

탄화수소 누출로 인한 위험분석 평가 방법론 연구

A Methodology for Assessing Risk from Released Hydrocarbon

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ABSTRACT

This study presents a method for calculating the concentration of hydrocarbon releases in enclosed areas using empirical equations of evaporation rate. The approach of the method is to estimate the hydrocarbon exposure concentration in the air under conditions assumed. A methodology for assessing risk was suggested to individual risk assessment to exposed workers or others by probit expressions. The toxicity criteria and available human exposure data were examined and guidelines for risk assessment suggested for benzene-air and toluene-air systems. The value of probit constants with mole fractions of lethal concentrations in a mixture of hydrocarbons and a non-toxic substance was predicted. The probit values calculated with mole fractions can be used to estimate guidelines to prevent toxicity within enclosed working areas.

국문 요약

본 연구는 밀폐된 지역에서 경험적 증발속도식을 사용하여 누출된 탄화수소의 농도 계산방법을 소개하였다. 그 접근방법은 가정된 조건하에서 공기중의 탄화수소 누출 농도를 추정하여 노출된 근로자나 사람의 개인적 위험평가를 위해 probit 표현식에 의한 위험평가 방법론을 제시하였다. 벤젠-공기 및 톨루엔-공기계에 대해 독성기준과 사람의 노출데이터가 조사되어 졌고, 위험평가를 위한 가이드라인을 제시하였다. 탄화수소와 비독성 물질의 혼합물의 치사농도의 물분율에 따른 probit 상수값들을 예측하였다. 물분율에 따라 계산된 probit값은 밀폐된 작업구간에서 독성을 예방하기 위한 가이드라인을 추정하기 위해 사용될 수 있다.

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1. Introduction

Accidental releases of hazardous and toxic chemicals can present a serious risk to a worker's safety and to the environment in workplaces. Also, in spill incidents arising from open containers or storage tanks containing hydrocarbon fractions, it is important to know how long an explosive or flammable and toxic chemical concentration can exist. A particular concern is the generation of a vapour concentration from the release, which may cause health and safety hazards. To establish the exposure concentration, the source strength or the evaporation rate of the chemical substances must be determined. Evaporation rates are important to understand the process of evaporation from hydrocarbons spilled on water or on land, and the rate of evaporation of such hydrocarbons into the air.

The aim of this study is concerned with the estimates of the concentration of hydrocarbon releases in the open, such as a pool of volatile liquid within an enclosed working area. A further aim of the study is to develop the methodology for undertaking a risk analysis from hydrocarbon spills or releases using existing mass-transfer coefficient equations which consider temperature, spill area and ventilation rate or wind speed. The hydrocarbon exposure concentration is estimated in the air from empirical equations of the evaporative rate. We suggest a methodology for individual risk assessment of exposed workers or others by the use of probit expressions. The probit analysis can be used in quantitative risk analysis to predict the effect of toxic exposure.

2. Equations of the Mass Transfer Coefficient and the Rate of Evaporation

Knowledge of rate of evaporation is important both from of the practical viewpoint of cleaning up spills and for developing predictive models. The molar flux of an evaporative component *i* from a hydrocarbon spill is generally given as¹⁾:

$$N_i = \frac{K_m(P_i - P)}{RT} \dots\dots\dots (1)$$

where *N_i* is the evaporative molar flux of *i* [mol/m² sec], *K_m* is the overall mass-transfer coefficient [m/sec], *R* is the universal gas constant [8.314 Pa m³/mol K], *T* is the absolute temperature of the bulk air [K], *P_i* is the hydrocarbon vapor pressure of component *i* at the interface [Pa], and *P* is the hydrocarbon partial pressure in the atmosphere, which is usually taken as zero far from the spill.

Mackey and Matsugu²⁾ collected evaporation data for cumene. The data were used to correlate the gas-phase mass-transfer coefficient as a function of wind speed and pool size by the equation.

$$K_m = 0.0292 U^{0.78} X^{-0.11} Sc^{-0.67} \dots\dots\dots (2)$$

where *K_m* is the mass-transfer coefficient, *U* is the wind speed, *X* is the pool diameter or the scale size of the evaporation area, and *Sc* is the Schmidt number. They noted that the evaporation process is more complex for hydrocarbon mixtures, being dependent on the liquid diffusion characteristics with a liquid-phase diffusion resistance being present.

Reed³⁾ reported on the development of an evaporation equation from oil spills. He used the expression of previous workers²⁾ to approach the estimate of the mass-transfer coefficient.

$$K = 0.029 W^{0.78} D^{-0.11} Sc^{-0.67} \sqrt{(M_w + 29)/M_w} \dots\dots\dots (3)$$

where *K* is the mass transfer coefficient, *W* is the wind speed [m/hr], *D* is the spill diameter [m], *Sc* is the Schmidt number [-],

and M_w is the molecular weight of the vapor.

Fingas⁴⁾ has reviewed the physics and predictive modeling of oil spill evaporation. He noted that most workers used boundary layer equations adapted from water evaporation work. These equations predict a constant evaporation mass transfer rate dependent on scale size and wind speed. He also reported that there are only three frequently used schemes currently employed in models. The most commonly used method is that of evaporation exposure as proposed by Stiver and Mackay⁵⁾. One form of the model requires a constant mass-transfer coefficient and a vapor pressure for each oil fraction. The second most commonly used method is that of applying fractionated cut data. This method is applied by using the distillation curves to estimate parameters for the Makay equations. The third most common method is to assume a loss rate that is estimated from the oil property's data and presumes that the loss progresses linearly or logarithmically with time.

The evaporation rate of a volatile liquid such as hydrocarbon substance is expected to be a function of the saturation vapor pressure. A more generalized expression for the evaporation rate of a liquid into stagnant air from equation (1) is rewritten by :

$$E_m = \frac{M_w K A (P^0 - P)}{RT} [kg/sec] \dots\dots\dots (4)$$

where E_m is the evaporation rate [kg/sec], M_w is the molecular weight of the volatile substance [kg/kg-mol], K is the mass transfer coefficient [m/sec], P^0 is the vapor pressure of the substance [kPa], P is the partial pressure of the substance in air [kPa], R is the universal gas constant [8.314 kPa m³/kg-mol K], and T is the temperature of the liquid [K]. For many cases, P^0 is much greater than P . the equation is reduced to :

$$E_m = \frac{M_w K A P^0}{RT} [kg/sec] \dots\dots\dots (5)$$

3. Model to Estimate the Concentration of Hydrocarbon in an Enclosed Area

The best method to determine the concentration of the amount of volatile vapors is to measure the vapor concentration directly. Estimates of the vapor concentrations of hydrocarbon are frequently required in risk analysis in open systems such as a pool of volatile liquid, an opening to a storage tank or an open container of volatile liquid within working areas. The concentration of hydrocarbon in a small enclosed place can be calculated from a mass balance shown in Fig. 1. The mass balance of the volatile hydrocarbon species is as follows :

$$V \frac{dC}{dt} = E_m - M_f Q_v C \dots\dots\dots (6)$$

where Q_v is the ventilation flow rate of air [m³/sec], V is the specific enclosure volume [m³], E_m is the evaporation rate of hydrocarbon [kg/sec], A is the pool area [m²], C is the local concentration of volatile hydrocarbon into the air [kg/m³], and M_f is the non-ideal mixing factor [-].

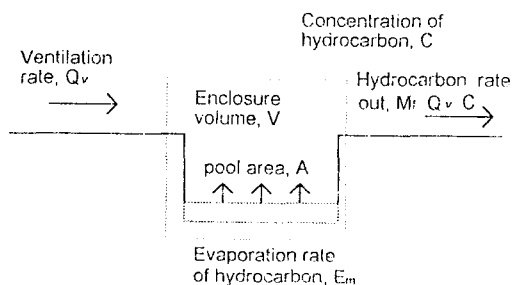


Fig. 1 Mass balance on volatile hydrocarbon in enclosure area

Since at steady state the accumulation term is zero, if the vaporization rate and the volumetric flow rate of air are assumed to be constant with time, The equation is reduced

to C,

$$C = \frac{E_m}{M_f Q_v} [kg/m^3] \dots\dots\dots (7)$$

The mixing factor M_f determined by tracer gas techniques. Ideal or perfect mixing takes place when M_f is 1, the non-ideal mixing factor ranges from 0.1-0.5 for most practical situations⁶⁾.

To obtain the more convenient concentration C in ppm, the application of the ideal gas law parameters substituted in equation (7) yields the following equation :

$$C = \frac{E_m RT}{M_f Q_v PM_w} \times 10^6 [ppm] \dots\dots\dots (8)$$

This equation (8) includes the following assumptions. The condition assumed is steady state and not considered heat transfer around the chemical pool, including the effect of ground conduction. The concentration calculated from this equation could be both a minimum average concentration and a maximum average concentration estimated in the specific volume of the air, depending upon the mixing factor. Also, the equation (8) is used to calculate the ventilation rate required to sweep an enclosed area.

To estimate the concentration from equation (7), it is the most important know how to obtain the rate of evaporation. The rate of vaporization of volatile liquid such as hydrocarbons is expected to be a function of the saturation vapor pressure. An expression for the evaporation rate of a liquid into stagnant air is given by equation (4) or (5) in the previous section. These equations were used to calculate the evaporation rate or a source term rate of volatile from open vessel or from liquid releases where the material is stagnant. To estimate the concentration in ppm of volatile substance, equations (5) and (8) can be combined, and equation (9) reduces to :

$$C = \frac{KAP^o}{M_f Q_v P} \times 10^6 [ppm] \dots\dots\dots (9)$$

To obtain the concentration of volatile from hydrocarbon releases from equation (9), the mass-transfer coefficient can be calculated based on the gas-phase mass-transfer, which can be estimated using the Thibodeaux's relationship⁷⁾. The mass-transfer coefficient for one species, K , may be related to that for another K_{ref} by :

$$\frac{K}{K_{ref}} = \left(\frac{D}{D_{ref}} \right)^n \dots\dots\dots (10)$$

All other factors included in K_{ref} for reference species, such as fluid type, temperature, flow rate, location, depth, and so on, must remain constant for one species. where D is the gas-phase diffusion coefficient and n is a constant. Values of n can be derived from the mass transfer theories and suggested $n=1$ for stagnant-film model and $n=2/3$ for boundary layer theory. The gas-phase coefficients can be estimated from the molecular weights of a reference substance and a volatile substance⁸⁾ as follows :

$$\frac{D}{D_{ref}} = \sqrt{\frac{M_{ref}}{M}} \dots\dots\dots (11)$$

Equations (10) and (11) can combined with molecular weights terms for the stagnant-film model and the boundary layer theory, respectively.

For the stagnant-film model,

$$K = K_{ref} \left(\frac{M_{ref}}{M} \right)^{1/2} \dots\dots\dots (12)$$

For the boundary layer theory

$$K = K_{ref} \left(\frac{M_{ref}}{M} \right)^{1/3} \dots\dots\dots (13)$$

where water with a mass-transfer coefficient of 8.3×10^{-3} m/sec at 25°C as a reference substance (K_{ref}) was used^{6,9)}.

To calculate the gaseous diffusion coefficients, empirical correlations to predictions of

the diffusion coefficient have been developed by many authors. In this study, empirical relations as given by Fuller *et al.*^{10,11)} were used :

$$D = (10^{-3}) \frac{T^{1.75}(1/M_1 + 1/M_2)^{1/2}}{p[(\sum_i V_{i1})^{1/3} + (\sum_i V_{i2})^{1/3}]^2} \quad [cm^2/sec] \dots\dots\dots (14)$$

in which T is in degrees Kelvin, p is in atmospheres, and the V_{ij} are the molar volumes of substance j . The vapor pressure p^* of the hydrocarbon with temperature was calculated from the Antoine equation¹²⁾ as follows :

$$\ln(p^*) = A - \frac{B}{C+T} \quad \dots\dots\dots (15)$$

where A, B, C are constants for each substance, and T is the temperature in Kelvin.

3.1 The Probit Approach for Risk Analysis

The use of safety and risk assessment studies has improved considerably over recent years. A quantitative risk analysis is one of the tools that can be used to assess the risk of an industrial accident. A risk analysis generally consists of the following steps¹³⁾ :

- 1) Identification of potential hazards for releases of hazardous materials,
- 2) Calculation of physical effects of the those releases,
- 3) Calculation of consequences due to those effects,
- 4) Calculation of the probabilities of steps 1-3,
- 5) Final risk evaluation: combine consequences and probabilities and give risk-reducing recommendations for determination of accident scenarios.

It is common in quantitative risk analysis to evaluate toxicity risks by calculation of the percentage of the population exposed that show a certain health effect. The probit model

is widely used in risk analysis to predict the probability of death or injury associated with the human response to an exposure to excess energy or toxic materials. The probit equation is a mathematical relationship between the response fraction and the probit(=probability unit), which takes into account that the response is non-linear. The probit variable Y is related to the probability P given by¹⁴⁾ :

$$P = \frac{1}{(2\pi)^{1/2}} \int_{-\infty}^{Y-5} \exp\left(-\frac{u^2}{2}\right) du \quad \dots (16)$$

This equation provides a relationship between the probability P and the probit variable Y . The probit relationship transforms the sigmoid shape of the normal response versus dose curve into a straight line when plotted using a linear probit scale. For individual releases, the probit approach is particularly suited and the probability of a given impact can be obtained using a probit expression.

The probit equations provide a means of relating the intensity of the toxic exposure to the degree or percentage response of the objects being exposed. The probit equation takes the form¹⁵⁾ :

$$Pr = a + b \ln(V) \quad \dots\dots\dots (17)$$

where Pr is the probit to a measure of the percentage of the vulnerable resource affected, and V is the independent variable as a function of the factor that causes injury or damage to the vulnerable resource, a, b are regression coefficients which are generally estimated from empirical data. For a toxic exposure V is the toxic load, which is expressed in terms of concentration and duration of exposure. The toxic load can be calculated by following equation :

$$Toxicload = \int_0^t [C(t)]^n dt \quad \dots\dots\dots (18)$$

where $C(t)$ is the concentration of the toxic substance as a function of time, n is a constant which is a function of specified

species, t is the exposure time. The toxic load is found to be described by a non-linear function of the form $(C^n t)$. For the concentration released is constant with time, the usual form when applied to toxic exposure situation is:

$$Pr = a + b \ln(C^n t) \dots\dots\dots (19)$$

where C is the concentration of the released vapor [ppm or mg/m³] and t is the time duration of the exposure [min].

4. Calculations and Estimation Method

The hydrocarbons chosen benzene and toluene in this study were selected as examples of assessing exposure to toxic vapors. Some physical properties such as a temperature, ventilation rate, and vapor pressure and pool area were estimated to calculate the rate of evaporation, and we could calculate the minimum and maximum concentrations. The parameters assumed for the risk analysis are as follows: the temperature is 5 to 30°C, the ventilation rate is 0.5 to 3m/sec, and the pool size on a rectangular basis is 0.75 to 2.0m².

The mass-transfer coefficient can be calculated from equations (2), (3), or (10) by the empirical equations of their author's^{2,3,6-9)}. The released concentration of hydrocarbon into the air could be estimated from the evaporation equation (5) and equations (8) or (9). A risk assessment of toxicity from released or spilled hydrocarbons was estimated by using the probit approach method as above mentioned.

5. Results and Discussion

5.1 Estimation of the Released Concentration of Benzene and Toluene

Knowledge of the mass-transfer coefficient

is important to estimate the concentration of the hydrocarbons. In this study, the mass-transfer coefficient was calculated by the empirical equations mentioned in previous section. Various methods of calculating the mass-transfer coefficient were compared. The mass-transfer coefficient calculated by the boundary layer theory in the equations was the largest value. Because a worst case must reflect an 'upper bound' of a wide range of conditions expected in the workplace and yet be rational and realistic about those conditions, the largest values for a mass-transfer coefficient are employed in a risk assessment, also, due to a maximum probability of fatality on a worker for a given exposure. Accordingly, the mass-transfer coefficient calculated by the boundary layer theory was used. The minimum and maximum concentrations of hydrocarbons with parameters were calculated from equations (9) and (13).

Fig. 2 shows typically the steady-state concentration variations with temperature in benzene-air and toluene-air systems at a constant pool area of 1.5m² and a constant ventilation rate of 1m/sec. The minimum concentration of both benzene and toluene is not affected relatively by an increase with rising

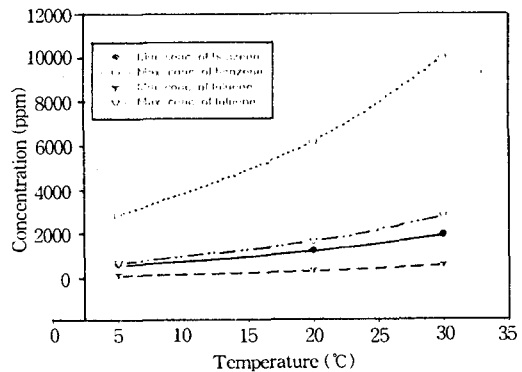


Fig. 2 Steady-state concentration variation with temperature in benzene-air and toluene-air system (pool area=1.5m², ventilation rate=1m/sec)

the temperature, but the maximum concentration of benzene increases sharply with temperature more than with toluene. It is due to the higher vapour pressure of benzene compare with toluene. Fig. 3 shows typically the steady-state concentration variations with the ventilation rate at a constant temperature of 20°C and a constant pool area of 2m². The concentrations of benzene and toluene are diminished with an increase of the ventilation rate, because of the displacement of vapour. As shown in Fig. 3, since an increment of the ventilation rate can reduce the generated concentration, it is useful to remove the contaminant before workers are exposed. The concentration variation with the pool area at a constant temperature of 20°C and a ventilation rate of 1m/sec was typically shown in Fig. 4. The hydrocarbon concentration increases linearly with increasing the pool area. A maximum concentration of benzene shows a high linear variation with the pool area due to the higher vapour pressure. As discussed above, the hydrocarbon concentration generated in an enclosed working area can be estimated and expressed in terms of the mixing factor and the evaporation rate as a function of temperature, ventilation rate and pool area.

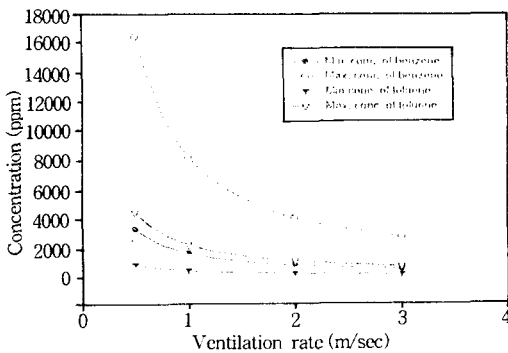


Fig. 3 Steady-state concentration variation with ventilation rate in benzene-air and toluene-air system (temperature=20°C, pool area=2m²)

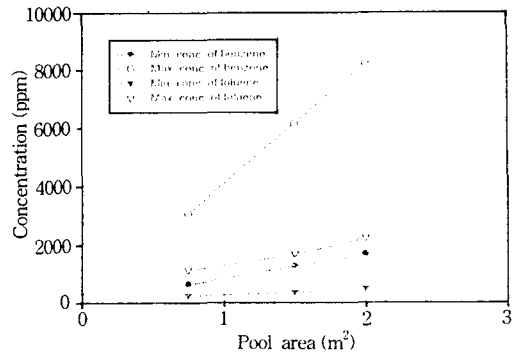


Fig. 4 Steady-state concentration variation with pool area in benzene-air and toluene-air system (temperature=20°C, ventilation speed=1m/sec)

5.2 Approaches to Estimating Toxicity Data of Exposure

The probit equation for toxic assessment is highly dependent on the type of toxic substance released. Probit constants in its equations are known for only a limited number of substances, and are mainly based on animal toxicity data. The Center for Chemical Process Safety (CCPS) and the Netherlands Organization of Applied Scientific Research (TNO) provide values of the constants in the probit expression¹⁶⁾. Some examples of the CCPS probit constants are listed in Table 1^{15,17)}.

Table 1 The CCPS probit constants examples for the lethal toxicity

Substance	a	b	n
Acrolein	-9.931	2.049	1.00
Ammonia	-35.900	1.850	2.00
Benzene	-109.780	5.300	2.00
Bromine	-9.040	0.920	2.00
Carbon tetrachloride	-6.290	0.408	2.50
Chlorine	-8.290	0.920	2.00
Hydrogen cyanide	-29.420	3.008	1.43
Hydrogen sulfide	-31.420	3.008	1.43
Phosgene	-19.270	3.686	1.00
Sulfur dioxide	-15.670	2.100	1.00
Toluene	-6.794	0.408	2.50

Probit equations are established using a

series of toxicity data for a specific health effect and exposure route and preferably for one species. Toxicity data have to be obtained for a range of exposure times and concentrations. In this study, the probit variables for the benzene and toluene are calculated by the CCPS probit constants as shown in Table 1.

Fig.5 and Fig.6 show the plot of probit variable versus concentration with time in benzene-air and toluene-air systems under experimental conditions, respectively. The most often available endpoint was the Lethal Concentration Low (LCLo). In applying the LCLo, the question needs to be resolved of what percentages response it can be considered to represent¹⁸⁾. We assumed it represented 0.1% and 1.0% exposure for fatality due to get the Immediately Dangerous to Life and Health (IDLH) level¹⁹⁾. In figures 5 and 6, the solid straight lines present transformation percentages of fatality of 0.1% and 1.0% by probit values, respectively. The predicted fatality using probit equations could be obtained from Fig.5 and Fig.6. The developed toxicity criteria and available human exposure data were examined and guidelines for workers suggested in shown Table 2. The sign '<' denotes concentration less than the values. The generated concentration can be controlled to

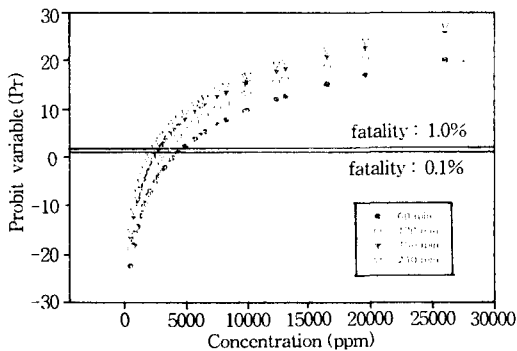


Fig. 5 Plot of probit variable versus concentration with time in benzene-air system

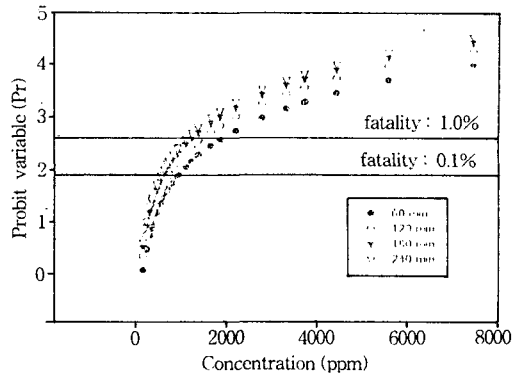


Fig. 6 Plot of probit variable versus concentration with time in toluene-air system

Table 2 Guidelines' examples for worker's toxicity from predicted exposure

Substance	Value of Concentration [ppm]	Time [min]	Predicted % fatality for stated conditions
Benzene	5227	60	<1.0
	4864	60	<0.1
	3696	120	<1.0
	3439	120	<0.1
	3018	180	<1.0
	2808	180	<0.1
	2614	240	<1.0
	2432	240	<0.1
Toluene	2088	60	<1.0
	988	60	<0.1
	1583	120	<1.0
	748	120	<0.1
	1346	180	<1.0
	636	180	<0.1
	1200	240	<1.0
	567	240	<0.1

reduce toxicity by ventilation with fresh air. The guidelines can be used to estimate the exposure concentration into the air for a worker standing near a pool of volatile hydrocarbon.

5.3 Probit Expression Constants for Mixture of Hydrocarbon and a Non-toxic Substance

The values of constants in probit equations are given for pure substances by the CCPS or

TNO. Values of probit constants are not given for a mixture of a toxic and non-toxic or binary mixtures. As mentioned in the previous section, a ventilation system to remove the contaminant before workers are exposed may be effective. Air can usually be used a displacement. If a non-toxic substance in air mixture exists, the probit constants to estimate toxicity are needed to adjust the constant. The probit values calculated from the probit equation (19) is the overall concentration of the mixture in the air - not the concentration of pure toxic substance. The probit equation in terms of overall mixture concentration is written as follows :

$$Pr = a + b [(C_{mix} x)^n t] \dots\dots\dots (20)$$

where C_{mix} is the total concentration of the mixture in air and x is the mole fraction of toxic substance within the mixture. The mixture probit equation can be rearranged to :

$$Pr = a_{mix} + b \ln (C_{mix}^n t) \dots\dots\dots (21)$$

$$\text{where } a_{mix} = a + b \ln (x^n) \dots\dots\dots (22)$$

The value of the probit constant depends on the mole fraction of toxic substance in the mixture and can be calculated using the equation (22). But the values of the toxic constants b and n are independent of the relative concentration of the toxic substance in the mixture. That is same values of b and n in the pure toxic substance component of the mixture.

Table 3 shows the probit constants with the mole fraction for mixture of a hydrocarbon and a non-toxic substance. The probit constants can be used as part of estimating the fatality of the toxic exposure for the mixture of hydrocarbon and non-toxic substance. Using the probit constants in Table 3, the comparison of values of the probit variables with mole fraction to mixture fatality was examined numerically and the typical

probit variables with mole fraction of concentration selected for toluene at exposure time of 60min. is shown in Table 4. The value of the probit variables was decreased with decreasing mole fraction than those of pure substance, resulting in the diluted concentration. We used the probit constants with mole fraction in shown Table 3 and calculated to assess the likelihood of a fatality of a worker. The probit values calculated with mole fraction can be decided the guidelines of the toxicity within an enclosed working area.

Table 3 Probit constants with mole fraction for mixture of hydrocarbon

Substance	Mole fraction	a	b	n
Benzene	0.1	-134.187	5.300	2.00
	0.2	-126.840	5.300	2.00
	0.3	-122.542	5.300	2.00
	0.4	-119.493	5.300	2.00
	0.5	-117.127	5.300	2.00
	0.6	-115.195	5.300	2.00
	0.7	-113.561	5.300	2.00
	0.8	-112.145	5.300	2.00
	0.9	-110.896	5.300	2.00
	1.0	-109.780	5.300	2.00
Toluene	0.1	-9.143	0.408	2.50
	0.2	-8.436	0.408	2.50
	0.3	-8.022	0.408	2.50
	0.4	-7.729	0.408	2.50
	0.5	-7.501	0.408	2.50
	0.6	-7.315	0.408	2.50
	0.7	-7.158	0.408	2.50
	0.8	-7.022	0.408	2.50
	0.9	-6.901	0.408	2.50
	1.0	-6.794	0.408	2.50

Table 4 Comparison of values of the probit variables with mole fraction and Concentration selected for toluene at exposure time of 60min

Concentration [ppm]	Probit variables / Mole fraction				
	0.2	0.4	0.6	0.8	1.0
4436	1.80	2.51	2.92	3.21	3.44
2218	1.09	1.80	2.21	2.51	2.73
1109	0.39	1.09	1.51	1.80	2.03
739	-0.3	0.68	1.09	1.39	1.61

6. Conclusions

The simple model to estimate the concentration of hydrocarbon generated in an enclosed working area described. The model could be expressed in terms of the mixing factor and the evaporation rate as a function of temperature, ventilation rate and pool area. The fatality of 0.1% and 1.0% by transformation percentages of probit variables predicted for benzene-air and toluene-air systems. The toxicity criteria and available human exposure data were examined and suggested guidelines for workers. The comparison of values of the probit constants with mole fractions for mixture of hydrocarbon and a non-toxic substance was examined numerically and obtained the typical probit variables with mole fraction of concentration. The probit values calculated with mole fraction can used to be decided the guideline for the toxicity within enclosed working area.

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