 nm wavelength [29]. The pH was determined using a pH meter (WTW 720, Inolab, Germany). To reduce energy consumption costs for aeration, the air-to-water ratio ((G/L) of 60:1, 70:1, and 80:1) was chosen based upon other studies to compare with the higher air-to-water ratio [30]. The air-to-water ratio in three sets was adjusted by an air and water rotameter. A gas alert Extreme  $\text{NH}_3$  portable single-gas sensor (from BW Technologies) with a measurable range of 0-100 ppm and a resolution of 1 ppm was used for ammonia gas detection.

### 2.3. Calculations of the mass transfer coefficient of ammonia

The ammonia stripping method is based on the mass transfer of ammonia from the fluid phase ( $\text{NH}_4^+$ ) into the gas phase ( $\text{NH}_3$ ). These two shapes of ammonia are in thermodynamic equilibrium in accordance with Eq. (1):



The dispersion of the  $\text{NH}_4^+/\text{NH}_3$  is a function of the pH [31]. To appraise the impact of the pH, stripping temperature, and kinetics of ammonia removal, the values of  $\alpha_{\text{NH}_3}$  and  $pK_a$  can be obtained from Eqs. (2) and (3):

$$\alpha_{\text{NH}_3} = \frac{1}{1 + 10^{pK_a - \text{pH}}} \quad (2)$$

$$pK_a = 0.0897 + (2729/T) \quad (3)$$

where  $\alpha_{\text{NH}_3}$  is a fraction of free ammonia in total ammonia ( $\text{NH}_3 + \text{NH}_4^+$ ),  $K_a$  is the acid hydrolysis constant, and  $pK_a$  is  $-\log K$  (the negative logarithm of stoichiometric of acid hydrolysis constant of  $\text{NH}_4^+$ ) on the temperature scale,  $T$  (K) [32-34]. The release of the ammonia gaseous form ( $\text{NH}_3$ ) in the solution is a function of Henry's constant according to Eq. (4):

$$H_c = \frac{C_G}{C_L} \quad (4)$$

where  $H_c$  is Henry's constants,  $C_G$  is the concentration of ammonia in the gaseous phase ( $\text{NH}_3$ ) and  $C_L$  is the concentration of ammonia in the liquid phase ( $\text{NH}_4^+$ ), respectively [35].  $H_c$  is affected by temperature according to Van't Hoff's equation (Eq. 5):

$$\text{Log } H_c = \frac{-H^\circ}{RT} + k \quad (5)$$

where  $H^\circ$  is the enthalpy change resulting from the dissolution of the compound in water,  $R$  is the universal gas constant,  $T$  is the absolute temperature (K), and  $k$  is the compound dependent constant [34]. The stripper factor ( $S$ ) is the capacity to remove the contaminant during the stripper process that is dependent on Henry's constants and the air-to-water ratio (G/L) [23,36]. The stripper factor is calculated based on Eq. (6):

$$S = H_c \cdot \frac{G}{L} \quad (6)$$

where  $G$  and  $L$  are the volumetric flow rates of gas and liquid, respectively. The mass transport of ammonia from the fluid phase to the gas phase is affiliated with air supply rate, the area of fluid-gas interaction, and the concentration difference of  $\text{NH}_3$  gas in the air [34, 14]. According to the two-film model, the mass transport of ammonia from the fluid phase to the gas phase in the air stripping process can be calculated according to the equation presented by Matter-Mueller et al. (Eq. (7):

$$-Ln \frac{C_{L_t}}{C_{L_0}} = \frac{Q_G H_c}{V_L} \left[ 1 - \exp\left(\frac{-K_L a V_L}{H_c Q_G}\right) \right] t \quad (7)$$

where  $C_{L_t}$  and  $C_{L_0}$  are respectively the ammonia concentrations in the fluid phase at any time and the initial ammonia concentrations in the fluid phase in  $\text{gr}/\text{m}^3$ ,  $Q_G$  is the gas flow rate (l/h),  $H_c$  is the dimensionless Henry's constants,  $V_L$  is the total volume of the fluid (l),  $a$  is the interface area per unit volume of fluid ( $\text{m}^2/\text{m}^3$ ).  $K_L$  is the overall mass transfer coefficient of ammonia in  $\text{h}^{-1}$  [37, 24]. The mass transfer coefficient for ammonia removal can be obtained by first-order kinetics according to Eq. (8):

$$C_i = C_0 e^{-K_L t} \quad (8)$$

where  $C_i$  is the concentration of the eliminated compound (mg/l) at a given time(t),  $C_0$  is the initial concentration of the eliminated compound (mg/l),  $K_L$  is the removal rate constant, and  $t$  is the stripping time [34,38].

### 2.4. Analysis

The statistical and operational aspects of the air stripping system, including temperature, initial ammonium ions concentration, pH, and the air-to-water ratio (G/L), were analyzed (encompassing 590 data). To estimate the efficacy of variation in the kinetic and mass transfer of ammonia in the air stripping method and in order to statistically analyze the linear relationship and correlation coefficient of the data, their heat map diagram was first plotted with GraphPad Prism 8; then, for a more accurate analysis, four linear regression models consisting of Model 1 (univariate regression model), Model 2 (multivariate regression model/comparing two parameters), Model 3 (multivariate linear regression model/comparing four parameters), and Model 4 (multivariate linear

regression model by subgroup) were used. The subset analysis was carried out to estimate the best practical achievement of the system at various air-to-water ratios (G/L). In the linear regression models,  $K_L$  is the dependent variable, and the independent variables are temperature, initial  $\text{NH}_4^+$ , and pH. The amount of change in  $K_L$  for 1 standard deviation (SD) variation was reported in the independent variable based on B (Unstandardized Coefficients) and Beta (Standardized Coefficients). The error bar indicates the 95% confidence interval (CI). All the analyses were performed in STATA 14.

### 3. Results and discussion

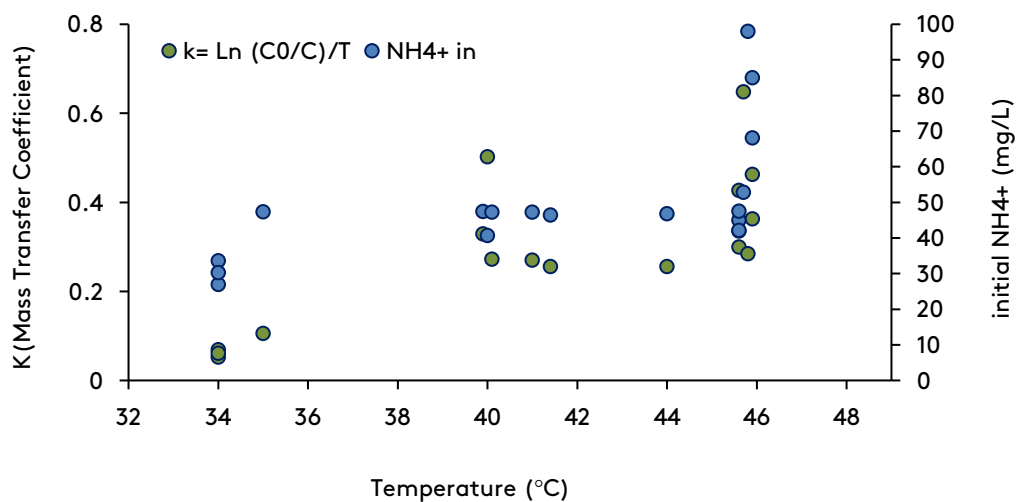
#### 3.1. Impact of temperature

Table 2 shows the impact of temperature changes on the acid hydrolysis constant ( $K_a$ ), the negative log ( $pK_a$ ) of  $\text{NH}_4^+/\text{NH}_3$  (aq) equilibrium, and Henry's constants for ammonia removal in the artificial solution by the air stripping process based on Eqs. (2) and (3). The results show that  $pK_a$  is a function of temperature and decreases with a rise in temperature based on Eq. (3). Furthermore, Henry's constants increase with raising the temperature due to the decrease in the dissolvability of the gaseous form ( $\text{NH}_3$ ) as the free-ammonia concentration, according to Eq. (5). This result clearly indicates that Henry's constants, which is an important index of a compound's potential for the elimination of ammonium ions in the air stripping method, increase with a rise in temperature [23,39]. The impact of temperature in three sets ( $34.25 \pm 0.44$ ,  $38.57 \pm 3.4$ ,  $40.5 \pm 7.68$  in  $^\circ\text{C}$ ) on the  $K_L$  at various initial  $\text{NH}_4^+$  concentrations (mg/l) was investigated. The maximum  $K_L$  ( $0.64825 \text{ h}^{-1}$ ) was obtained at  $45.7 \text{ }^\circ\text{C}$ , while the lowest  $K_L$  ( $0.0528 \text{ h}^{-1}$ ) was observed at  $34 \text{ }^\circ\text{C}$  (Figure 2). Furthermore, the result of the effect of temperature and pH on  $\alpha_{\text{NH}_3}$  (the fraction of free ammonia in total ammonia ( $\text{NH}_3 + \text{NH}_4^+$ )) based on Eq. (2) is given in Figure 3. And this result indicates that the increase in temperature and pH raises the  $\alpha_{\text{NH}_3}$  (the gaseous form ( $\text{NH}_3$ )). The heat map diagram (Figure 4) shows the correlation coefficient of the influential parameters (pH, temperature, and initial  $\text{NH}_4^+$ ) and mass transfer coefficient for ammonia removal in the artificial

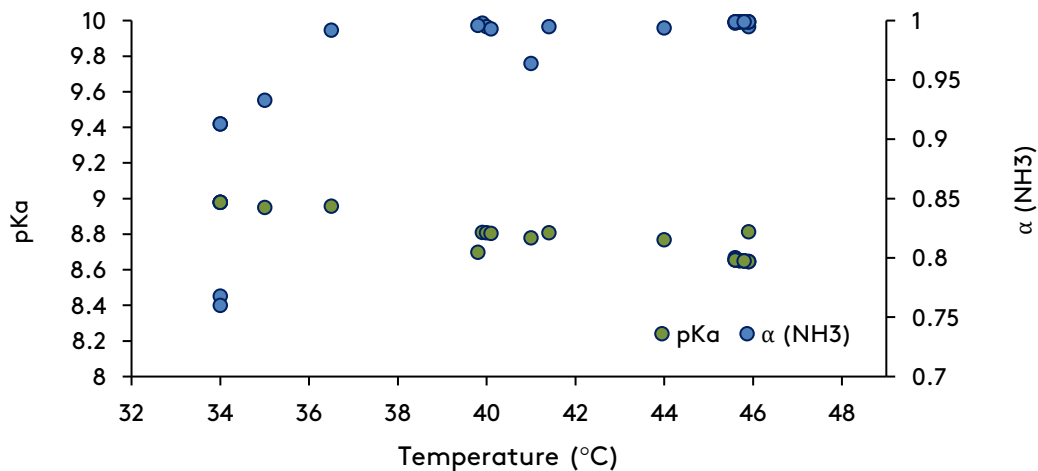
solution with the range of  $\text{NH}_4^+$  concentration, which are similar to those of a municipal WWTP by the air stripping process. The results of Model 1 showed that there was a significant definite correlation between temperature and  $K_L$  ( $B=0.004$ ; 95% CI: 0.002-0.005; Beta=0.175; P-value=0.000). This result indicated that with a 1 standard deviation enhancement in a unit of temperature,  $K_L$  increased 0.175%. Therefore, by increasing the operating temperatures, a better  $K_L$  was achieved due to a decrease in the solubility of the gaseous form ( $\text{NH}_3$ ) in the wastewater and an increase in Henry's constant with rising temperature, according to Eq. (5). This relationship in Eq. (5) expresses that the dissolvability of  $\text{NH}_3$  gas in air stripping is a function of operating temperature, and increasing temperature leads to a decrease in the dissolvability of  $\text{NH}_3$  gas by increasing Henry's coefficient. This result is similar to that of other studies that show a higher  $K_L$  in the air stripping method was obtained by enhancing the molecular dispersion in both liquid and gas films due to the temperature increase [40,41,32,33,3]. In a study by Zhu et al., the maximum of  $K_L$  was obtained as  $0.0146 \text{ min}^{-1}$  at  $60 \text{ }^\circ\text{C}$  [22]. In the study accomplished by Pouladi et al., the highest reaction rate was obtained at the highest temperature [42]. In Model 2, a comparison of the effect of the temperature and pH parameters on the mass transfer coefficient of ammonia was significant, and  $K_L$  increased with a rise in the unit of temperature (0.109%) and pH (0.454%) values (variation in  $K_L$  for 1 standard deviation variation in the independent variable). The results of this model showed that by comparing the concurrent effect of these two parameters on  $K_L$ , this effect increased. However, the impact of pH ( $B=0.099$ ; 95% CI: 0.083-0.114; P-value=0.000) was more significant than temperature ( $B=0.002$ ; 95% CI: 0.001-0.004; P-value=0.003) for the mass transfer of ammonia from the solution. The results showed a higher mass transfer coefficient of ammonia was obtained at higher temperatures and pH values. The increase in pH and temperature led to an increase in the fraction of free ammonia (gaseous form ( $\text{NH}_3$ )) in total ammonia ( $\text{NH}_4^+/\text{NH}_3$  (aq)) and a decrease in the dissolvability of  $\text{NH}_3$  gas in the solution. These results are consistent with other similar studies [33].

**Table 2.** Impact of temperature variation on pKa, Ka, and Henry’s constants (Hc).

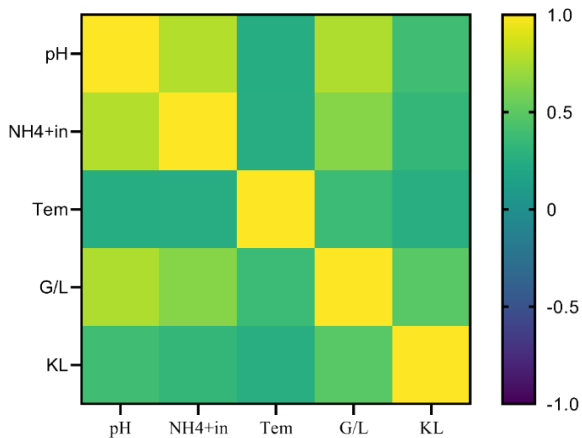
Temperature (°C)	pKa	Ka	Hc
34	8.97	$1.04966085 \times 10^{-9}$	$2.31309466 \times 10^{-5}$
35	8.95	$1.12178596 \times 10^{-9}$	$2.62800711 \times 10^{-5}$
40	8.8	$1.55399637 \times 10^{-9}$	$6.62067383 \times 10^{-5}$
45.9	8.64	$2.25298344 \times 10^{-9}$	$1.20157415 \times 10^{-4}$



**Fig. 2.** Impact of temperature and initial ammonium ions concentration on  $K_L$ .



**Fig. 3.** Impact of temperature and pH on  $\alpha_{NH_3}$ .



**Fig. 4.** Heat map diagram of the influential parameter on  $K_L$ .

### 3.2. Impact of initial ammonia concentration and pH

The efficacy of pH ( $9.7 \pm 0.26$ ,  $10.93 \pm 0.16$ ,  $11.94 \pm 0.32$ ) and the initial ammonia ion ( $\text{NH}_4^+$ ) concentration (26.98–47.34, 19.49–47.48, and 41–98 in mg/l) on  $K_L$  in the air stripping method were investigated. These results revealed that the minimum  $K_L$  for  $\text{NH}_4^+$  concentration (inlet=33.63 mg/l, outlet=31.9 mg/l) at a pH of 9.48 was  $0.0528 \text{ h}^{-1}$ , and the maximum  $K_L$  for an initial  $\text{NH}_4^+$  concentration pH of 12.2 (inlet =52.81 mg/l, outlet =3.95 mg/l) was  $0.64825 \text{ h}^{-1}$ . The impact of ammonium ion ( $\text{NH}_4^+$ ) concentration and pH on  $K_L$  is illustrated in Figure 5. These results indicate that the maximum of  $K_L$  ( $0.10569$ ,  $0.50284$  and  $0.64825 \text{ h}^{-1}$ ) is obtained at a pH of 10.1, 11.13, and 12.2, and a further increase in pH (12.2) does not change the mass transfer constant. The results of the univariate regression model showed that increasing pH raised  $K_L$ , and a significant definite correlation was observed between pH and  $K_L$  ( $B=0.102$ ; 95% CI: 0.087–0.118; Beta=0.47; P-value=0.000). This result indicated that with a 1 standard deviation increase in a unit of pH,  $K_L$  increased by 0.47%. Many studies have reported that pH greatly affects the conversion of the fluid phase ( $\text{NH}_4^+$ ) into the gaseous form ( $\text{NH}_3$ ); also, the transfer of the chemical reaction acid-base balance to the left side, according to Eq. (1), leads to the driving force of  $K_L$  and ammonia elimination efficiency [39,35,32,33]. Moreover, many studies reported that at higher pH (around 11–12), the stripper

process has higher efficiency for ammonia removal [11]. In a study conducted by Zhu et al.,  $K_L$  increased from  $0.0014$  to  $0.0040 \text{ min}^{-1}$  with a rise in pH from 10 to 12 [22]. The plotting of  $-\ln(C/C_0)$  versus stripping time ( $t$ ) showed a good relative coefficient between  $-\ln(C/C_0)$  and the stripping time (h) ( $R^2 = 0.97$ ) (Figure 6). The maximum of  $K_L$  ( $0.64825 \text{ h}^{-1}$ ) was obtained at a 4 h contact time with an initial  $\text{NH}_4^+$  concentration (52.81 mg/l), and increasing the concentration above this amount (from 53.55 to 98 mg/l) did not increase  $K_L$  (varied from  $0.38866$  to  $0.28472 \text{ h}^{-1}$ ) (Figure 7). These results were in compliance with those of another study, showing that by raising the ammonium concentration from 75 to 500 mg/l,  $K_L$  varied from  $0.0858$  to  $0.0804 \text{ h}^{-1}$ , and the mass transfer coefficient was not significantly affected by increasing the initial  $\text{NH}_4^+$  concentration [41]. Thus, it can express that the elimination performance in the air stripping method is dominated by dissemination through a gaseous film that is in theory dependent on the concentration of the volatile compound in solution [41,24]. The results of Model 1 showed that there was a significant relationship between the initial  $\text{NH}_4^+$  and  $K_L$  ( $B=0.002$ ; 95% CI: 0.001–0.002; Beta=0.236; P-value=0.000). This result indicated that with a 1 standard deviation enhancement in a unit of initial ammonium ion concentration,  $K_L$  increased by 0.236%. In Model 2, the linear regression statistical analysis between pH and the initial concentration of ammonia (multivariate regression model) indicated that the stripping pH ( $B=0.0.129$ ; 95% CI: 0.107–0.150; P-value=0.000) and the initial concentration of ammonia ( $B=-0.001$ ; 95% CI: 0.002–(-0.001); P-value=0.000) were two factors affecting the mass transfer constant; this effect increased for pH and decreased for the initial concentration of ammonia. Thus,  $K_L$  increased with a rise per unit of pH (0.592%) and decreased with an increase per unit of the initial ammonium ion concentration (-0.175%) (a variation in  $K_L$  for 1 standard deviation variation in the independent variable). Consequently, the pH had the greatest impact on the transformation of ammonium ions ( $\text{NH}_4^+$ ) to ammonia gas ( $\text{NH}_3$ ) in accordance with the ammonium ion ( $\text{NH}_4^+$ )/ ammonia gas ( $\text{NH}_3$ ) equilibrium in Eq. (1).

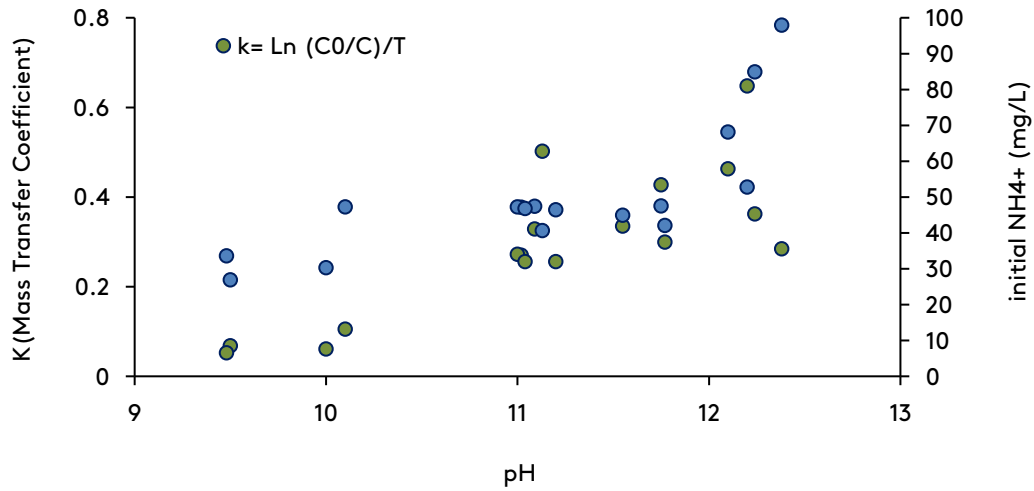


Fig. 5. Impact of initial ammonium ion concentration and pH on  $K_L$ .

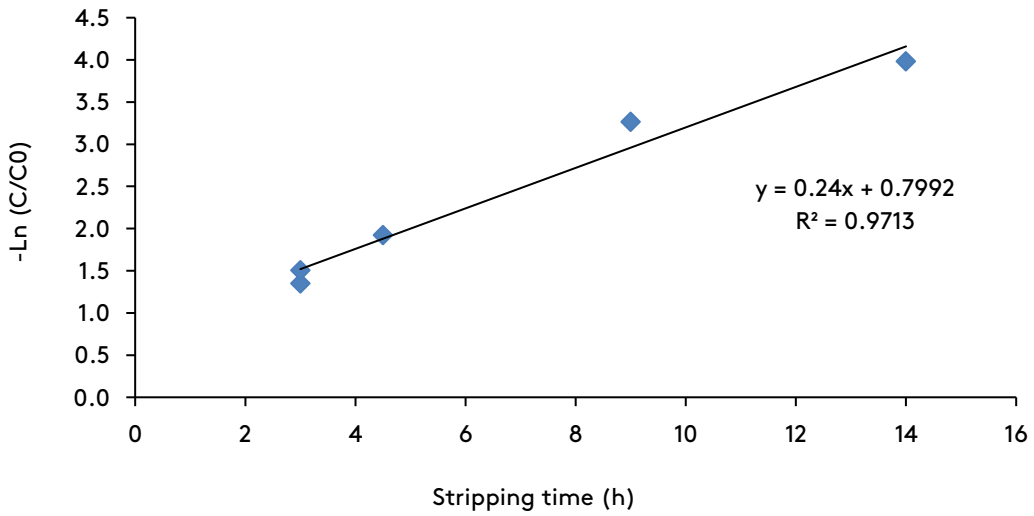


Fig. 6. Rate of  $-\ln (C/C_0)$  versus stripping time.

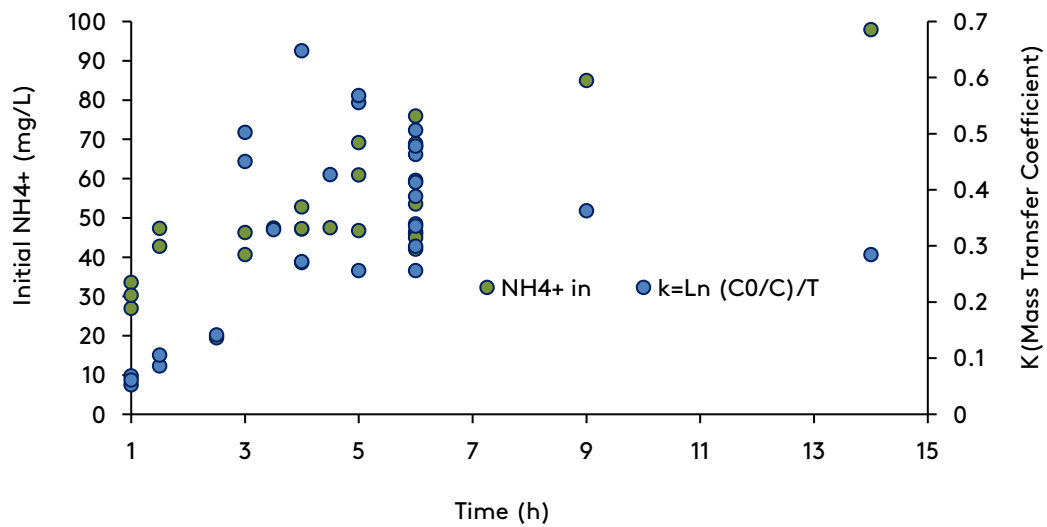


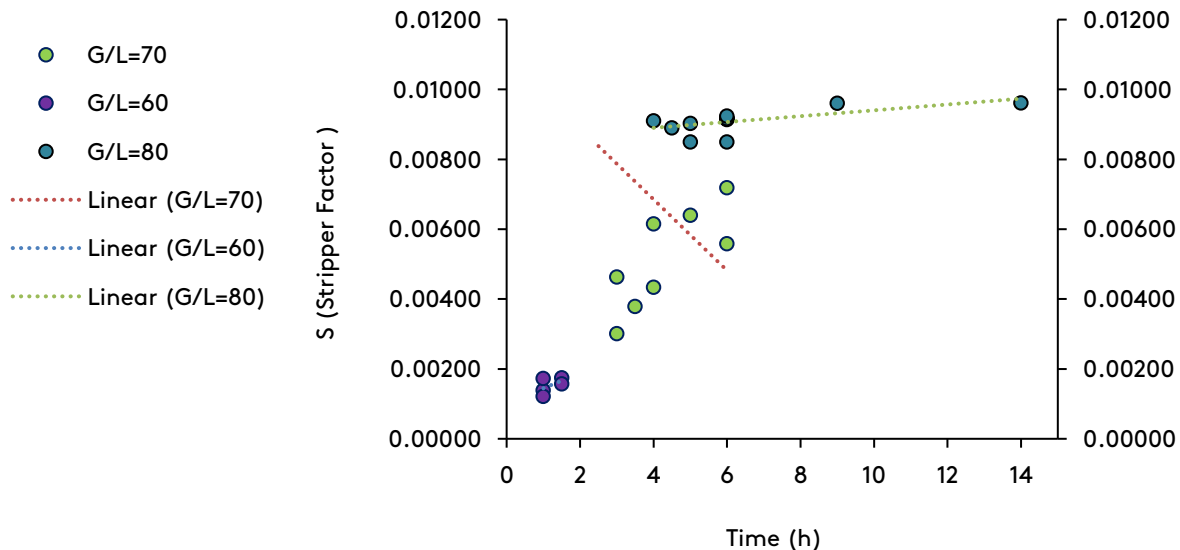
Fig. 7. Impact of initial ammonium ions concentration on  $K_L$ .



### 3.3. Impact of airflow

The air-to-water ratio was a significant and influential parameter on the stripper factor and  $K_L$ . The experimental values of the air to the water ratio (G/L) (60:1, 70:1, and 80:1) in this study were selected in accordance with other studies regarding the air stripping method [30]. Figure 8 reveals that increasing the air-to-water ratio from 60:1 to 80:1 increases the stripper factor and ammonium ion removal capacity of the artificial wastewater according to Eq. (6). The results also indicate that increasing the G/L ratio (60:1, 70:1, and 80:1) raises the mass transfer of ammonia ( $0.10569$ ,  $0.50284$ , and  $0.64825 \text{ h}^{-1}$ ) in the artificial solution at operation times of 1.5, 3.5, and 4 h, respectively (Figure 9). It clearly shows that the

mass transfer constant decreases by increasing time, according to Eq. (8). The results of Model 1 showed that there was a significant definite correlation between the G/L ratio and  $K_L$  ( $B=0.013$ ; 95% CI: 0.011-0.015; Beta=0.509; P-value=0.000). This result indicated that with a 1 standard deviation increase in a unit of G/L ratio,  $K_L$  increased by 0.509%. This could be due to the fact that increasing the G/L ratio accelerated the mass transfer of ammonia from the fluid phase ( $\text{NH}_4^+$ ) into the gas phase ( $\text{NH}_3$ ) according to the two-film model by raising the interfacial area and decreasing the gas phase resistance; many studies have reported that an upper air-to-water ratio (G/L) reduces mass transfer endurance and promotes the efficiency of stripping of ammonia [35,43,22].



**Fig. 8.** Impact of the air-to-water ratio (G/L) on the stripper factor (S).

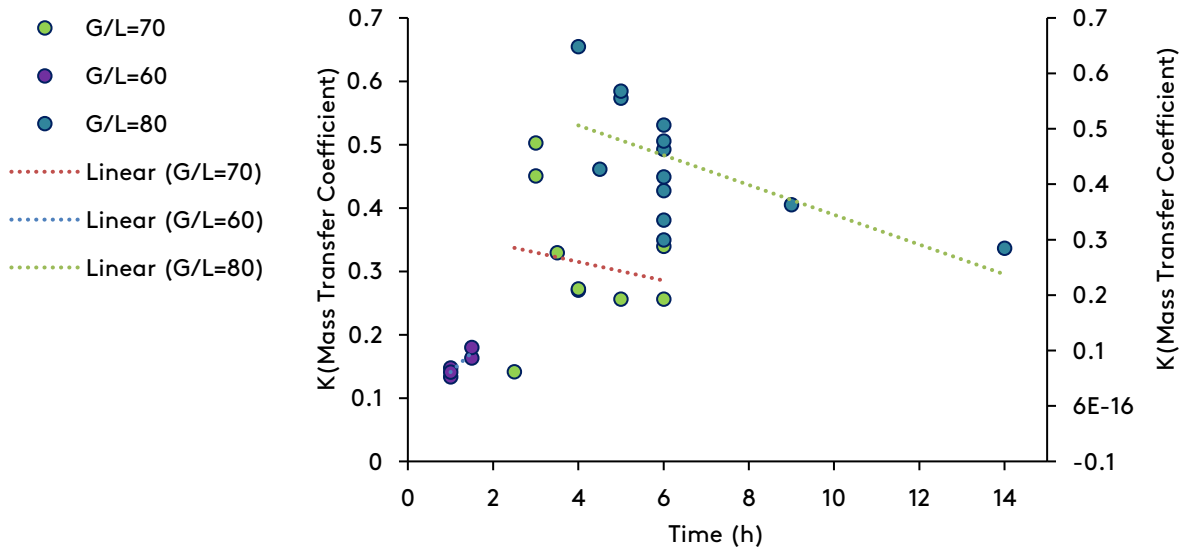


Fig. 9. Impact of the air-to-water ratio (G/L) on  $K_L$ .

In accordance with the results of the effects of air-to-water ratio (G/L) on  $K_L$  (Figure 9.), the maximum mass transfer coefficient ( $0.64825 \text{ h}^{-1}$ ) was achieved at four hours; then, by increasing time to 14 h, the constant rate of mass transfer decreased, indicating a decrease in the mass transfer coefficient by increasing time, according to Eq. (8). The reason could be that in the first four hours, the  $K_L$  value increased and in the subsequent hours, the  $K_L$  value decreased as the maximum of  $K_L$  ( $0.64825 \text{ h}^{-1}$ ) was obtained at the 4 hr contact time with the initial  $\text{NH}_4^+$  concentration ( $52.81 \text{ mg/l}$ ). These results indicated that increasing the initial  $\text{NH}_4^+$  concentration and stripper time will not lead to an increase in  $K_L$ . These results are similar to those of another study reporting that the mass transfer coefficient of ammonia decreases with raising time to 24 h [44]. In Model 2, the linear regression comparing the effect of the air-to-water ratio (G/L) and temperature parameters on  $K_L$  was significant; therefore,  $K_L$  increased with a rise in the unit of the G/L ratio (0.494%) and temperature (0.079%) values (a variation in  $K_L$  for 1 standard deviation variation in the independent variable). However, the effect of the G/L ratio ( $B=0.013$ ; 95% CI: 0.011-0.015;  $P$ -value=0.000) was more

significant than temperature ( $B=0.002$ ; 95% CI: 0.000-0.003;  $P$ -value=0.03) for the mass transfer of ammonia from the solution. This result indicated that the combined effect of high temperature and the G/L promoted  $K_L$  and, consequently, the elimination performance in the air stripping process that is in line with other studies [45,46]. Moreover, in Model 2, where there is the comparison of the effect of parameters of the G/L ratio ( $B=0.011$ ; 95% CI: 0.007-0.015;  $P$ -value=0.000) and pH ( $B=0.021$ ; 95% CI: 0.011-0.053;  $P$ -value=0.19) on the mass transfer coefficient of ammonia, a significant definite correlation was obtained between the G/L and  $K_L$ ;  $K_L$  increased by a rise in the unit of the G/L ratio (0.423%) (a variation in  $K_L$  for 1 SD variation in the independent variable), while there was no significant relationship between pH and  $K_L$ . In Model 3, the impact of influential parameters (pH, temperature, initial ammonium ion concentration, and G/L ratio) on  $K_L$  was investigated. Table 3 displays the variation in  $K_L$  per 1 SD increase or decrease in the unit of pH, temperature, initial  $\text{NH}_4^+$  concentration, and G/L ratio in the unadjusted univariate and multivariate regression model. The error bar indicates the 95% confidence interval.

**Table 3.** Variation in  $K_L$  per 1 standard deviation (SD) increase or decrease in a unit of pH, temperature, and initial ammonia ion ( $\text{NH}_4^+$ ) concentration.

	pH	Initial concentration (mg/l)	Temperature (°C)	G/L ratio
	Variations (95% CI) √/	Variations (95% CI)	Variations (95% CI)	Variations (95% CI)
Model 1	0.102 (0.087-0.118)	0.002 (0.001-0.002)	0.004 (0.002-0.005)	0.013 (0.011-0.015)
Model 3	0.048 (0.012-0.084)	-0.001 (-0.002-0.000)	0.001 (0.000-0.003)	0.01 (0.006-0.014)

Model 1: Unadjusted univariate regression model.

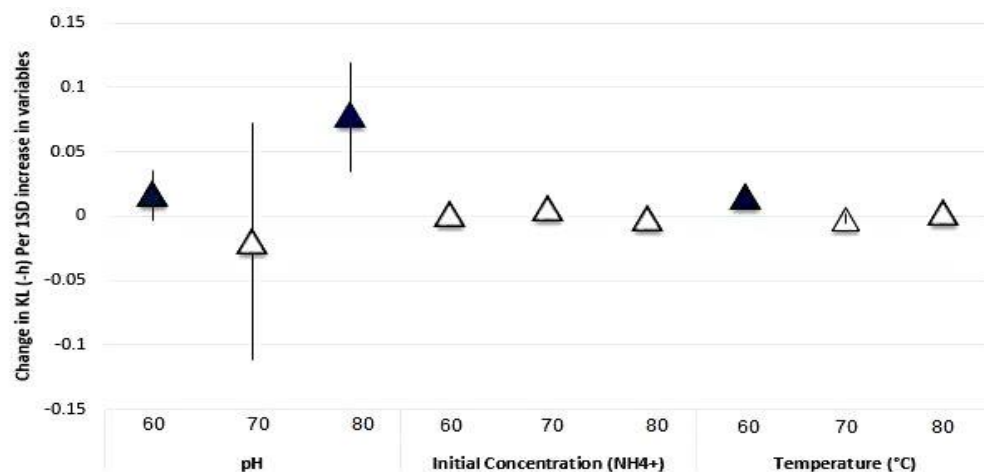
Model 3: Unadjusted multivariate regression model.

In this study, the statistical analysis results showed there were significant relationships between  $K_L$  and the temperature, pH, initial ammonium ion, and G/L ratio. Likewise, four linear regression models were used to predict and show changes in  $K_L$  based on changes in variables and the mentioned operating conditions, including temperature, initial ammonium ion concentration and G/L ratio, 4 linear regression models were used. The beta coefficients in Model 1 (adjusted univariate regression model) showed that with 1 standard deviation increase per unit of the pH, initial ammonium ion ( $\text{NH}_4^+$ ), temperature, and the G/L ratio, the  $K_L$  increased by 0.47%, 0.236%, 0.175%, and 0.509%, respectively. These results indicated that in the univariate regression model and evaluation of the effect of individual variables on  $K_L$ , the most effective operating conditions were the air-to-water ratio, pH, initial ammonium ion concentration, and temperature. In Model 2, the results showed that in the concurrent comparison of the air-to-water ratio with temperature, pH, and initial ammonium ion concentration, the most effective factor was the G/L ratio. Besides, the beta coefficients in Model 3 (adjusted multivariate linear regression model) that compared four parameters showed that  $K_L$  with a 1 standard deviation increase per unit of pH, temperature, and the G/L ratio increased by 0.221%, 0.071%, and 0.38%, respectively, and the initial ammonium ion decreased by 0.138%. And this led to the increasing effect of pH on  $K_L$  due to the increased conversion of ammonium ion ( $\text{NH}_4^+$ ) to gaseous form ( $\text{NH}_3$ ) in equilibrium ( $(\text{NH}_4^+) / (\text{NH}_3(\text{aq}))$ ). The reason for the increasing effect of temperature on the increase in  $K_L$  was due to the decrease in the solubility of ammonia gas in solution as a result of the increase in Henry's constants by increasing the temperature. Also, the increasing effect of the air-

to-water ratio was due to the effect of the G/L ratio in accelerating the transfer of ammonium ions from the soluble phase to the gas phase, according to the two-layer model. The effect of initial ammonium ion on  $K_L$  in comparison with the simultaneous effect of the four factors was not incremental, according to the results presented in this study. The results of Model 3 showed that in simultaneously comparing the four factors and operating conditions affecting  $K_L$ , the most influential parameters influencing  $K_L$  were the G/L ratio, pH, temperature, and initial ammonium ion. A comparison of the results of Models 1, 2, and 3 showed that the air-to-water ratio was the most effective factor on  $K_L$ , which indicates the importance of G/L ratios in accelerating the mass transfer of ammonia from the liquid phase to the gas phase. Figure 10 depicts the variation in  $K_L$  with a 1 standard deviation increase in a unit of pH, initial ammonium ion ( $\text{NH}_4^+$ ) concentration, and temperature at various G/L ratios. The results are related to Model 4 (an unadjusted multivariate regression model by subgroups (various air-to-water ratios (60:1, 70:1, and 80:1)), and the error bar indicates a 95% confidence interval. The P-values for pH, initial ammonium ion concentration, and temperature in this model are respectively 60 (0.086, 0.011, 0.004), 70 (0.665, 0.000, 0.396), and 80 (0.000, 0.000, 0.032). Based on the results, no significant relationships are found between pH and  $K_L$  in the G/L ratio 60 and between the pH, temperature, and  $K_L$  in the G/L ratio 70. Moreover, the beta coefficients (an adjusted multivariate regression model) for pH, initial  $\text{NH}_4^+$  concentration, and temperature in Model 4 are 60 (0.259%, 0.451%, 0.344%), 70 (-0.032%, 0.549%, -0.063%), and 80 (0.205%, -0.243%, 0.101%), respectively. The results of this model show that in the air-to-water ratio of 60 and 80, the initial  $\text{NH}_4^+$

concentration and pH are the most effective factors on the mass transfer constant. As mentioned earlier, many studies have reported the air-to-water ratio required to remove ammonium ions by the air stripping process as >2000:1. In the air stripping method, increasing the air-to-water ratio makes it necessary to use air and water blowers and pumps with a higher power, increasing energy consumption costs. The results of this study

on the impact of the air-to-water ratio (G/L) (60:1, 70:1, and 80:1) on the kinetics and  $K_L$  revealed that air-to-water ratio (G/L) of <2000:1 (60:1, 70:1, and 80:1) can be used successfully for the elimination of ammonium ions via the air stripping method in municipal WWTPs. Thus, the energy costs required to supply aeration flow are reduced compared to other studies with a higher air-to-water ratio of >2000:1.



**Fig. 10.** Variation in  $K_L$  per 1 standard deviation increase or decrease in a unit of pH, initial ammonium ion concentration, and temperature various air-to-water ratio (60:1, 70:1, and 80:1). The results are an unadjusted multivariate regression model. Error bar indicates 95% CI.

Table 4 displays a comparison of the current study and other studies on  $K_L$  in wastewater with a low

concentration of ammonia by the air stripping method.

**Table 4.** Comparing the  $K_L$  in wastewater with a low concentration of ammonia by air stripping method.

Wastewater Type	Ammonia concentration (mg/l)	$K_L$ ( $h^{-1}$ )	Temperature ( $^{\circ}C$ )	(G/L) ratio	pH	time (h)	Reference
Synthetic Wastewater- Packed bed	33.63 -47.34	0.0528- 0.10569	34.25±0.44	60	9.7±0.26	1-1.5	This work
	19.49 -40.68	0.14188- 0.50284	38.57±3.4	70	10.93±0.16	2.5-3	
	52.81-98	0.64825- 0.28472	40.5±7.68	80	11.94±0.32	4-14	
Synthetic Wastewater- Jet loop reactor	100-500	0.588	50	*0.13	11	13	[41]
		0.288	40	0.13	11	21	
Synthetic Wastewater- Jet loop reactor	100-500	0.3055-09613	20-50	2250	11	2.5-7.5	[47]
Acetylene purification	125±2	0.876	60	*0.5	12	2	[22]
wastewater- Packed bed	125±2	0.75	50	0.5	12	3	
	125±2	0.504	40	0.5	12	5	

\*  $Qa/V$  [ $m^3 / (h \cdot L)$ ];  $Qa$ : Airflow rate;  $V$ : Solution volume.

#### 4. Conclusions

The results of this study revealed that  $pK_a$  and Henry's constants were a function of temperature and the decreasing and increasing with a rise in temperature, respectively. Furthermore, the stripper factor increased with the air-to-water ratio (G/L). According to the results, the minimum ( $0.0528 \text{ h}^{-1}$ ) and maximum ( $0.64825 \text{ h}^{-1}$ ) of  $K_L$  for wastewater with a range of  $\text{NH}_4^+$  concentration, like that of a municipal WWTP in the air stripping method, was obtained in the operating status (initial ammonium ion concentration of  $33.63 - 52.81 \text{ mg/l}$ , pH of  $9.48 - 12.2$ , temperature of  $34 - 45.7 \text{ }^\circ\text{C}$ , and G/L ratio of  $60:1 - 80:1$ ) within 1 to 4 h. The results of the statistical analysis showed there were significant relationships between  $K_L$  and the temperature, pH, initial ammonium ions ( $\text{NH}_4^+$ ), and G/L ratio. In addition, the results of the adjusted linear regression based on variation in  $K_L$  per 1 standard deviation change in the independent variable in Models 1, 2, 3, and 4 are as follows:

1. Model 1: (G/L ratio > pH > initial  $\text{NH}_4^+$  concentration > Temperature)
2. Model 2: (pH, Temperature: pH > Temperature), (G/L ratio, pH: G/L ratio > pH), (G/L ratio, Temperature: G/L ratio > Temperature), and (pH, initial  $\text{NH}_4^+$  concentration: pH > initial  $\text{NH}_4^+$  concentration).
3. Model 3: (G/L ratio > pH > Temperature > initial  $\text{NH}_4^+$  concentration).
4. Model 4: 60 (initial  $\text{NH}_4^+$  concentration > Temperature > pH), 70 (initial  $\text{NH}_4^+$  concentration > Temperature > pH), and 80 (pH > Temperature > initial  $\text{NH}_4^+$  concentration).

A comparison of Models 1, 2, and 3 showed that the air-to-water ratio was the most effective factor on  $K_L$ . Furthermore, the effects of the air-to-water ratio, pH, and temperature increased in Model 3, leading to the acceleration and conversion of ammonium ions ( $\text{NH}_4^+$ ) to gaseous form ( $\text{NH}_3$ ). However, the effect of the initial  $\text{NH}_4^+$  concentration decreased. In regression Model 4, the initial  $\text{NH}_4^+$  concentration and pH at a low (60:1) and high (80:1) G/L ratio were the most influential factor on  $K_L$ , respectively. The results of this study revealed that the air-to-water ratio (60:1, 70:1, and 80:1) could be used successfully to eliminate ammonium ions from municipal WWTPs,

leading to lower energy costs for the required aeration in the air stripping method.

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