

บทที่ 4

MASS TRANSFER COEFFICIENT FOR MEK VOLATILIZATION FROM WASTEWATER

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ABSTRACT

Methyl Ethyl Ketone (MEK) is used in many industries including pharmaceutical, plastics, paints, and lubricants. The wide spread used of MEK has resulted in their appearance in industrial wastewater which consequently emitted to atmosphere via volatilization process. The monitoring of MEK released from wastewater to atmosphere is then necessary. The aim of this research was to investigate the effect of the wind speed on mass transfer process of MEK from wastewater. The volatilizations of MEK from wastewater and from pure water were investigated in pilot scale volatilization tank, 50 cm wide, 100 cm long and 20 cm deep. The wind speeds were varied from 0 to 4.42 m/s. The water temperature, the air temperature and the relative humidity were monitored for all experiments and they were 29 ± 1 °C, 31 ± 1 °C and 70-80%, respectively. The BOD₅, COD and SS of wastewater used in this study were 45-98 mg/L, 2320-2960 mg/L and 27-62 mg/L, respectively. The overall mass transfer coefficient, $K_{OL}a$, values determined under a range of conditions were then used to evaluate the effect of the wind speed on the mass transfer process. The results showed that the relationship between $K_{OL}a$ and the wind speeds falls into two regimes with a break at the wind speed of 2.4 m/s. This corresponds to the Reynolds number of 1.4×10^5 , which approximately characterized the change of airflow from laminar to turbulence. In laminar regime, $K_{OL}a$ was slightly increased with increasing Reynolds number and the linear relationship between $K_{OL}a$ and the Reynolds number was observed. In turbulence regime, $K_{OL}a$ increased more rapidly. The relationship between $K_{OL}a$ and the Reynolds number was also linear but has a distinctly higher slope. Moreover, $K_{OL}a$ obtained from MEK volatilization from wastewater was much lower than that obtained from pure water, particularly in turbulence regime. Based on this work, we can conclude that the wind speed and wastewater characteristic (i.e., uncontrolled organic masses) are the important parameters in controlling the volatilization of MEK from wastewater. Finally, the $K_{OL}a$ based on wastewater experiments were used to develop the $K_{OL}a$ correlation. The use of this correlation for predicting of MEK emission rate from wastewater was discussed.

Keyword: MEK, Mass Transfer Coefficient, Volatilization, Emission

1. INTRODUCTION

Methyl Ethyl Ketone (MEK) is present in municipal wastewater due to discharges from domestics, commercial and industrial sources. A major concern on toxic VOCs in municipal wastewater is their potential air emission from wastewater treatment plants (Escalas et.al.,2003). The emission of VOCs to atmosphere is a growing area of concern because,

apart from their potential toxicity, the highly reactive nature of some of VOCs contributes to the photochemical formation of ozone in the lower atmosphere (Binnie et al., 2002). Air emission (i.e., volatilization), biodegradation and sorption are main process affecting VOC fate in wastewater treatment plants. Air emission has found to prevail in both primary and secondary clarifier in wastewater

treatment plant (Corsi et al., 1995). The fluxes of VOCs emission from wastewater were measured using flux chamber (Sadek, et al., 1996) or estimated by using existing fate model (Bianchi, et al., 1998). The VOCs emission rate from wastewater depending on environmental conditions such as the wind speed and, water temperature (Bunyakan, et al, 2001), water depth (Lunney, at al., 1985) and suspended solid in wastewater (Binnie et al., 2002). The exist model for predicting VOC emission rate required mass transfer coefficient. The mass transfer coefficients for prediction VOC emission from wastewater, used currently, were developed base on volatilization of VOCs from water (Mackay and Yeun, 1983; Liss and Merlivat, 1986). To our knowledge, there are no mass transfer coefficient correlations for predicting of VOCs emission rate from wastewater, which actually developed base on emission rate of VOCs from wastewater.

To develop such mass transfer coefficient, the understanding of volatilization process of VOCs from wastewater is required. The aim of this research was to investigate the effect of wind speed on mass transfer process of VOCs from wastewater. The novelty of this work is not the method used to determine the mass transfer coefficient but rather a use of wastewater instead of pure water as previously done and reported in the literatures.

In this work we present the volatilization of Methyl Ethyl Ketone (MEK) from wastewater and from pure water. The experiments are conducted in a pilot volatilization tank where the wind speed over the water surface can be varied and controlled at desired values. This type of setup is able to investigate the effect of the wind speed on mass transfer coefficients of MEK from both wastewater and pure water.

2. METHODS AND MATERIALS

2.1 Materials

Wastewater was taken from wastewater treatment facility at the Prince of Songkla University Hat Yai Songkhla Thailand. The characteristic of wastewater is shown in table 1.

Table 1. Wastewater characteristic

Properties	mg/l
BOD ₅	45-98
COD	2320-2960
Suspended solids (SS)	27-62

MEK (99.5%, Merck) is used in the experiments without any further purification.

2.2 Analysis

The liquid samples were analyzed using a gas chromatography (GC). The GC (Model HP9806, Hewlett Packard), equipped with a capillary column, a flame ionization detector and an integrator (Model 9860), was operated at an injection temperature of 180 °C, a detector temperature of 200 °C and an oven temperature of 50 °C. The GC column was a (HP-FAAP, Polyethylene glycol-TPA modified, Hewlett Packard), 30 m capillary glass column with an inside diameter of 0.25 mm. Helium was used as a carrier gas at a flow rate of 0.2 mL/min. This analytical system gave excellent resolution for MEK.

2.3 Volatilization experiment

The experiments for unsteady state volatilization of MEK from wastewater were conducted in the pilot volatilization tank. The experimental set-up is comprised of two main pieces of equipments; the volatilization tank (50 cm wide, 100 cm long, and 20 cm deep) and the wind generated system, as shown in figure 1.

2.3.1 Solution preparation

Prior to starting an experiment, the wastewater and the water contained MEK was prepared by dissolving MEK in wastewater or water to the desired concentration. The volumes of the wastewater and water were 100 L.

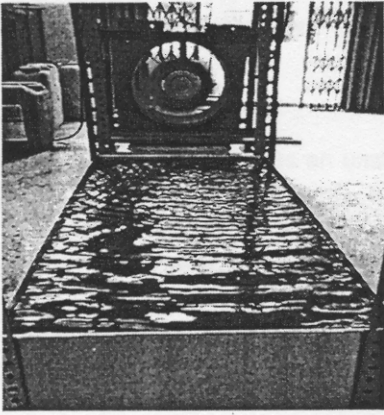


Figure 1. Pilot volatilization tank equipped with a wind generated system

2.3.2 Effect of the wind speed

The unsteady state volatilizations of MEK from wastewater and water were investigated at various wind speed. The wind speed was generated by a fan mounted just above water surface at the end of the volatilization tank (see figure 1). The speed of the fan was controlled by an inverter (Novem Inverters NSP series, Novem Engineering). The wind speeds were measured by using air velocity meter (Model TRI-Sense, Cole-Parmer Instrument Co.) at 10 cm above water surface for 15 positions across the water surface. The average value from 15 data points was then used as the average wind speeds at 10 cm (U_{10cm}) for each run. The U_{10cm} in the range of 0-4.42 m/s were used in this investigation. The water temperature, air temperature and relative humidity were monitored for all experiments using humidity/temperature meter (Model TRI-Sense, Cole-Parmer Instrument Co.) and they were 29 ± 1 °C, 31 ± 1 °C and 70-80%, respectively. The BOD₅, COD and SS of wastewater used in this study were 50-98 mg/L, 2320-2960 mg/L and 27-38 mg/L, respectively. The water depth of 20 cm, corresponding to the water volume of 100 L, was employed. The initial MEK concentrations in wastewater and in water were 600-759 mg/L and 659-756 mg/L, respectively.

2.3.3 Volatilization time and liquid sampling

The volatilization time was 5 hr. During experiment, liquid samples were collected periodically, from the middle of the tank, at 10, 30 and 60 minute in the first hour and every hour for the rest of volatilization time. Each liquid sample was collected in a 20 mL glass vial, capped immediately with a Teflon-lined septum, marked and placed in cooler. Each sample was filtrated and analyzed for MEK concentration. The concentration-time data of each run is used to determine the overall mass transfer coefficient of MEK.

2.3.4 Data analysis

Overall mass transfer coefficients for MEK volatilization from wastewater and from pure water are determined using two-film model, which is express as equation (1)

$$R_{MEK} = K_{OL} a \left(C_{MEK,L} - \frac{HC_{MEK,G}}{RT} \right) \quad (1)$$

where $K_{OL} a$ = overall mass transfer coefficient, m^3/s

a = surface area of wastewater, m^2

$C_{MEK,L}$ = concentration of MEK in liquid phase, mol/m^3

$C_{MEK,G}$ = concentration of MEK in gas phase, mol/m^3

H = Henry law constant, $atm \cdot m^3$

R = universal gas constant
= $8.21 \times 10^{-5} atm \cdot m^3 / mol \cdot K$

T = absolute temperature, K.

The airflow over the water surface removed MEK as it volatile, maintaining the MEK concentration in gas phase above the wastewater surface at zero. Equation (1) reduces to

$$R_{MEK} = K_{OL} a C_{MEK,L} \quad (2)$$

The material balance around the volatilization tank, illustrated in figure 1, can be expressed as equation (3)

$$R_{MEK} = -V \frac{dC_{MEK,L}}{dt} \quad (3)$$

where V = volume of volatilization tank, m^3

t = volatilization time, s

Combining equation (2) and (3) gives

$$\frac{dC_{MEK,L}}{dt} = -\frac{K_{OL}a}{V} C_{MEK,L} \quad (4)$$

The integration of equation (4) gives an unsteady state concentration profile

$$C_{MEK,L} = C_{MEK,Lo} \exp\left(-\frac{K_{OL}a}{V} t\right) \quad (5)$$

or
$$\ln \frac{C_{MEK,L}}{C_{MEK,Lo}} = -\frac{K_{OL}a}{V} t \quad (6)$$

Providing that the concentration of MEK in volatilization tank is uniform, the overall mass transfer coefficient can be obtained from concentration-time data as shown by equation (6), using linear regression technique.

3. RESULTS AND DISCUSSION

3.1 Concentration as function of water depth

The concentrations of MEK were measured at 3 points, which were 5, 10, and 15 cm, below the water surface. The typical concentration-time data measured at difference water depths for the volatilization of MEK from wastewater and from pure water are shown in figure 2 and 3, respectively. From these figures, the concentration of MEK in volatilization tank was approximately uniform and the equation (6) can be applied to determine the overall mass transfer coefficient, $K_{OL}a$. Moreover, the lower degree of uniform concentration, observed in wastewater, as compared to that of pure water, may arise from the presence of organic mass in wastewater, which may act as a mass transfer barrier in liquid phase.

3.2 Concentration profiles and $K_{OL}a$ determination

The $K_{OL}a$ for volatilization of MEK from wastewater and from water were determined for each experiment by performing a linear regression analysis between $\ln(C_{MEK,L}/C_{MEK,0})$ against t according to equation (6).

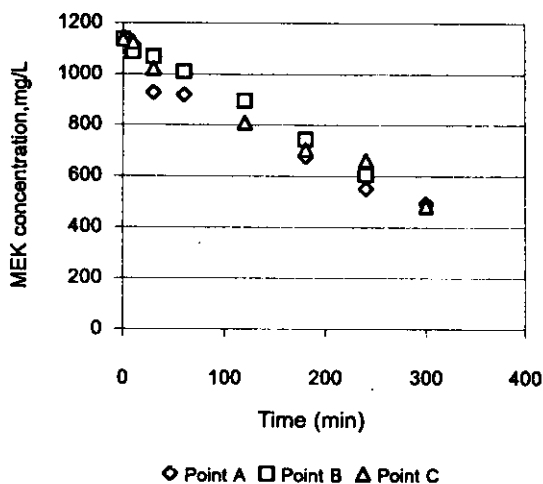


Figure 2. MEK concentrations as function of time measured at various depths of wastewater. Point A, B and C were 15, 10, and 5 cm below water surface, respectively

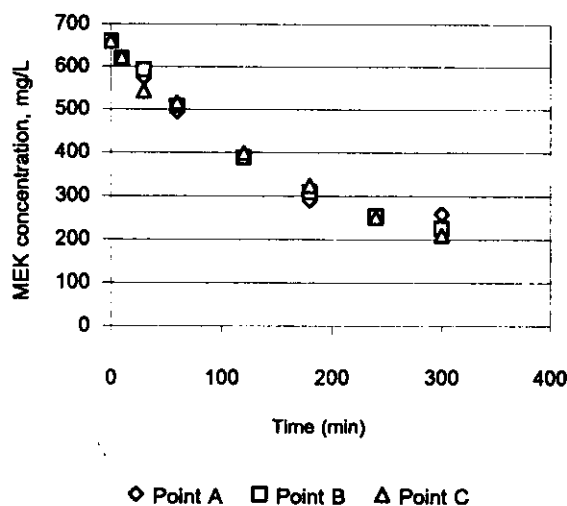


Figure 3. MEK concentrations as function of volatilization time measured at various depths of pure water. Point A, B and C were 15, 10, and 5 cm below water surface, respectively

The experimental results fit the model predictions with a correlation coefficient of more than 0.99 for all experiments. Figure 4 shows a typical plot of results for an experiment of wastewater. The results indicate that volatilization of MEK from wastewater followed first-order kinetics with respect to MEK concentration in bulk water. The $K_{OL}a$ determined under a range of wind speed were then used to evaluate the effects of wind speed on the mass transfer process.

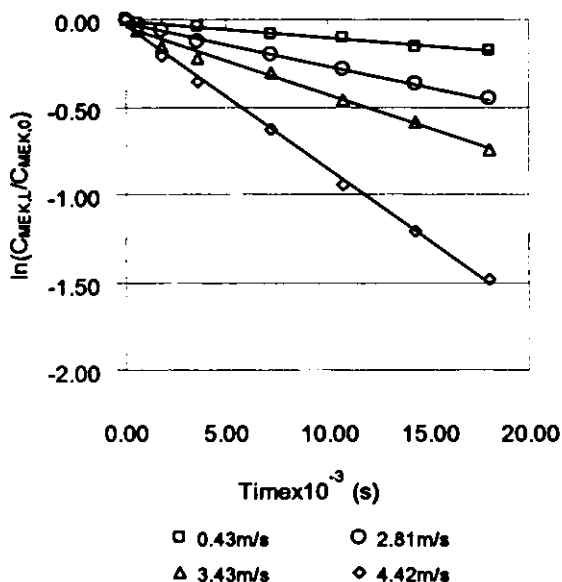


Figure 4. Typical plot of $\ln(C_{MEK,L} / C_{MEK,0})$ vs. t for volatilization of MEK from wastewater at various wind speeds.

3.3 The effect of the wind speed on $K_{OL}a$

The effect of the wind speed on $K_{OL}a$ for volatilization of MEK from wastewater and from water is shown in figure 5. The data show that the relationship between $K_{OL}a$ and wind speed falls into two regimes with a break at the wind speed range of 2-3 m/s.

At low wind speed regime, $U_{10cm} \leq 2.40$ m/s, the $K_{OL}a$ of MEK was slightly increased as the wind speed increased.

At high wind speed regime, $U_{10cm} > 2.40$ m/s, the $K_{OL}a$ of MEK was strongly influenced by the wind speed. The relationship between $K_{OL}a$ and the wind speed is linear but has a distinctly higher slope as compared to that of low wind speed regime. The transition of $K_{OL}a - U_{10cm}$ relationship at 2.40 m/s are agree well with the results previously reported by a number of researchers, which usually found at 2-3 m/s (Bunyakan, et al., 2001; Wanninkhof, et al., 1991). The high wind speed, greater than 2.40 m/s, provided a shear stress at air-water surface that is high enough to set the interface and liquid below in motion resulting

in high turbulence and the appearance of waves. Consequently, $K_{OL}a$ were increased significantly with increasing wind speed in this regime.

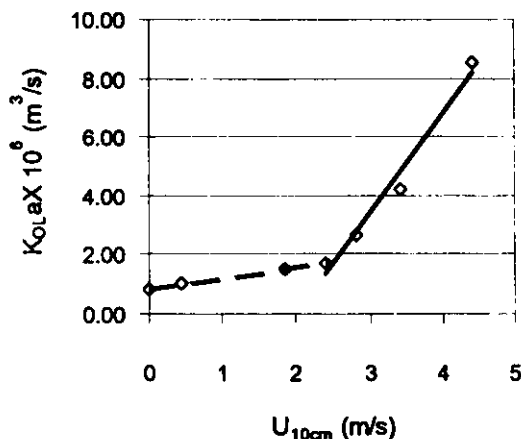


Figure 5. $K_{OL}a$ of MEK as a function of the wind speed ($T_{wastewater} = 29 \pm 1$ °C, Water depth = 20 cm and wastewater properties: BOD 50-98 mg/L, COD = 2320-2960 mg/L and SS = 27-38 mg/L)

The extent of turbulence at air-water interface can be represented by the Reynolds number calculated at 10 cm above the water surface, Re_{10cm} , (Bunyakan, et. al, 2005) as given by equation (7).

$$Re_{10cm} = \frac{\rho U_{10cm} L}{\mu} \quad (7)$$

where U_{10cm} = average wind speed over the water surface measured at 10 cm above water, m/s,

ρ = air density, kg/m³,

μ = air viscosity, kg/(m s), and

L = length of water surface, m.

The $K_{OL}a$ of MEK are plotted as a function of the Reynolds number, Re_{10cm} , in figure 6. From this figure we can see that the change in slope of relationship between $K_{OL}a$ and Re_{10cm} occurred at Re_{10cm} values of 1.40×10^5 which approximately characterized the change of airflow from laminar to turbulence.

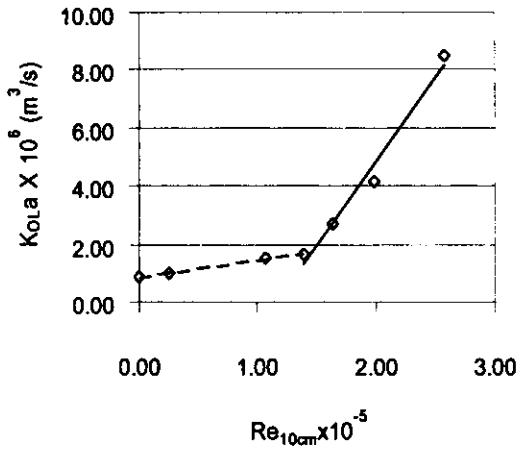


Figure 6. $K_{OL}a$ of MEK as a function of the Reynolds number ($T_{wastewater} = 29 \pm 1$ °C, Water depth = 20 cm and wastewater properties: BOD 50-98 mg/L, COD = 2320-2960 mg/L and SS = 27-38 mg/L)

In laminar regime, $Re_{10cm} \leq 1.40 \times 10^5$, the $K_{OL}a$ was found to slightly increase with increasing Reynolds number. A linear regression between $K_{OL}a$ and the Reynolds number was carried out and yielded the following relation ($R^2 = 0.99$).

$$K_{OL}a = 6.0 \times 10^{-12} Re_{10cm} + 8.4 \times 10^{-7} \quad (8)$$

$$Re_{10cm} \leq 1.40 \times 10^5$$

In turbulence regime, $Re_{10cm} > 1.40 \times 10^5$, The $K_{OL}a - Re_{10cm}$ relationship is also linear but has a distinctly higher slope. A linear regression between $K_{OL}a$ and the Reynolds number was carried out and yielded the following relation ($R^2 = 0.99$).

$$K_{OL}a = 5.8 \times 10^{-11} Re_{10cm} - 6.8 \times 10^{-6} \quad (9)$$

$$Re_{10cm} > 1.4 \times 10^5$$

3.4 The comparison between $K_{OL}a$ from wastewater and from pure water

The comparison between $K_{OL}a$ for volatilization of MEK from wastewater and that of pure water is shown in figure 7. It can be seen that the $K_{OL}a$ of wastewater were lower than those of pure water for all investigated ranges of Reynolds number.

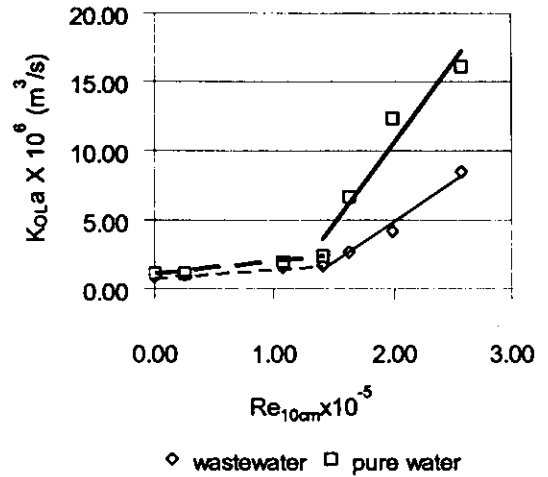


Figure 7. The comparison between $K_{OL}a$ of MEK volatilization from wastewater (BOD 50-98 mg/L, COD = 2320-2960 mg/L and SS = 27-38 mg/L) and that from pure water at $T_{water} = 29 \pm 1$ °C and a water depth of 20 cm.

In laminar regime, the difference between $K_{OL}a$ of MEK volatilization from wastewater and those from water was average about 27% and it was independent of the wind speed with about 29%, 25% and 28% being difference at wind speed of 0, 1.86 and 2.4 m/s, respectively. This implies that the presence of organic mass in wastewater is the key parameter controlling the mass transfer from wastewater. The organic mass in wastewater acts as a barrier to mass diffusion, thus reduces mass transfer rate and the $K_{OL}a$. The independent on the wind speed of % difference between $K_{OL}a$ of wastewater and that of water also implies that the wind speed in this regime provides very low extent of turbulence in water column.

In turbulence regimes, however, the difference between $K_{OL}a$ of wastewater and that of water were higher than that found in laminar regime. This implies that the organic masses in wastewater were acted as both the turbulence suppression and as a barrier to mass transfer. The high extent of turbulence caused high mass transfer rate in water and thus $K_{OL}a$ while the presence of organic mass in wastewater resulted

in lower extent of turbulence, at the same wind speed, which then led to much lower in $K_{OL}a$ as compared to pure water. Since the amount of organic masses in waste water were the same for all wind speed, the turbulence suppression capability of this organic mass decreased with increasing wind speed as indicated by the higher in the difference between the $K_{OL}a$ of wastewater and those of water were observed with increasing wind speed.

3.5 Mass transfer coefficient for predicting MEK volatilization rate from wastewater

The mass transfer coefficient for predicting volatilization rate of MEK was developed based on the $K_{OL}a$ from wastewater. To account for the effect of water temperature on mass transfer coefficient, the film theory is used. The mass transfer coefficient varies with the first power of the molecular-diffusion coefficient. By applying the reference-substance concept (Rathbun and Tai, 1987) and the film theory, the $K_{OL}a$ correlation of MEK are given by equation (10).

$$K_{OL}a(T) = K_{OL}a(T_R) \left(\frac{D_{MEK-liquid}(T)}{D_{MEK-liquid}(T_R)} \right) \quad (10)$$

where $D_{MEK-liquid}(T)$ and $D_{MEK-liquid}(T_R)$ are the diffusion coefficient of MEK in water at any interested temperature (T) and at reference temperature (T_R), in cm^2/s , respectively. The diffusion coefficient of MEK at reference temperature of 29 °C, is $1.05 \times 10^{-5} cm^2/s$. Combining equation (8), (9) and (10), the $K_{OL}a$ for MEK at any temperature can be predicted for laminar and turbulence flow as given by equations (11) and (12), respectively.

$$K_{OL}a(MEK) = [6.0 \times 10^{-12} Re_{10cm} + 8.4 \times 10^{-7}] \left(\frac{D_{MEK-liquid}(T)}{1.05 \times 10^{-5}} \right), \quad Re_{10cm} \leq 1.40 \times 10^5 \quad (11)$$

$$K_{OL}a(MEK) = [5.8 \times 10^{-11} Re_{10cm} - 6.8 \times 10^{-6}] \left(\frac{D_{MEK-liquid}(T)}{1.05 \times 10^{-5}} \right), \quad Re_{10cm} > 1.40 \times 10^5 \quad (12)$$

3.6 Emission rate model of MEK

The emission rate of MEK as function of Reynolds number can be obtained by substituting $K_{OL}a$ from equation (11) and (12) into equation (2). Thus the rates of MEK emission from wastewater are given by equation (13) and (14) for laminar and turbulence airflow over the water surface, respectively.

$$R_{MEK} = [6.0 \times 10^{-12} Re_{10cm} + 8.4 \times 10^{-7}] \left(\frac{D_{MEK-liquid}(T)}{1.05 \times 10^{-5}} \right) C_{MEK,L}, \quad Re_{10cm} \leq 1.40 \times 10^5 \quad (13)$$

$$R_{MEK} = [5.8 \times 10^{-11} Re_{10cm} - 6.8 \times 10^{-6}] \left(\frac{D_{MEK-liquid}(T)}{1.05 \times 10^{-5}} \right) C_{MEK,L}, \quad Re_{10cm} > 1.4 \times 10^5 \quad (14)$$

where R_{MEK} is the MEK emission rate (mol/s) and $C_{MEK,L}$ is the concentration of MEK in wastewater (mol/m^3).

3.7 Simulate MEK emission rate from wastewater

Application of equations (13) and (14) is demonstrated through an example. Consider the wastewater contains MEK at the concentration of 500 mg/L. The wastewater treatment pond is 50 m long and the wind speeds at 10 cm above the water surface were varied from 0-4.42 m/s. The wastewater temperatures were varied from 25-35 °C. While the air temperature was assumed at 30 °C. The emission rate of MEK is need for air pollution management purposes.

The first step is to compute the necessary physical parameters of the VOC. The diffusion coefficients of MEK in water at various temperatures were computed and shown in table 1.

The next step is to estimate the Re_{10cm} according to equation (7). The Re_{10cm} for various wind speed are listed in table 2.

Table 1. Properties of MEK

T_{water} (°C)	$D_{MEK} \times 10^5$ (cm ² /s)	$\frac{D_{MEK}(T)}{D_{MEK}(T_R)}$	MW (g/gmol)
25	1.032	0.987	72.107
30	1.049	1.003	
35	1.067	1.020	

Table 2. The Reynolds number at air temperature of 30°C

U_{10cm} (m/s)	L (m)	$Re_{10cm} \times 10^6$	Flow Characteristic
0.5	50	1.45	Turbulence
1.0	50	2.91	
1.5	50	4.36	
2.0	50	5.81	
2.5	50	7.27	
3.0	50	8.72	
3.5	50	10.17	
4.0	50	11.63	

Since calculated Re_{10cm} were greater than 1.4×10^5 , equation (14) is then applied. The predicted MEK emission rate as function of the wind speed and the wastewater temperature are presented in figure 8. The emission rates of MEK are strongly depended on wind speed, as expected, and only slightly varied with wastewater temperature for simulated range of temperature, 25-35°C.

4. CONCLUSION

The volatilizations of MEK from wastewater and from pure water were investigated in pilot scale volatilization tank. The $K_{OL}a$ were determined under a range of conditions and were used to evaluate the effect of the wind speed on the mass transfer process. The results showed that the relationship between $K_{OL}a$ and the wind speeds falls into two regimes with a break at the wind speed of 2.4 m/s. This corresponds to the Reynolds number of 1.4×10^5 , which

approximately characterized the change of airflow from laminar to turbulence.

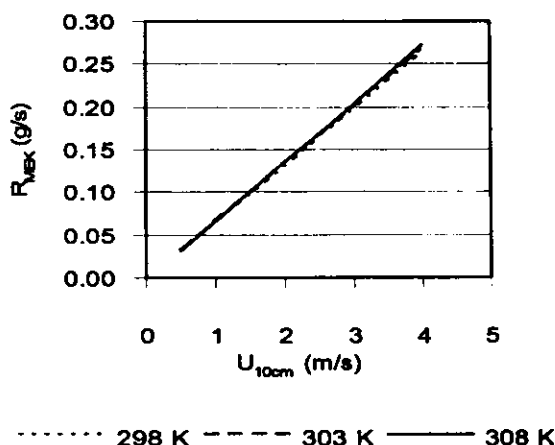


Figure 8. Predicted MEK emission rate as function of wind speed and wastewater temperature ($C_{MEK-L} = 400$ mg/L)

In laminar regime, $K_{OL}a$ was slightly increased with increasing Reynolds number and the linear relationship between $K_{OL}a$ and the Reynolds number was observed.

In turbulence regime, $K_{OL}a$ increased more rapidly. The relationship between $K_{OL}a$ and the Reynolds number was also linear but has a distinctly higher slope.

Moreover, $K_{OL}a$ obtained from MEK volatilization from wastewater were much lower than that from pure water, particularly in turbulence regime. The wind speed and wastewater characteristic (i.e., uncontrolled organic masses) are the important parameters in controlling the volatilization of MEK from wastewater. Finally, the $K_{OL}a$ based on wastewater experiments were used to develop the $K_{OL}a$ correlation. The use of this correlation for predicting of MEK emission rate from wastewater has been discussed.

5. ACKNOWLEDEMENTS

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