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Effects of initial chlorobenzene concentration, air flowrate and temperature on mass transfer of chlorobenzene by air stripping

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ABSTRACT

Volatile organic compounds (VOCs) are widely used in various industrial processes and caused water pollutions. VOCs removal from raw water is essential for water works to guarantee drinking water security. Air stripping column experiment was conducted to assess the effects of chlorobenzene (CB) initial concentration, air flowrate and temperature on the mass transfer of CB during air stripping. Air–water mass transfer of CB was quantified using the air–water mass transfer coefficient which was obtained from the simulation of a lumped parameter model. Experimental results demonstrated that air–water overall mass transfer coefficients (K_L) obtained from this study ranged from 10^{-1} and 10^{0} min^{-1}. From a correlation analysis, K_L was found directly proportional to the initial concentration of CB, air flowrate and temperature. The correlation result of K_L with air flowrate and temperature demonstrates that the air–water overall mass transfer coefficients was strongly affected by the temperature of the bulk liquid.

Keywords: Volatilization; VOCs; Air stripping; Mass transfer; Effect

1. Introduction

Volatile organic compounds (VOCs) , such as trihalomethanes, chlorinated solvents etc. are chemical materials widely used in industrial activities [1]. The improper disposal of VOCs led to serious water pollution and to the necessity of finding effective remediation technologies for its removal. This is particularly urgent when the water is utilized as drinking water as the VOCs has a potential risk to human health [2].

The removal of VOCs is an intractable problem for waterworks, especially during a sudden pollution episode such as VOCs spilling in raw water. The cleanup of the sudden contaminated waters is often difficult and expensive, for it usually involves treating dilute VOCs solution. Traditionally, the treatments for removing VOCs in water include air stripping, adsorption, advanced oxidation, distillation, anaerobic/aerobic biological treatment, bioreactor and supported liquid membrane [3–5]. These methods have some shortcomings and limitations. For example, adsorption is economic only at low VOCs levels due to the high cost of the adsorbents and their regeneration system [6]. In comparison, distillation is economic only at higher VOCs levels. Biological treatment is a clean method, but is time consuming and effective only at low VOCs levels. Thereby biological treatment cannot meet the demand of emergency rapid response to water pollution. Advanced oxidation

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is efficient for specific compound, but it may form new products that are more harmful than the original one [7]. Although supported liquid membrane could possess high flux using hollow fibers, it often suffers from the membrane stability [6,8–10]. Air stripping is assumed to be the best known method, technically and economically for removing VOCs from ground water to meet drinking water regulations [11]. In addition, air stripping process can be operated in water intakes or distributing well, and is applicable to most waterworks in China.

In general, air-stripping is a process where VOCs adsorbs to the air–water surface from the liquid, and then the adsorbed solute molecules find ways to evaporate into the air [12]. The bulk concentration of the pollutant is gradually reduced. The successful design of air-stripping systems in emergency conditions requires an understanding of the mass transfer coefficients for a given organic compound that affect the removal efficiency. There are special studies [13,14], in which the coefficients for VOCs were measured as there is an opinion that commercial design information is not specifically related to trace quantities of organic contaminants in water. However, the effects of air flowrate and temperature on mass transfer of air-stripping process for drinking water emergency treatments have been largely ignored in consideration.

The main focus of this study is to experimentally establish a basic understanding of air stripping efficiency for VOCs removal with respect to air flowrate variation and temperature changes. An estimate of mass transfer coefficient will provide further insights into the effects of air flowrate and temperature on VOCs mass transfer during air bubbling. In this study, chlorobenzene (CB) was used as a typical pollutant of VOCs, for it has been detected frequently in lots of surface water.

2. Experimental

2.1. Materials

Analytical grade CB (minimum 99.8% purity) was chosen as the target contaminants. Raw water was collected in a surface water source from Mopanshan reservoir (Harbin, Heilongjiang Province), and the water quality is shown in Table 1. CB-contaminated water was prepared by adding predetermined amount of CB into raw water without any other solvent. According to the previous studies [15], air–water partition coefficients (K_{iaw}) of CB at different temperature were given in table 2.

Table 1
Raw water characteristics

Raw water source	pH	UV_{254} (cm^{-1})	DOC ($mg\ l^{-1}$)	SUVA ($l\ (m \cdot mg)^{-1}$)	Turbidity (NTU)
Mopanshan reservoir	7.7	0.18	6.1	2.95	0.98

Table 2
Air–water partition coefficients (K_{iaw}) of CB at different temperature

Temperature	0°C	5°C	10°C	15°C	25°C	30°C
K_{iaw}	0.048	0.062	0.079	0.101	0.158	0.197

2.2. Analysis

All collected samples were filtered using a 0.45 μm glass fiber filter to remove suspended solid. CB was determined by high performance liquid chromatography (Waters Associates, U.S.A.), using a variable wavelength 2478 UV dual channel detector. The chromatographic condition was: flowrate 0.5 ml min^{-1}, injection volume 1 μl, UV wavelength 254 nm, C_{18} column (25–35 μm particles, 100 Å pore sizes, 4.6 × 250 nm, RS tech Corporation), mobile phase: methanol/water is 70:30 (v/v).

2.3. Experimental procedures

Stripping experiments were conducted in a bubbled column (60 mm internal diameter × 170 mm height). During the stripping, 400 ml contaminated raw water was filled in the column, and air was introduced into the bottom of the vessel through a microporous diffuser. Air flowrate (1.0–3.0 l min^{-1}) was set stably via the using of high-precision rotameter.

The effects of initial CB concentration, temperature and air flowrate on CB removal during the air stripping were investigated according to the following procedure: experimental tests with variable initial CB concentrations (1–10 mg l^{-1}, the Chinese drinking water quality standards set the maximum allowable concentration of CB at 0.3 $mg \cdot l^{-1}$) were performed under an air flowrate of 2.5 l min^{-1} at 288 K. The air stripping kinetics studies were performed at five different flowrates (1, 1.5, 2, 2.5, and 3 l min^{-1}) under a desired temperatures (0, 5, 10, 15, 25, and 30°), with CB initial concentration of 7 mg l^{-1}. Samples were taken at selected time intervals and the system operation time was 15–40 min.

3. Results and discussion

Figs. 1, 3 and 5 present typical experimental results showing the change of CB concentration over time at different temperature and various air flowrate during the air bubbling. Experimental results indicated that the concentration of CB decreased rapidly during the initial steps of the air stripping operation (0–4 min), and finally kept constant. The rapid depletion of CB at initial step was ascribed to the faster volatilization rate of CB at higher concentration. The asymptotic aqueous CB concentration was reached between 4 and 40 min after a start of bubbling, implying that a quasi-steady condition for the volatilization of CB was reached for all the experimental runs. This low concentration continued until an undetectable value was reached after a period of stripping time, meanwhile the qualitative shape of CB concentration resembled to be field application results. In field-scale systems such aqueous effluent concentrations may be due to rate-limiting nonequilibrium mass transfer processes such as sorption, CB dissolution, and retarded CB diffusion in the aqueous phase.

3.1. Lumped parameter model

All experiments in this study were based on the hypothesis that the aqueous concentration of CB in the stripping column was uniform prior to the air bubbling. Once the air was introduced into the stripping system, the concentration gradient of CB was formed around the bubbles, and hence CB present in the bulk water volatilized from the air–water interface to air. In the stripping system, air bubbles were assumed to be spherical (in the same diameter) and evenly distributed throughout with a same velocity. A mass balance for the solute system gives the transfer rate as:

$$V_L \frac{dC_L}{dt} = Q(C_{G0} - C_{Gf}) \tag{1}$$

where Q is the gas flowrate (l·min^{-1}), V_L is the volume of the liquid (L), C_L is the CB concentration in the liquid phase (mg·l^{-1}), t is time (min), C_{G0} and C_{Gf} are CB concentration in the entrance gas and exit gas (mg·l^{-1}), respectively. Since CB concentration in the entrance gas was assumed to be zero, i.e., $C_{G0} = 0$, Eq.(1) may be simplified as:

$$V_L \frac{dC_L}{dt} = -\,QC_{Gf} \tag{2}$$

CB diffusion from liquid to air can be described by the two-film theory as listed below:

$$Q \cdot dC_G = A \cdot k_L \cdot \alpha \cdot (C_L - C_{Le}) \cdot dz \tag{3}$$

where z is the height of liquid (m), A is the cross section area (m^2), k_L is the gas–liquid mass transfer coefficient, α is the gas–liquid interfacial area (m^2 m^{-3}), and C_{Le} is CB concentration in the liquid (mg l^{-1}), which is assumed to be in equilibrium with CB concentration in gas.

Since the relationship between C_G and C_{Le} follows Henry's law equation (Eq.(4)) in air stripping system, which can be described as:

$$C_G = K_{iaw} C_{Le} \tag{4}$$

where K_{iaw} is the dimensionless air–water partition coefficient.

Incorporating the Henry's law equation into Eq. (3), the mass balance for CB diffusion from liquid to air can be integrated with the boundary conditions $C_G(z = 0) = C_{G0}$ and $C_G(z = H) = C_{Gf}$, then gives:

$$\int_{C_{G0}}^{C_{Gf}} \frac{dC_G}{(C_L - \frac{C_G}{K_{iaw}})} = \frac{k_L \alpha A}{Q} \int_0^H dz \tag{5}$$

As $C_{G0} = 0$, $AH = V$ and C_L is constant in the integrating process, Eq.(5) takes the following explicit form:

$$C_{Gf} = K_{iaw} C_L \left[1 - \exp\left(\frac{-k_L \alpha V}{QK_{iaw}} \right) \right] \tag{6}$$

Then, by substituting Eq. (6) into Eq. (2), the integral form of Eq. (2), obtained with the boundary conditions $C_L(t = 0) = C_{L0}$ and $C_L(t = t) = C_{Lt}$, can be expressed as follows:

$$\int_{C_{L0}}^{C_L} \frac{dC_L}{C_L} = \frac{-QK_{iaw}}{V} \left[1 - \exp\left(\frac{-k_L \alpha V}{QK_{iaw}} \right) \right] \int_0^t dt \tag{7}$$

From Eq. (7) we have:

$$C_L = C_{L0} \exp\left\{ \frac{-QK_{iaw}}{V} \left[1 - \exp\left(\frac{-k_L \alpha V}{QK_{iaw}} \right) \right] t \right\} \tag{8}$$

Defining K_L as the overall mass transfer coefficient (min^{-1}), $K_L = k_L\alpha$, Eq. (8) can be expressed as follows:

$$C_L = C_{L0} \exp\left\{ \frac{-QK_{iaw}}{V} \left[1 - \exp\left(\frac{-K_L V}{QK_{iaw}} \right) \right] t \right\} \tag{9}$$

The predicted CB concentration in liquid were compared with the experimentally determined VOCs concentration, and K_L values were adjusted to minimize the sum of square of deviations. The best combination of K_L that gave the minimum sum of square of deviations was selected.

The effects of initial CB concentration, air flowrate and temperature on CB removal during the air stripping are separately presented in Figs. 1, 3 and 5. The lumped parameter model derived above was used to fit the measured data. Experimental results demonstrated that the simulated CB concentrations were correlated to the

Table 3
Air–water overall mass transfer coefficient for various initial CB concentrations (Conditions: air flowrate = 2.5 l min^{-1}, solution temperature = 15°C)

C_0 (mg·l^{-1})	K_L (min^{-1})	R^2
1	1.143 ± 0.106	0.994
3	1.321 ± 0.100	0.997
5	1.390 ± 0.129	0.997
7	1.432 ± 0.196	0.993
10	1.663 ± 0.262	0.995

measured results quite well ($R^2 > 0.99$), which implied that the lumped parameter model are potentially useful for predicting and interpreting mass transfer in air stripping. The distributions of K_L are summarized in Tables 3 and 4, and the order of magnitude of the obtained K_L was between 10^{-1} and 10^0 min^{-1}. The curve fitting approach adopted here can provide an accurate estimate of aqueous CB concentration during air bubbling.

3.2. Effect of initial CB concentration

The increase in initial CB concentration from 1 to 10 mg l^{-1} enhanced the removal of CB significantly during the air stripping (Fig. 1), as well as increased the K_L values from 1.143 to 1.663 min^{-1} (Table 3). The positive effect of increasing initial CB concentration is related to a large concentration gradient of CB concentration on the air–liquid interface, which is feasible for the diffusion of CB during air stripping. In addition, a linear relationship was found between the obtained K_L and the initial concentration of CB (Fig. 2), which is essential for choosing K_L under different pollution concentration in practical operation. However, Hassane et al. found that the overall mass transfer coefficient was mainly constant for initial feed chloroform concentrations ranging from 50 to 1000 ppm according to the study of mass transfer in the membrane air stripping process [8]. This result is inconsistent with the finding obtained above. This discrepancy may be caused by the imperfect reliability of the different models used to estimate the overall mass transfer coefficient of air stripping.

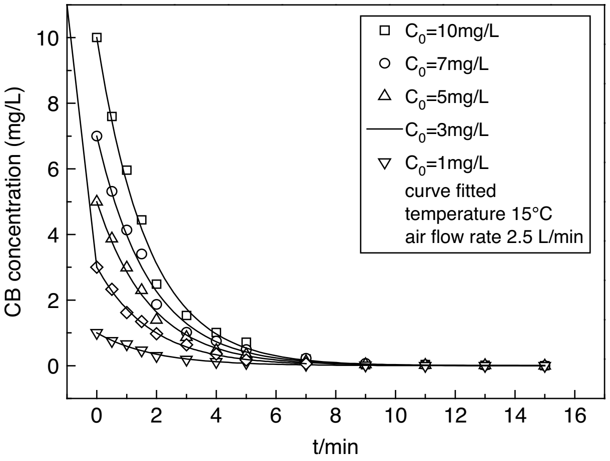

Fig. 1. Effluent of initial CB concentration.

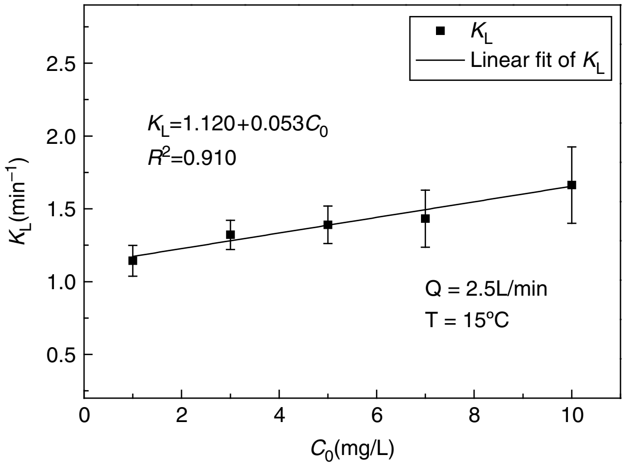

Fig. 2. The correlation of air–water mass transfer coefficient (K_L) with initial CB concentration (C_0).

3.3. Effect of air flowrate

Aqueous CB showed a higher CB removal efficiency during the air stripping with increasing air flowrate (Fig. 3). In comparison to the condition of flowrate at 2–3 min^{-1}, the effect of air flowrate on CB removal was more significant under a lower flowrate (1–2 l min^{-1}) operation, especially for higher temperature, ascribing to the nonlinear increasing of interfacial area while the air flowrate increased. Traditionally, the decreasing of bubble size (or the increasing in bubble concentration) would lead to an increase in air–aqueous interfacial area at lower air flowrate. However, frequency bubble collision at higher air flowrate condition would increase the diameter of air bubbles during the air stripping, as a result, the interfacial area does not linearly increased with an increase in air flowrate [16,17]. Several studies have also reported similar observation (VOCs removal vs. air flowrate) during the air stripping using conventional technologies [18–20]. For example, early work by Verma et al. [21] revealed that the removal efficiency of 1,2-dichloroethane from contaminated ground water was nonlinearly dependent on the air flowrate while the cascade cross flow and countercurrent air strippers were employed.

Another significant observation is that a roughly linear relationship was found between CB removal efficiency and air flowrate at low temperature of 0°C and 5°C (R^2 = 0.89). In comparison, the effects of flowrate on

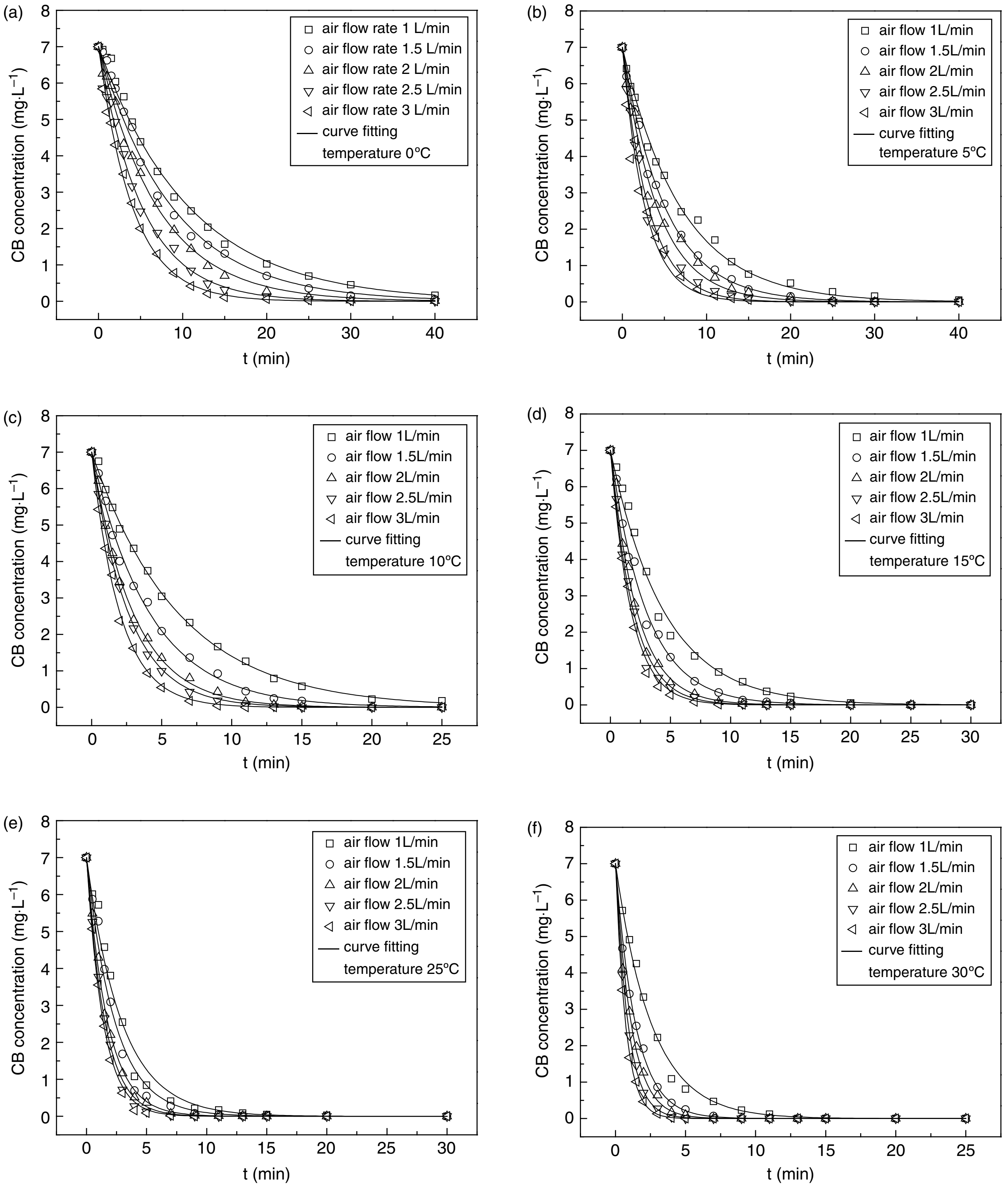

Fig. 3. Aqueous CB concentration profiles for various times during air stripping at various air flowrate (the initial CB concentration was 7 mg l^{-1}).

CB removal were not so significant at higher temperature (15–30°C), especially at higher flowrates. This result indicated that the removal of CB by air stripping at high temperature condition would reach a high level even at a relatively lower air flowrate, which is necessary to consider in the design and operation of air stripping system.

The estimated air–water mass transfer coefficients, K_L, for five air flowrates at six temperatures are given in Table 4. The correlation of K_L with air flowrate at varied temperatures is presented in Fig. 4. Similar to the effect of air flowrate on CB removal, K_L values are directly proportional to air flowrate (Table 4 and Fig. 4), which would be expected from the discussion above and previous studies [20]. Chao et al. [22] showed in a two-dimensional laboratory scale experiment that the air–water mass transfer coefficient was found to be directly proportional to the air flowrate. Since the total air–water interface surface area is proportional to the number and size of the air bubbles, an increase in air flowrate will result in an increase in the K_L values.

Table 4
The overall air–water mass transfer coefficients for air stripping experiments (conditions: air flowrate rang is 1–3 l min^{-1}, temperature range is 0–30°C, initial CB concentration 7 mg l^{-1})

Air flowrate (l min)	Temperature (°C)	K_L (min^{-1})	R^2
1	0	0.177	0.993
1.5	0	0.180	0.993
2	0	0.216	0.997
2.5	0	0.307	0.995
3	0	0.416	0.997
1	5	0.403	0.995
1.5	5	0.427	0.998
2	5	0.455	0.992
2.5	5	0.610	0.998
3	5	0.650	0.994
1	10	0.432	0.998
1.5	10	0.498	0.998
2	10	0.741	0.998
2.5	10	0.786	0.998
3	10	1.044	0.998
1	15	0.580	0.993
1.5	15	0.822	0.996
2	15	0.977	0.994
2.5	15	1.027	0.996
3	15	1.139	0.996
1	25	0.793	0.985
1.5	25	0.823	0.994
2	25	1.016	0.997
2.5	25	1.079	0.997
3	25	1.114	0.998
1	30	0.801	0.995
1.5	30	2.089	0.998
2	30	2.256	0.997
2.5	30	2.871	0.999
3	30	3.908	0.999

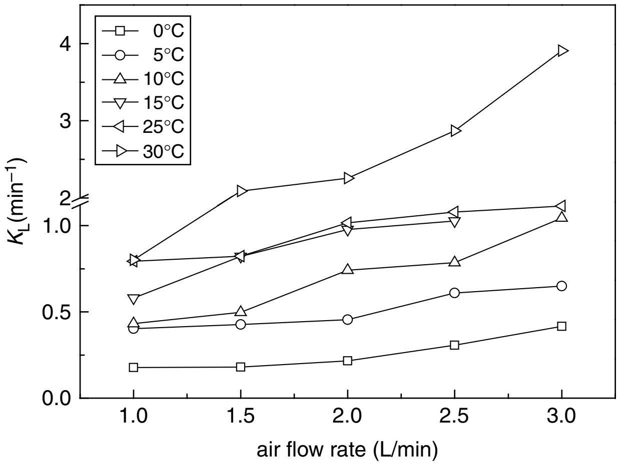

Fig. 4. The correlation of air–water mass transfer coefficient (K_L) with air flowrate at different temperature.

3.4. *Effect of temperature*

Experimental temperatures (ranged from 0°C to 30°C) was chosen in accordance with the operating conditions in waterworks. Fig. 5 presents the variation of the aqueous CB concentration as a function of stripping time for the given air flowrates at different temperatures. Results indicated that the removal efficiency of CB during air stripping increased with increasing temperature within the examined range at each air flowrate condition. Presented result confirmed the well-known and often experimentally observed increase in VOCs removal by stripping with the rise in aqueous temperature. In previous works, it was concluded that the increased removal of VOCs by stripping is mainly due to an increase of VOCs vapor pressure when temperature is increased [23–25]. Furthermore, It should be noted that low temperature condition (0–5°C) effected the overall CB reduction considerably at a given air flowrate, whereas less significantly for higher temperature conditions (15–30°C). This relationship between temperature variation and the amount of the removed contaminant was also observed by Simon et al. [26]. They concluded that higher temperature in the batch stripping of swine manure increases the removal efficiency, and that temperature difference of about 10 K, at higher temperature range, does not significantly influence the stripping efficiency.

The effects of temperatures on K_L, at different flowrates, are shown in Fig. 6. As expected, temperature plays an important role on mass transfer phenomena as K_L increases as temperature is increased (Fig. 6), which

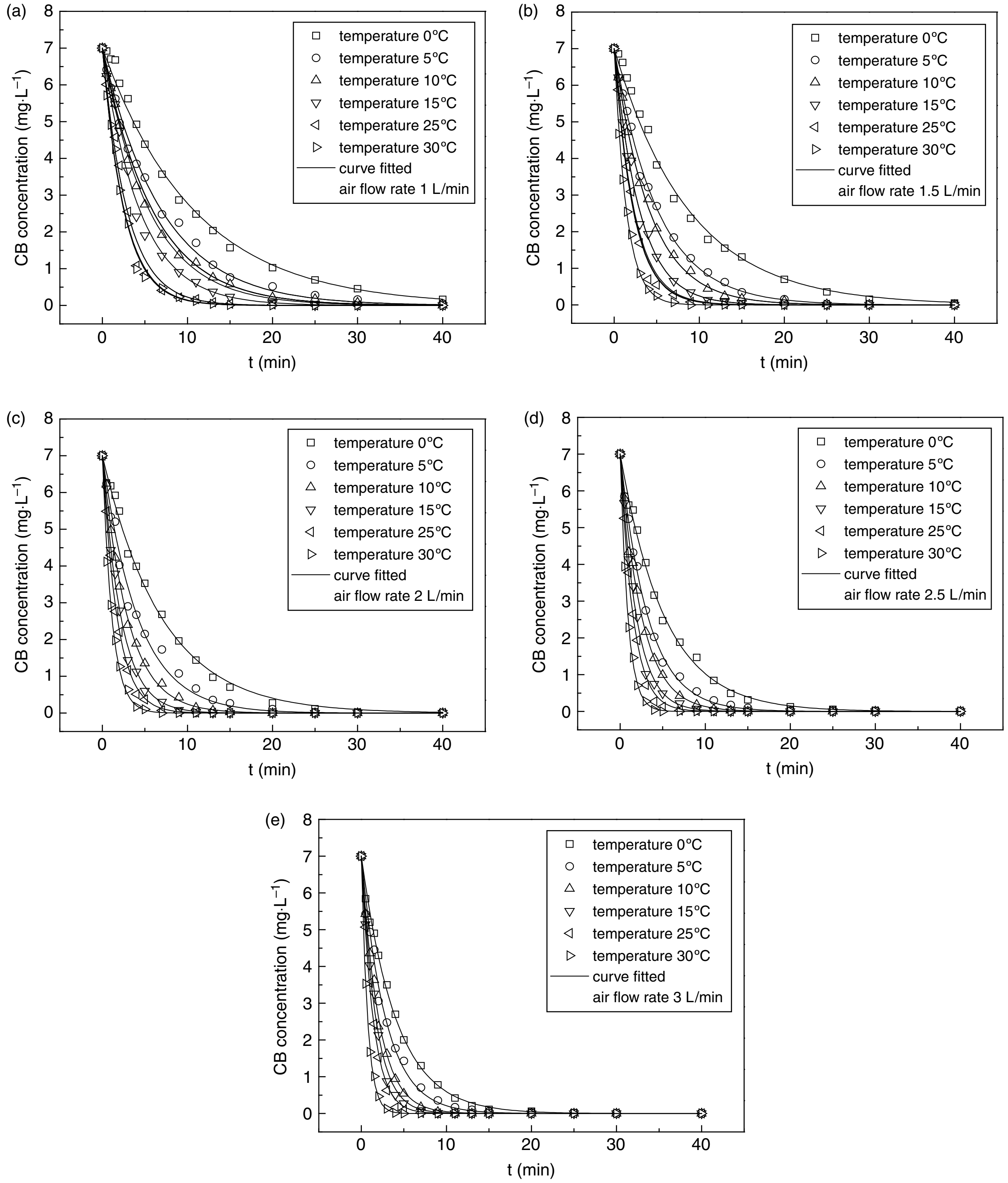

Fig. 5. Aqueous CB concentration profiles for various times during air stripping at different temperature (The initial CB concentration was 7 mg l^{-1}).

is more evident at higher temperature conditions (25–30°C). Previous studies revealed that the change of temperatures influences on the physical properties of both the liquid and gas, and has a significant impact on the mass transfer process [27]. For example, an increase in temperature would lead to a pronounced reduction of liquid viscosity and surface tension of air bubbles, resulting in (1) formation of small and stable bubbles and (2)

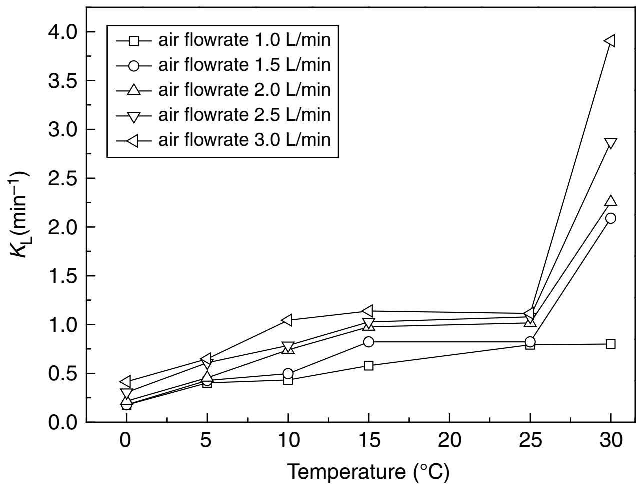

Fig. 6. The correlation of air–water mass transfer coefficient (K_L) with temperature at different air flowrate.

increases the probability of coalescence. It is this duality that alters the interfacial area and influences significantly on the mass transfer process [16,28]. In addition, the physical properties of the liquid and gas change with increasing temperature has also a great influence on diffusion coefficient. Viscosity decrease leads to a decrease in the thickness of the stagnant film at gas/liquid interface, resulting in a lower mass transfer resistance, and hence increases diffusion coefficient [27]. Both effects conduct to an increase in K_L.

Furthermore, it is interesting that a jump of K_L values was found at around 30°C for a given air flowrate as illustrated (Fig. 6), which indicates that stripping at higher temperature could achieve a higher CB removal efficiency even at lower air flowrate. As a result, the removal efficiency of CB could varied insignificant for different air flowrates stripping, which was similar to the discussion in Sect. 3.3. In addition, the effect of temperature on K_L values was more significant than that of air flowrate in the range from 1 to 3 l min^{-1}. In terms of practical implications, experimental results concerning temperature strongly suggested that the chosen of air flowrate is temperature dependent, is essential for the optimum design and operation of air stripping system.

4. Conclusions

Air stripping experiments were conducted to investigate the effects of initial CB concentration, air flowrate and temperatures on the mass transfer of CB. The following conclusions are drawn based on presented experimental results:

1. The lumped parameter model revealed that the K_L values ranged from 10^{-1} to 10^0 min^{-1} in this study, and K_L was found to be linearly proportional to initial CB concentration, but nonlinearly dependent on the air flowrate and temperature.
2. The effect of temperature on K_L values was more significant than that of air flowrate in the range of applicability in this study.
3. The effect of air flowrate on CB removal was less significant at higher temperature (15–30°C), especially for higher flowrates. Moreover, lower temperatures (0–5°C) has more significant effect on the overall CB reduction in comparison to the higher temperatures condition (15–30°C) at a given air flowrate.
4. The chosen air flowrate is temperature dependent for air stripping, which should be considered in the design and operation of air stripping system.

Acknowledgments

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