ORIGINAL ARTICLE

Factors affecting the volatilization of volatile organic compounds from wastewater

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Abstract

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This study aimed to understand the influence of the wind speed (U_{10cm}), water depth (h) and suspended solids (SS) on mass transfer coefficient ($K_{OL}a$) of volatile organic compounds (VOCs) volatilized from wastewater. The novelty of this work is not the method used to determine $K_{OL}a$ but rather the use of actual wastewater instead of pure water as previously reported. The influence of U_{10cm}, h, and SS on $K_{OL}a$ was performed using a volatilization tank with the volume of 100-350 L. Methyl Ethyl Ketone (MEK) was selected as a representative of VOCs investigated here in.

The results revealed 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. At $U_{10cm} \leq 2.4$ m/s, $K_{OL}a$ was slightly increased linearly with increasing U_{10cm}. For $U_{10cm} > 2.4$ m/s, $K_{OL}a$ increased more rapidly. The relationship between $K_{OL}a$ and U_{10cm} was also linear but has a distinctly higher slope. For the $K_{OL}a$ dependency on water depth, the $K_{OL}a$ decreased significantly with increasing water depth up to a certain water depth after that the increase in water depth

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had small effect on $K_{OL}a$. The suspended solids in wastewater also played an important role on $K_{OL}a$. Increased SS resulted in a significant reduction of $K_{OL}a$ over the investigated range of SS. Finally, the comparison between $K_{OL}a$ obtained from wastewater and that of pure water revealed that $K_{OL}a$ from wastewater were much lower than that of pure water which was pronounced at high wind speed and at small water depth. This was due the presence of organic mass in wastewater which provided a barrier to mass transfer and reduced the degree of turbulence in the water body resulting in low volatilization rate and thus $K_{OL}a$. From these results, the mass transfer model for predicting VOCs emission from wastewater should be developed based on the volatilization of VOCs from wastewater rather than that from pure water.

Key words : VOCs, volatilization, mass transfer coefficient, MEK

บทคัดย่อ

จรัญ บุญกาญจน์ แสนสุข แซ่อึ่ง จรรยา อินทมณี และ จันทิมา ชั่งสิริพร

ปัจจัยที่มีผลต่อการระเหยของสารอินทรีย์ระเหยจากน้ำเสีย

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การศึกษานี้มีวัตถุประสงค์เพื่อทำความเข้าใจถึงอิทธิพลของความเร็วลม (U_{10cm}) ความลึกของน้ำ (h) และของแข็งแขวนลอย (SS) ต่อสัมประสิทธิ์การถ่ายโอนมวลรวม ($K_{OL}a$) ของสารอินทรีย์ระเหย (VOCs) ที่ระเหยจากน้ำเสีย ความใหม่ของงานวิจัยนี้ไม่ได้อยู่ที่วิธีการหาค่า $K_{OL}a$ หรือการหาความสัมพันธ์ระหว่าง $K_{OL}a$ กับตัวแปรต่าง ๆ ข้างต้น แต่อยู่ที่การศึกษาโดยใช้น้ำเสียจริงแทนการใช้น้ำบริสุทธิ์ดังเช่นในงานวิจัยที่มีรายงานไว้ก่อนหน้านี้ การศึกษาอิทธิพลของ U_{10cm}, h, และ SS ต่อ $K_{OL}a$ ดำเนินการในถังระเหยขนาด 100-350 L สารอินทรีย์ระเหยที่เลือกศึกษาคือ เมททิลเอททิล คีโตน (Methyl Ethyl Ketone, MEK)

ผลการศึกษาพบว่า ความสัมพันธ์ระหว่าง $K_{OL}a$ กับ U_{10cm} แบ่งออกเป็นสองช่วงโดยมีจุดเปลี่ยนที่ U_{10cm} เท่ากับ 2.4 m s^{-1} ที่ $U_{10cm} \leq 2.4$ m s^{-1} $K_{OL}a$ เพิ่มขึ้นอย่างช้า ๆ แบบเชิงเส้นกับความเร็วลมที่เพิ่มขึ้นที่ $U_{10cm} >$ 2.4 m s^{-1} $K_{OL}a$ เพิ่มขึ้นอย่างรวดเร็ว ความสัมพันธ์ระหว่าง $K_{OL}a$ กับ U_{10cm} ยังคงเป็นแบบเชิงเส้นแต่มีความชันสูงกว่ามาก ผลการศึกษาอิทธิพลของความลึกของน้ำต่อ $K_{OL}a$ พบว่า $K_{OL}a$ ลดลงกับความลึกที่เพิ่มขึ้นจนถึงความลึกค่าหนึ่ง จากนั้นพบว่าการเพิ่มขึ้นของความลึกมีผลต่อ $K_{OL}a$ น้อย ส่วนผลของ SS ต่อ $K_{OL}a$ พบว่าการเพิ่มของ SS มีผลทำให้ $K_{OL}a$ ลดลงอย่างมีนัยสำคัญตลอดช่วงของ SS ที่ศึกษา สุดท้ายจากการเปรียบเทียบระหว่าง $K_{OL}a$ ที่ได้จากการระเหยของ MEK จากน้ำเสียและจากน้ำบริสุทธิ์ พบว่า $K_{OL}a$ ที่ได้จากน้ำเสียมีค่าต่ำกว่าค่าที่ได้จากน้ำบริสุทธิ์มาก โดยความแตกต่างนี้มีสาเหตุหลักมาจากสารอินทรีย์ที่แขวนลอยอยู่ในน้ำเสีย ซึ่งสามารถกีดกันการถ่ายโอนมวลและลดความปั่นป่วนในน้ำ ทำให้อัตราการระเหยและ $K_{OL}a$ ในกรณีของน้ำเสียต่ำกว่ากรณีของน้ำบริสุทธิ์มาก ดังนั้นแบบจำลองสำหรับทำนายการระเหยของ VOCs จากน้ำเสียควรพัฒนาจากข้อมูลการระเหยของ VOCs จากน้ำเสียจริงมากกว่าการใช้ข้อมูลการระเหยของ VOCs จากน้ำบริสุทธิ์

ภาควิชาวิศวกรรมเคมี คณะวิศวกรรมศาสตร์ มหาวิทยาลัยสงขลานครินทร์ อำเภอหาดใหญ่ จังหวัดสงขลา 90112

Volatile organic compounds (VOCs) are present in municipal wastewater due to discharges from domestic, commercial and industrial sources. Volatilization, biodegradation and sorption are the main processes affecting VOCs fate in wastewater treatment plants. VOCs volatilization has found to prevail in both primary and secondary clarifier in wastewater treatment plant (Corsi *et al.*, 1991). Although, the VOCs emission fluxes from wastewater treatment plants can be measured using a flux chamber, the estimation of VOCs emissions using a mass transfer model is preferred. Existing mass transfer models used for estimating VOCs emissions from wastewater, however, are developed based on the volatilization of VOCs from pure water (Mackay and Yeun, 1983; Lunney *et al.*, 1985; Bunyakan *et al.*, 2001). Moreover, most of the existing models included only the effect of the wind speed above water surface and water temperature. These parameters did not relate to any dimension of wastewater basin such as length, width and depth of water basin which was theoretically known to influence mass transfer rate in other systems (Cussler, 1984)

The objective of this study was to understand the roles of the wind speed above water surface, the water depth and the suspended solid on mass transfer coefficient of MEK that allowed for the determination of these effects on VOCs volatilization rate 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 investigated by other researchers (Mackay and Yeun, 1983; Lunney *et al.*, 1985; Bunyakan *et al.*, 2001).

Methods and Materials

1. Materials

Wastewater was taken from the wastewater treatment facility at Prince of Songkla University Hat Yai, Songkhla, Thailand. The characteristics of the wastewater are shown in Table 1.

MEK was selected as VOCs representative in this study since the volatilization of MEK from water is governed by both gas and liquid films. In other words, the volatilization rate of MEK from water is between that of VOCs whose the volatilization rates are controlled by gas film and liquid film, respectively. MEK (99.5%, Merck) was used in the experiments without any further purification.

Table 1. Wastewater characteristics analyzed from 27 water samples

Properties	mg/l
BOD_5	45-98
COD	2320-2960
Suspended solids (SS)	27-62

2. Analysis

The liquid samples were analyzed using 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 180ºC, a detector temperature of 200ºC and an oven temperature of 50ºC. 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.

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 two main pieces of equipments; the volatilization tank (50 cm wide, 100 cm long, and 5-70 cm deep) and the wind generator system, as shown in Figure 1.

3.1 Solution preparation

Prior to starting an experiment, the wastewater and the pure water containing MEK was prepared by dissolving MEK in wastewater or water to the desired concentrations. The volumes of the wastewater and water were varied from 25 to 350 L depending on water depth, which varied from 5-70 cm.

3.2 Effect of the wind speed

The unsteady state volatilizations of MEK from wastewater and water were investigated at various wind speeds. 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 the water surface for 15 positions across the water surface.

Figure 1. Pilot volatilization tank equipped with a wind generator system
[Color figure can be viewed in the electronic version]

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±1ºC, 31±1ºC and 70-80%, respectively. The BOD_5, 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.

3.3 Effect of water depth

The unsteady state volatilizations of MEK from wastewater and water were investigated at various water depths. The water depths were varied from 5 to 70 cm, corresponding to water volume varying from 25 to 350 L. To prevent the accumulation of MEK near the water surface, the wind speed over the water surface was kept constant at 2.81 m/s for all experiments. The water temperature was at room temperature (29±1ºC). The BOD_5, COD and SS of wastewater used in this study were 45-58 mg/L, 2868-3285 mg/L and 42-62 mg/L, respectively. The initial concentrations of MEK in wastewater and in water were in the same range as those for wind speed investigation.

3.4 Effect of suspended solids

The unsteady state volatilizations of MEK from wastewater were investigated at various suspended solids. The water depths were at 20 cm, corresponding to water volume of 100 L. The water temperature was at room temperature (29±1ºC). The SS of wastewater were varied from 0 to 40 mg/L. The initial concentrations of MEK in wastewater were between 659 and 1096 mg/L while the wind speed over the water surface was kept constant at 2.81 m/s for all experiments.

3.5 Volatilization time and liquid sampling

The volatilization time was 5 hr. During the experiment, liquid samples were collected periodically from the middle of the tank at a predetermined depth from water surface depending on total water depth. The sampling periods were 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 filtered and analyzed for MEK concentration. The concentration-time data of each run was used to determine the overall mass transfer coefficient of MEK.

3.6 Data analysis

The overall mass transfer coefficients for MEK volatilization from wastewater and from pure water were determined from concentration-time data using a two-film model and mass balance around the volatilization tank. Providing that the concentration of MEK in the volatilization tank is uniform, the overall mass transfer coefficient can be obtained from the slope of a plot between ln [$C_{MEK,L}$ / $C_{MEK,o}$] as shown by equation (1). The details were given elsewhere (Bunyakan *et al.*, 2001)

$$\ln\frac{C_{MEK,L}}{C_{MEK,Lo}}=\frac{K_{OL}a}{V}t \qquad (1)$$

where $K_{OL}a$ = overall mass transfer coefficient of MEK, m^3/s
$C_{MEK,L}$ = concentration of MEK in liquid phase at any time t, mol/m^3
$C_{MEK,Lo}$ = concentration of MEK in liquid phase at $t = 0$, mol/m^3
V = volume of volatilization tank, m^3
t = volatilization time, s

Results and Discussion

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

The volatilization of MEK from wastewater and pure water was investigated at various wind speeds varied from 0-4.42 m/s where the water depth was constant at 20 cm. In order to determine $K_{OL}a$ the concentration-time data were measured. The typical concentration-time data measured at different depth from water surface, at the middle of the volatilization tank, for the volatilization of MEK from wastewater and pure water, at U_{10cm} of 2.81, are shown in Figure 2. It is clearly shown from Figure 2 that the volatilization rate of MEK from wastewater was significantly lower than that of pure water as indicated by a lower slope of concentration-time data. The concentrations of MEK were found uniform over the entire water body with a relative standard deviation (RSD%) less than 10%. Thus equation (1) can be applied for $K_{OL}a$ determination.

The effect of the wind speed on $K_{OL}a$ for MEK volatilization from wastewater is shown in

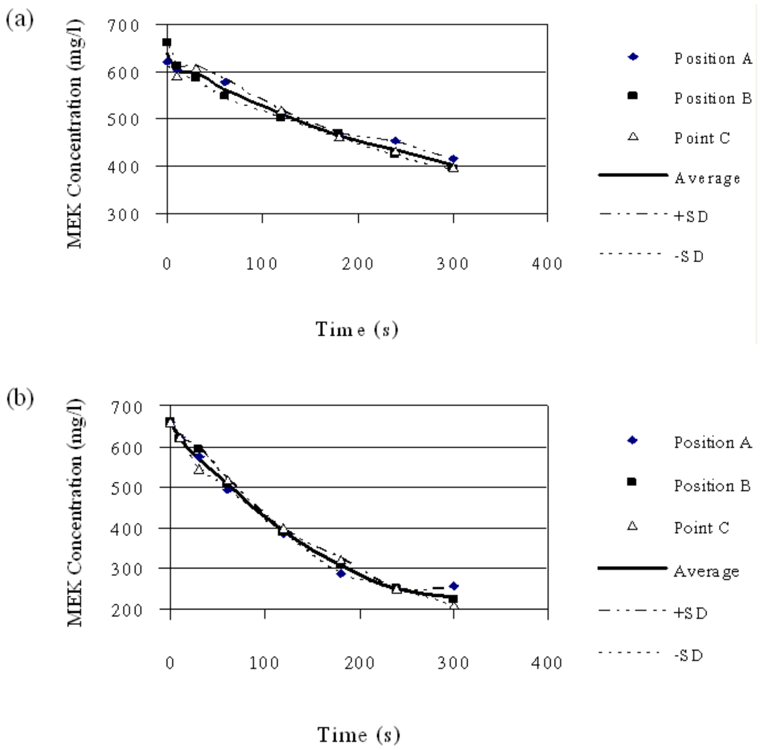

Figure 2. MEK concentrations as function of time measured at various water depths (Point A, B and C were 15, 10, and 5 cm below water surface, respectively); (a) wastewater and (b) pure water. U_{10cm} = 2.81 m/s.

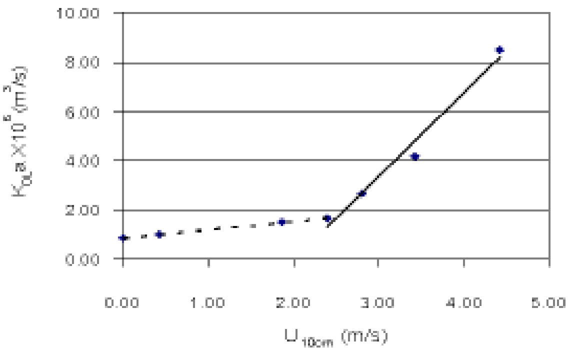

Figure 3. $K_{OL}a$ of MEK as a function of the wind speed ($T_{wastewater}$ = 29±1ºC, Water depth = 20 cm and wastewater properties: BOD 50-98 mg/L, COD = 2320-2960 mg/L and SS = 27-38 mg/L)

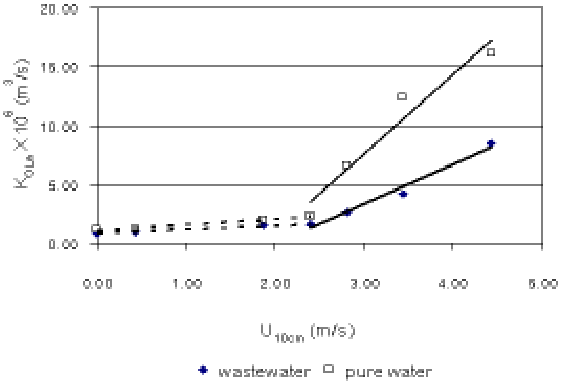

Figure 4. The comparison between $K_{OL}a$ of MEK volatilization from wastewater (BOD 50-98 mg/L, COD = 2320-2960 mg/L and SS = 27-38 mg/L) and that from pure water at T_{water} = 29±1ºC and a water depth of 20 cm.

Figure 3. From Figure 3, the data show that the relationship between $K_{OL}a$ and U_{10cm} fell into two regimes with a break at the wind speed range of 2.4 m/s. At low wind speed regime, $U_{10cm} \leq 2.40$ m/s, $K_{OL}a$ of MEK slightly increased as the wind speed was increased. At the high wind speed regime, $U_{10cm} > 2.40$ m/s, $K_{OL}a$ of MEK was strongly influenced by the wind speed. The relationship between $K_{OL}a$ and the wind speed was linear but had a distinctly higher slope compared with that of the low wind speed regime. The transition of $K_{OL}a$-U_{10cm} relationship at 2.40 m/s agreed well with the results previously reported by a number of researchers, who mostly found the transition 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 was high enough to set the interface and liquid below in motion resulting in high turbulence and the appearance of waves. Consequently, $K_{OL}a$ increased significantly with increasing wind speed in this regime.

The comparison between $K_{OL}a$ for MEK volatilization from wastewater and from pure water is shown in Figure 4. It can be seen that the $K_{OL}a$ of wastewater were lower than those of pure water for all wind speeds. The ratios of $K_{OL}a$ (water) to $K_{OL}a$ (wastewater) are shown as function of U_{10cm} in Figure 5. It can be seen from this figure that the ratios of $K_{OL}a$ (water) to $K_{OL}a$ (wastewater) were

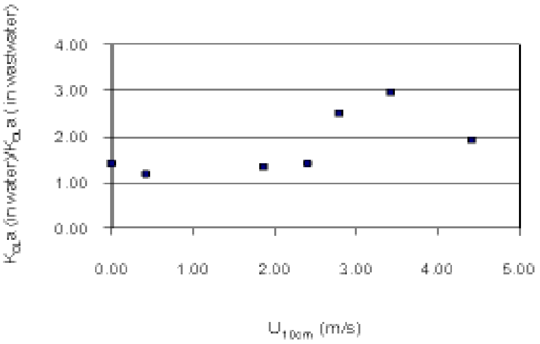

Figure 5. The ratio $K_{OL}a$ (in water) / $K_{OL}a$ (in wastewater) of MEK as function of the wind speed at T_{water} = 29±1ºC and a water depth of 20 cm.

roughly constant at 1.33 over the low wind speed regime ($U_{10cm} \leq 2.40$ m/s). However, at the high wind speed regime, $U_{10cm} > 2.40$ m/s, the ratio of $K_{OL}a$ (water) to $K_{OL}a$ (wastewater) were found to increase with increasing wind speed up to the wind speed of 3.4 m/s and then decrease slightly. This may attributed to the fact that the turbulence in the water body increased with increasing wind speed. The organic masses suspended in wastewater acted as both the turbulence suppression medium and as a barrier to mass transfer through diffusion process resulting in a significant reduction of mass transfer as compared to that in pure water. Thus the ratios of $K_{OL}a$ (water) to $K_{OL}a$ (wastewater) then increased with increasing wind speed up to a wind speed of 3.4 m/s. When the wind speed was higher than 3.4 m/s, the wind speed is high enough to overcome both the turbulence suppression and barrier effects. Thus the $K_{OL}a$ (wastewater) increased and the ratio of $K_{OL}a$ (water) to $K_{OL}a$ (wastewater) decreased as shown in Figure 5.

2. The effect of the water depth on volatilization rate of MEK

In the case where the water is fairly deep, the volatilization of VOCs from wastewater due to surface wind speed may depend on the water depth. In this research we have investigated the volatilization of MEK from pure water and wastewater for various water depths varied from 5-70 cm where the wind speed at the water surface was fixed at 2.81 m/s. The MEK concentrations in bulk liquid were measured at various positions along the water depth. The typical concentration-time data for volatilization of MEK from wastewater and pure water at high water depth are illustrated in Figures 6 and 7.

From Figure 6 and Figure 7 we can see that the volatilization rate, which is proportional to a slope of concentration-time data of MEK volatized from wastewater and pure water were found to depend on the water depth. The larger the water depth the smaller the volatilization rate was observed. However, the concentration of MEK even at the largest water depth still can be assumed as uniform concentration with a relative standard deviation (RSD%) less than 10% as evidenced in Figures 6 and Figure 7. Thus equation (1) can still be applied to determine the $K_{OL}a$ at various water depths investigated in this work.

3. The effect of the water depth on $K_{OL}a$

The effect of the water depth on $K_{OL}a$ for volatilization of MEK from wastewater and from water is shown in Figure 8. It is clearly shown from this figure that the $K_{OL}a$ decreased significantly with increasing water depth. Since, at high water depth, the degree of turbulence due to the surface wind was lower than that at the lower depth, the mass transfer coefficient was then decreased with increasing water depth. The organic masses presented in wastewater suppressed the turbulence and provided a barrier to mass transfer, thus $K_{OL}a$ of wastewater was significantly lower than that of

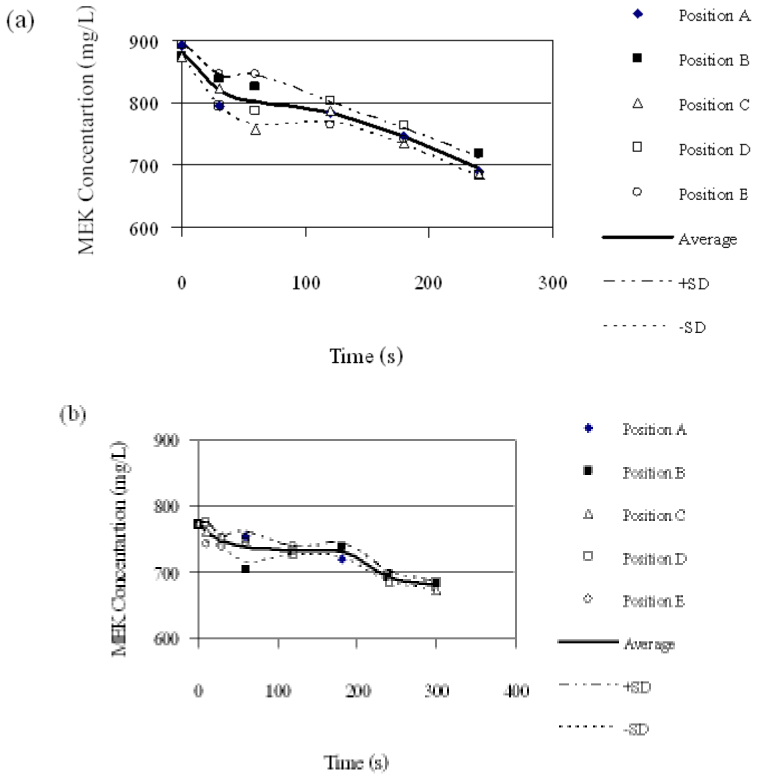

Figure 6. Typical concentration-time data for volatilization of MEK from wastewater at high water depth a) Total depth of 40 cm (A = 35 cm ,B = 30 cm, C = 20 cm, D = 10 cm and E = 5 cm below water surface) b) Total depth of 60 cm (A = 55 cm, B = 40 cm, C = 30 cm, D = 20 cm and E = 5 cm below water surface)

pure water, particularly at shallow water as shown in Figure 8. However, at water depth greater than the certain value of 30 cm, the degree of turbulence in water body was dominated by the water depth, the $K_{OL}a$ of wastewater and of pure water were then approximately the same as evidenced in Figure 8.

4. The effect of the suspended solid on $K_{OL}a$

From the previous results of the effect of the wind speed and the water depth on $K_{OL}a$, we believe that the suspended solid present in wastewater played an important role on the reduction of $K_{OL}a$ in wastewater as compared to pure water. The effect of suspended solids (SS) on $K_{OL}a$ was then investigated. $K_{OL}a$ were measured as function of SS presented in wastewater which was varied from 0-40 mg/L. The wind speed over the water surface and the water depth of wastewater were controlled at 2.81 m/s and 20 cm, respectively. The influence of SS on $K_{OL}a$ is illustrated in Figure 9.

It is clearly shown from Figure 9 that the $K_{OL}a$ significantly decreased with increasing SS in wastewater. There are three possible mechanisms which may be responsible for the decrease of $K_{OL}a$ with increasing SS. These are the adsorption of MEK on solid particles, the barrier to mass transfer and the suppression of turbulence in water by suspended solid particles. A number of researches have investigated adsorption of VOCs on suspended solid particles. Most of the previous research reported that the removal of VOCs due to adsorption onto suspended solid was considerably low and strongly depended on water temperature, the low temperature was preferred (Bianchi and Varney, 1998). Wakeham *et al.* (1983) reported

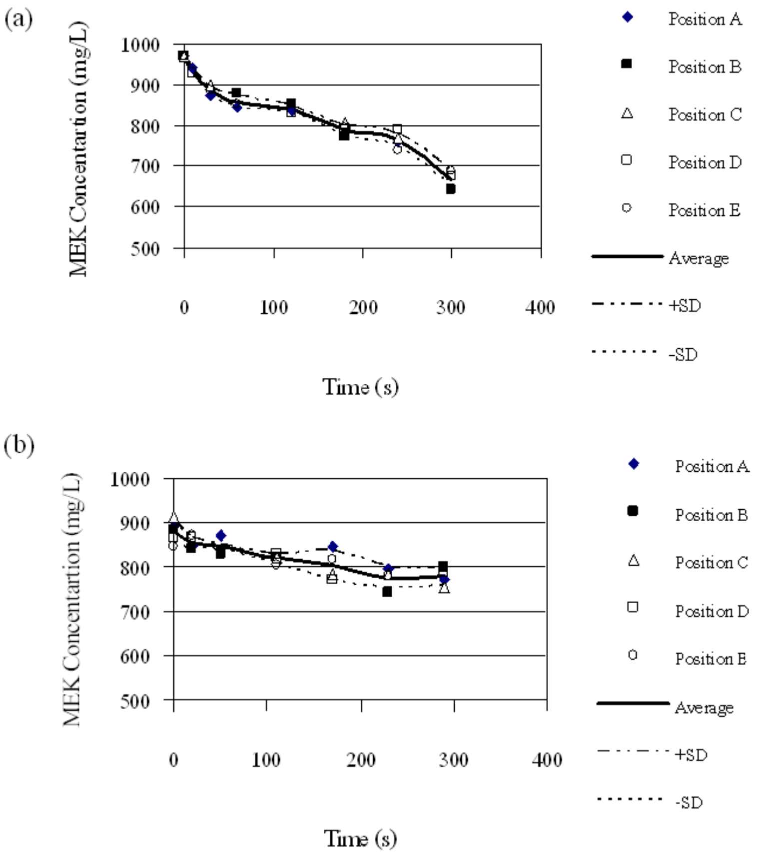

Figure 7. **Typical concentration-time data for volatilization of MEK from pure water at high water depth. a) Total depth of 40 cm (A = 35 cm ,B = 30 cm, C = 20 cm, D = 10 cm and E = 5 cm below water surface) b) Total depth of 65 cm (A = 60 cm, B = 50 cm, C = 35 cm, D = 20 cm and E = 5 cm below water surface)**

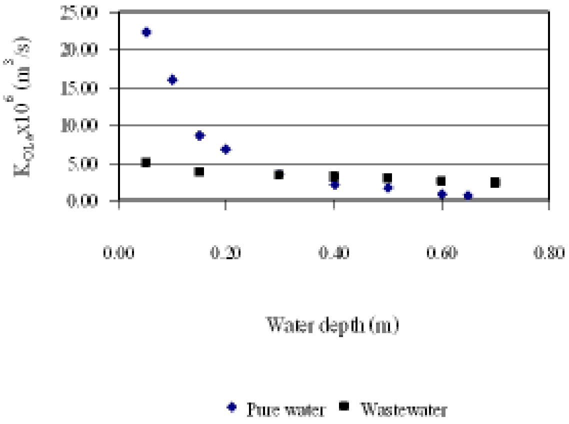

Figure 8. **The comparison between $K_{OL}a$ of MEK from wastewater and from pure water at various water depth.**

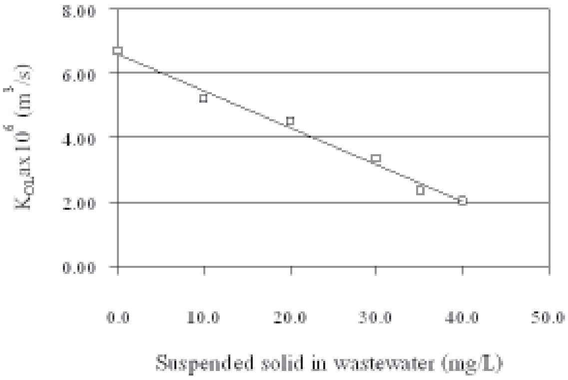

Figure 9. The effect of SS on $K_{OL}a$ of MEK at wind speed of 2.81 m/s, water depth of 20 cm and water temperature of 29±1ºC.

that only about 0.3% of VOCs such as toluene and tetrachloroethylene were adsorbed on to particulates. From these previous works we can see that the adsorption is a minor process for removal of VOCs from liquid. We then believed that the significantly decreased of $K_{OL}a$ in the presence of SS found in our investigation may be attributed mostly to the barrier effect and the turbulence suppression due to the presence of the organic solid in wastewater. In this case, the suspended solid in wastewater acts as a barrier for MEK transfer, reducing diffusion ability and eddy current (or the extent of turbulence) in bulk liquid phase, and thus reducing the volatilization rate and $K_{OL}a$.

Conclusion

The volatilizations of MEK from wastewater and from pure water were investigated in a pilot scale volatilization tank. The influence of the surface wind speed, the water depth and the suspended solid on $K_{OL}a$ were determined. The results revealed that the relationship between $K_{OL}a$ and the wind speeds fell into two regimes with a break at the wind speed of 2.4 m/s. At $U_{10cm} \leq 2.4$ m/s, $K_{OL}a$ was slightly increased with increasing U_{10cm} and a linear relationship between $K_{OL}a$ and U_{10cm} was observed. For $U_{10cm} > 2.4$, $K_{OL}a$ increased more rapidly. The relationship between $K_{OL}a$ and U_{10cm} was also linear but had a distinctly higher slope. The $K_{OL}a$ of MEK volatilized from wastewater decreased significantly with increasing water depth and the suspended solid in wastewater. The $K_{OL}a$ of MEK volatilized from wastewater were lower than those of pure water. These were pronounced at high wind speed and shallow water conditions. The lower $K_{OL}a$ found in wastewater as compared to pure water was due to the difference in water body characteristics. The presence of organics masses in wastewater provided a barrier to mass transfer and reduced the degree of turbulence in wastewater resulting in low volatilization rate and thus $K_{OL}a$. The mass transfer model for predicting VOCs emission from wastewater was then recommended to be developed from volatilization of VOCs from wastewater rather than those from pure water.

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