

## Review

# A literature review of the physics and predictive modelling of oil spill evaporation

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### Abstract

The literature on the physics and mathematical modelling of oil spill evaporation is reviewed. Two basic approaches to the mechanism of evaporation are proposed in the literature, first-order decay and boundary-layer limited. Several workers propose a first-order decay process which yields a logarithmic decrease in evaporation with time. Most workers use boundary-layer equations adapted from water evaporation work. These equations predict a constant evaporation mass transfer rate dependent on scale size and wind speed. The implementation of these equations in oil spill models is reviewed. Three primary approaches are adopted: use of a simplified logarithmic predictor, direct use of a boundary-layer model and use of a fractionated cut model. The last uses readily available distillation data and estimations of how each cut evaporates. A comparison of experimental data and prediction methods shows that the accuracy is very dependent on the particular oil properties.

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### 1. Introduction

Evaporation is the most important process that oils undergo after spillage. In a few days, light crudes or refined products can lose up to 75% of their volume. An understanding of evaporation is important both from the practical viewpoint of cleaning up spills and for developing predictive models. Many spill models incorporate evaporation as a component of their prediction. Evaporation is sometimes the only transformation process included in oil spill prediction models.

Although the process of evaporation of crude oils is basically simple, the application of evaporation equations in spill models is sometimes difficult. This is primarily a result of the paucity of input data required to make many of the equations operative.

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There are only three frequently used schemes currently employed in models. The most commonly used is that of evaporative exposure as proposed by Stiver and Mackay [1]. Difficulties with the implementation of this model are primarily with the input data. One form of the model requires a constant mass transfer coefficient and a vapour pressure for each oil. These parameters are not routinely measured for oil and must be estimated using other techniques. The second most commonly used method is that of applying fractionated cut data. The distillation curves are readily available. This method is applied by using the distillation curves to estimate parameters for the Mackay equations noted above or as a direct technique. The third most common method is to assume a loss rate which is estimated from the oil properties data and presume that the loss progresses linearly or logarithmically.

The basis for most of the evaporative work are the extensive studies on the evaporation of water [2, 3]. In fact, the currently used equations still employ portions of these equations. There may be several fundamental differences between the evaporation of a pure liquid such as water and for a multi-component system such as crude oil.

## 2. Physics and chemistry of oil evaporation

In 1964 Blokker [4] was the first to develop separate oil evaporation equations for oil evaporation at sea. His starting basis was theoretical. Oil was presumed to be a one-component liquid. The ASTM (American Society for Testing and Materials) distillation data and the average boiling points of successive fractions were used as the starting point to predict an overall vapour pressure. The average vapour pressure of these fractions was then calculated from the Clausius–Clapeyron equation to yield

$$\log \frac{p_s}{p} = \frac{qM}{4.57} \left( \frac{1}{T} - \frac{1}{T_s} \right), \quad (1)$$

where  $p$  is the vapour pressure at the absolute temperature  $T$ ;  $p_s$  is the vapour pressure at the boiling point  $T_s$  (for  $p_s$ , 760 mmHg was used),  $q$  is the heat of evaporation ( $\text{cal}^{-1} \text{g}$ ) and  $M$  is the molecular weight. The term  $qM/(4.57T_s)$  was taken to be nearly constant for hydrocarbons ( $= 5.0 \pm 0.2$ ) and thus the expression was simplified to

$$\log p_s/p = 5.0[(T_s - T)/T]. \quad (2)$$

From the data obtained the weathering curve was calculated, assuming that Raoult's law is valid for this situation and this gives  $qM$  as a function of the percentage evaporated. Pasquill's equation was applied stepwise, and the total evaporation time obtained by summation:

$$t = \frac{\Delta h D^{\beta}}{K_{ev} u^{\alpha}} \sum \frac{1}{pM}, \quad (3)$$

where  $t$  is the total evaporation time (h),  $\Delta h$  is the decrease in layer thickness (m),  $D$  is the diameter of the oil spill,  $\beta$  is a meteorological constant and is assigned a value of 0.11,  $K_{ev}$  is a constant for atmospheric stability and is taken to be  $1.2 \times 10^{-8}$ ,  $u$  is the wind speed ( $\text{m s}^{-1}$ ),  $\alpha$  is a meteorological constant and is assigned a value of 0.78,  $p$  is the vapour pressure at the absolute temperature  $T$  and  $M$  is the molecular weight of the component or oil mass.

A small wind tunnel was constructed and this equation tested against the evaporation of gasoline and a medium crude oil. The observed gasoline evaporation rate was much higher than was predicted and the crude oil rate was much lower than predicted. The times of evaporation, however, were relatively close and the equation was accepted for further use. The above equations were then incorporated into spreading equations to yield equations to predict the simultaneous spreading and evaporation of oil and petroleum products.

In 1973 Mackay and Matsugu [5] approached the problem by using the classical water evaporation and experimental work. The water evaporation equation was corrected to hydrocarbons using the evaporation rate of cumene. It was noted that the difference in constants was related to the enthalpy differences between water and cumene. Data on the evaporation of water and cumene have been used to correlate the gas phase mass transfer coefficient as a function of wind-speed and pool size by the equation

$$K_m = 0.0292U^{0.78}X^{-0.11}Sc^{-0.67}, \quad (4)$$

where  $K_m$  is the mass transfer coefficient in units of mass per unit time,  $U$  is wind speed, note that the power of 0.78 is the classical water evaporation-derived coefficient,  $Sc$  is the Schmidt number which is a dimensionless number representing the viscosity ratio of the evaporating material and air and  $X$  is the pool diameter or the scale size of evaporating area. Mackay and Matsugu noted that for hydrocarbon mixtures the evaporation process is more complex, being dependent on the liquid diffusion characteristics, a liquid phase diffusion resistance being present. Experimental data on gasoline evaporation were compared with computed rates. The computed rates showed fair agreement and suggest the presence of a liquid phase mass transfer resistance.

This work was subsequently extended by the same group [6] to show that the evaporative loss of a mass of oil spilled can be estimated using a mass transfer coefficient as shown above. This approach was investigated with some laboratory data and tested against some known mass transfer conditions on the sea. The conclusion was that this mass transfer approach could result in predictions of evaporation at sea.

In 1976 Butler [7] developed a model to examine evaporation of specific hydrocarbon components. The weathering rate was taken as being proportional to the equilibrium vapour pressure  $P$  of the compound and to the fraction remaining:

$$dx/dt = -kP(x/x_0), \quad (5)$$

where  $x$  is the amount of a particular component of a crude oil at time  $t$ ,  $x_0$  is the amount present of that same component at the beginning of weathering ( $t = 0$ ),  $k$  is an empirical rate coefficient and  $P$  is the vapour pressure of the oil component. Since petroleum is a complicated mixture of compounds,  $P$  is not equal to the vapour pressure of the pure compound, but neither would there be large variations in the activity coefficient as the weathering process occurs. For this reason, the activity coefficients were subsumed in the empirical rate coefficient  $k$ .  $P$  and  $k$  were taken as independent of the amount  $x$  remaining over a fairly wide range of oils. The equation was then directly integrated to give the fraction of the original compound remaining after weathering:

$$x/x_0 = \exp(-ktP/x_0). \quad (6)$$

The vapour pressure of individual components was fit using a regression line to yield a predictor equation for vapour pressure:

$$P = \exp(10.94 - 1.06N), \quad (7)$$

where  $P$  is the vapour pressure (Torr) and  $N$  is the carbon number of the compound in question. This combined with Eq. (6) yielded the following:

$$x/x_0 = \exp[-(kt/x_0)\exp(10.94 - 1.06N)], \quad (8)$$

where  $x/x_0$  is the fraction of the component left after weathering,  $k$  is the empirical constant,  $t$  is the time,  $x_0$  is the original quantity of the component and  $N$  is the carbon number of the component in question.

Eq. (8) predicts that the fraction weathered at a given time decreases more than exponentially with increasing carbon number. If the initial distribution of compounds is essentially uniform ( $x_0$  independent of  $N$ ), then the above equation predicts that the carbon number where a constant fraction (e.g. half) of the initial amount has been lost ( $x = 0.5x_0$ ) is a logarithmic function of the time of weathering:

$$N_{1/2} = 10.66 + 2.17 \log(kt/x_0), \quad (9)$$

where  $N_{1/2}$  is half the volume fraction of the oil.

The equation was tested using data from some patches of oil on the shoreline, whose age was known. The equation was able to predict the age of the samples relatively well. It was suggested that the equation was applicable to open water spills; however this was never subsequently applied in models.

Yang and Wang [8] developed an equation using the Mackay and Matsugu molecular diffusion process. The vapour phase mass transfer process was expressed by

$$D_{ie} = k_m(p_i - p_{i\infty})/[RT_s], \quad (10)$$

where  $D_{ie}$  is the vapour phase mass transfer rate,  $k_m$  is a coefficient that lumps all the unknown factors that affect the value of  $D_{ie}$ ,  $p_i$  is the hydrocarbon vapour pressure of fraction  $i$  at the interface,  $p_{i\infty}$  is the hydrocarbon vapour pressure of fraction  $i$  at infinite altitude of the atmosphere,  $R$  is the gas constant and  $T_s$  is the absolute temperature of the oil slick.

The following functional relationship was proposed:

$$k_m = aA^\gamma e^{qU}, \quad (11)$$

where  $A$  is the slick area,  $U$  is the overwater wind speed and  $a$ ,  $q$  and  $\gamma$  are empirical coefficients. This functional relationship was based on the results of past investigations, including, for instance, those of MacKay and Matsugu [5] who suggested the value of  $\gamma$  to be in the range from  $-0.025$  to  $-0.055$ . Further experiments were performed by Yang and Wang to determine the values of  $a$  and  $q$ . The results were found to be twofold. Experiments showed that a film formed on evaporating oils and that this film severely retarded evaporation. Before the surface film has developed ( $\rho_i/\rho_0 < 1.0078$ ),

$$K_{mb} = 69A^{-0.0055} e^{0.42U}. \quad (12)$$

After the surface film has developed ( $\rho_i/\rho_0 > 1.0078$ ),

$$K_{ma} = 1/5k_{mb}, \quad (13)$$

where  $\rho_0$  is the initial oil density and  $\rho_t$  is the weathered oil density at time  $t$ . The reduction of the evaporation rate was found to a drastic fivefold reduction after the formation of the surface film.

Drivas [9] compared the Mackay and Matsugu equation with data found in the literature and noted that the equations yielded predictions that were close to the experimental data. In 1982 Rheijnhart and Rose [10] developed a simple predictor model for the evaporation of oil at sea. They proposed the following simple relationship:

$$Q_{ei} = \alpha C_0 \quad (14)$$

where  $Q_{ei}$  is the evaporation rate of the component of interest,  $\alpha$  is a constant incorporating wind velocity and other factors taken as  $0.0009 \text{ m s}^{-1}$  and  $C_0$  is the equilibrium concentration of the vapour at the oil surface. Several pan experiments were run to simulate evaporation at sea and the data used to test the equation. No means were given for calculating the essential value  $C_0$ .

In 1986 Tkalin [11] proposed a series of equations to predict evaporation at sea:

$$E_i = \frac{K_a M_i P_{oi} x_i}{RT}, \quad (15)$$

where  $E_i$  is the evaporation rate of component  $i$  (or the whole) ( $\text{kg m}^{-2} \text{ s}^{-1}$ ),  $K_a$  is the mass transfer coefficient ( $\text{m s}^{-1}$ ),  $M_i$  is the molecular weight,  $P_{oi}$  is the vapour pressure of the component,  $R$  is the universal gas constant,  $x_i$  is the amount of component  $i$  at time  $t$  and  $T$  is the temperature in Kelvin. Using empirical data, relationships were developed for some of the factors in the equation:

$$P_{oi} = 10^3 e^A, \quad (16)$$

where

$$A = -(4.4 + \log T_b)[1.803\{T_b/T - 1\} - 0.803 \ln(T_b/T)], \quad (17)$$

where  $T_b$  is the boiling point of the hydrocarbon,

$$K_a = 1.25U \times 10^{-3}, \quad (18)$$

where  $U$  is the wind speed ( $\text{m s}^{-1}$ ). The equations were verified using empirical data from the literature.

The most frequently used work in spill modelling is that of Stiver and Mackay [1]. The work is based on some of the earlier work by Mackay and Matsugu [5] but significant additions were made. Additional information is given in a thesis by Stiver [12]. The formulation is initiated with assumptions about the evaporation of a liquid. If a liquid is spilled the rate of evaporation was given by

$$N = kAP/(RT), \quad (19)$$

where  $N$  is the evaporative molar flux ( $\text{mol s}^{-1}$ ),  $A$  is the area ( $\text{m}^2$ ),  $k$  is the mass transfer coefficient under the prevailing wind ( $\text{m s}^{-1}$ ),  $P$  is the vapour pressure of the bulk liquid,  $R$  is the gas constant ( $8.314 \text{ Pa m}^3 \text{ mol}^{-1} \text{ K}^{-1}$ ) and  $T$  is the environmental temperature (K). This equation was arranged to give

$$dF_v/dt = KAPv/(V_0RT), \quad (20)$$

where  $F_v$  is the volume fraction evaporated,  $t$  is time (s),  $P$  is the vapour pressure,  $v$  is the liquid's molar volume ( $\text{m}^3 \text{ mol}^{-1}$ ), and  $V_0$  is the initial volume of spilled liquid ( $\text{m}^3$ ).

Rearranging,

$$dF_v = [Pv/(RT)](KA dt/V_0), \quad (21)$$

or

$$dF_v = Hd\theta, \quad (22)$$

where  $H$  is Henry's law constant and  $\theta$  is the evaporative exposure.

The right-hand side of the second last equation has been separated into two dimensionless groups. The group  $KAt/V_0$  is termed evaporative exposure and was denoted as  $\theta$ . The evaporative exposure is a function of time, the spill area and volume (or thickness), and the mass transfer coefficient (which is dependent on the wind speed). The evaporative exposure can be viewed as the ratio of exposed vapour volume to the initial liquid volume.

The group  $Pv/(RT)$  or  $H$  is a dimensionless Henry's law constant or ratio of the equilibrium concentration of the substance in the vapour phase [ $P/(RT)$ ] to that in the liquid ( $1/v$ ).  $H$  is a function of temperature. The product  $\theta H$  is thus the ratio of the amount which has evaporated (oil concentration in vapour times vapour volume) to the amount originally present. For a pure liquid,  $H$  is independent of  $F_v$  and Eq. (22)

was integrated directly to give:

$$F_v = H\theta. \quad (23)$$

If  $K$ ,  $A$  and temperature are constant, the evaporation rate is constant and evaporation is complete ( $F_v$  is unity) when  $\theta$  achieves a value of  $1/H$ .

If the liquid is a mixture,  $H$  depends on  $F_v$  and the basic equation can only be integrated if  $H$  is expressed as a function of  $F_v$ ; i.e. the principal variable of vapour pressure is expressed as a function of composition. The evaporation rate slows as evaporation proceeds in such cases.

The equation was rewritten to incorporate constants from evaporation experiments:

$$F_v = (T/K_1) \ln(1 + K_1\theta/T) \exp(K_2 - K_3/T), \quad (24)$$

where  $F_v$  is the fraction evaporated,  $T$  is the ambient temperature in K and  $K_{1,2,3}$  are empirical constants. A value for  $K_1$  was obtained from the slope of the  $F_v$  vs.  $\log \theta$  curve from pan or bubble evaporation experiments. For  $\theta$  greater than  $10^4$ ,  $K_1$  was found to be approximately  $2.3T$  divided by the slope. The expression  $\exp(K_2 - K_3/T)$  was then calculated, and  $K_2$  and  $K_3$  determined individually from evaporation curves at two different temperatures. Variations of all the above equations have been used extensively by many other experimenters and for model application.

In 1989 Hamoda and co-workers [13] performed theoretical and experimental work on evaporation. An equation was developed to express the effects of API<sup>0</sup> (American Petroleum Institute gravity – a unit of density) of the crude oil, temperature and salinity on the mass transfer coefficient  $K$ :

$$K = 1.68 \times 10^{-5} (\text{API}^0)^{1.253} (T)^{1.80} e^{0.1441}, \quad (25)$$

where  $K$  is the mass transfer coefficient ( $\text{cm h}^{-1}$ ),  $T$  is the temperature (K) and  $e$  is the salinity in degrees salinity or parts per thousand. The exponents of the equation were determined by multiple linear regression on experimental data.

Quin and co-workers [14] weathered oils in a controlled environment and correlated the data with equations developed starting with Fick's diffusion law and the Clausius–Clapeyron equation. Crude oil was divided into a series of pseudo-fractions by boiling point. Each fraction was taken to be equivalent to an  $n$ -paraffin in evaporation behaviour. The  $n$ -paraffin distributions of a number of naturally weathered crude oils were determined by capillary gas–liquid chromatography. The actual evaporation determined by this procedure was compared with those generated by a computer simulation of weathering. Good agreement was obtained for oil film thicknesses between 10  $\mu\text{m}$  and 1 mm, weathered for periods of up to 4 weeks.

In 1991 Brown and Nicholson [15] studied the weathering of a heavy oil, bitumen. They compared experimental data using a large-scale weathering tank with two spill model outputs. In the FOOS model, the evaporative exposure concept is used in which the fraction of oil evaporated is given by a variant of the Mackay equation:

$$F = [\ln(P_0) + \ln(CE) + 1/P_0]/C, \quad (26)$$

where  $F$  is the fraction evaporated,  $P_0$  is the vapour pressure and  $C$  is an empirical constant. Further,

$$E = (K_m A v t) / (R T_e V_0), \quad (27)$$

where  $E$  is the evaporative exposure,

$$K_m = 0.0048 U^{0.78} Z^{-0.11} S_c^{0.67}, \quad (28)$$

$K_m$  is the mass transfer coefficient,  $R$  is the gas constant,  $A$  is the slick area,  $v$  is the oil molecular volume,  $t$  is the time,  $V_0$  is the initial slick volume,  $T_e$  is the environmental temperature,  $P_0$  is the initial liquid vapour pressure,  $U$  is the wind speed,  $Z$  is the pool size scale factor and  $Sc$  is the Schmidt number, taken as 2.7.

A comparison of the measured evaporation for a  $5 \text{ m s}^{-1}$  wind at an ambient temperature of  $20^\circ\text{C}$  was done with the equation above. A spill volume of  $100 \text{ m}^3$  was assumed. A value of about  $10^{-5} \text{ m}^3 \text{ mol}^{-1}$  was used for the average molar volume. The model generally described the observed evaporation quite well, particularly during the first few hours. Later, however, the model consistently predicted a higher evaporation than was observed. A simple method of correcting the equation was done by assuming that the vapour phase Schmidt number decreases slightly as the skin on the oil thickens. The evaporative exposure was modified to

$$K_m = (0.0025 - 0.000021t)U^{0.78}, \quad (29)$$

where  $K_m$  is the evaporative exposure (or mass transfer coefficient),  $t$  is time (h) and  $U$  is the wind speed ( $\text{m s}^{-1}$ ). The predicted evaporation compared favourably with the measured values.

The ASA model was also compared [15]. This model assumed that the oil consists of a series of components each with a distinct boiling point, API gravity and molecular weight. A mass transfer rate from the slick was then written for each component as

$$dm/dt = K_m P_i A F_i M_i / RT, \quad (30)$$

where  $dm/dt$  is the mass transfer rate,  $K_m$  is the mass transfer coefficient of Mackay,  $P_i$  is the vapour pressure,  $A$  is the spill area,  $F_i$  is the fraction of the slick remaining,  $M_i$  is the mass,  $R$  is the universal gas constant and  $T$  is the ambient temperature (K).

For this simulation, boiling points, volume per cent and API gravities were input for 13 boiling ranges. The general shape of the model curve agreed well with the measured data but the model predicts a slightly higher overall evaporation rate.

In 1992 Bobra [16] conducted laboratory studies on the evaporation of crude oils. The evaporation curves for several crude oils and petroleum products were measured under several different environmental conditions. These data were compared to the equation developed by Stiver and Mackay [1]. The equation used was

$$F_v = \ln[1 + B(T_G/T)\theta \exp(A - BT_0/T)]\{T/BT_G\}, \quad (31)$$



Table 1

Oil	Density g ml <sup>-1</sup> (15 °C)	Viscosity cP (15 °C)	Agreement to equation	T <sub>0</sub>	T <sub>G</sub>	A	B
Adgo	0.95	60	Poor	551	195	24	21
Alberta	0.84	10	Good	397	539	8	12
Amauligak	0.89	15	Moderate	471	370	12	15
Bent Horn	0.82	25	Poor	406	484	11	14
Diesel	0.83	3	Moderate	517	140	20	18
Endicott	0.92	85	Good	454	1400	-0.8	7
North Slope	0.89	25	Good	431	722	5	10
Panuke	0.78	1	Poor	268	368	7	11

where  $F_v$  is the fraction evaporated,  $T_G$  is the gradient of the modified distillation curve,  $A, B$  are dimensionless constants,  $T_0$  is the initial boiling point of the modified distillation curve and  $\theta$  is the evaporative exposure.

The constants for the above equation and the results from several comparison runs are summarized in Table 1.

This comparison showed that the Stiver and Mackay equation predicts the evaporation of most oils relatively well until the time exceeds about 8 h, after that it overpredicted the evaporation that occurred. The 'overshoot' can be as much as 10% evaporative loss at the 24 h mark. This is especially true for very light oils. The Stiver and Mackay equation was also found to underpredict or overpredict the evaporation of oils in the initial phases. Bobra noted that most oil evaporation follows a logarithmic curve with time.

### 3. Use of evaporation equations in spill models

Evaporation equations are the prime physical change equations used in spill models. This is because evaporation is the most significant change that occurs in an oil's composition. Many recent models (after 1980) use the Stiver and Mackay [1] approach. The equations developed by Mackay and co-workers can be implemented in a variety of ways. Often the difference in models is the manner in which the models are applied.

In 1976 Fallah and Stark [17] proposed a random model to predict the evaporation of oil at sea. The rate of evaporation from a free surface was given by

$$\frac{dV}{dt} = KA^\beta [U(z)]^\alpha (P_s - P_a), \quad (32)$$

where  $A$  is the surface area of liquid,  $U(z)$  is the wind speed at height  $z$  above the liquid surface,  $P_s$  is the saturation vapour pressure at liquid surface temperature,  $P_a$  is the

partial vapour pressure in the air upwind of the liquid surface,  $K$ ,  $\alpha$ , and  $\beta$  are constants and  $V$  and  $t$  are volume and time, respectively. This equation was combined with a probability density function and the Blokker equations described above. After a Mellin transform, the following equation was developed:

$$\Delta V_j = \frac{\pi}{4} K_e D_j^{2-\beta} U_j^\alpha (PM)_{j-1} \Delta t_j, \quad (33)$$

where  $\Delta V_j$  is the change in volume ( $\text{m}^3$ ),  $K_e$  is the evaporation coefficient (for hydrocarbons,  $K_e$  is  $1.2 \times 10^{-8} \text{ min}^{-1}$ ),  $\alpha$  and  $\beta$  are constants ( $\alpha = 0.78$  and  $\beta = 0.11$ ),  $D$  is the slick diameter,  $U$  is the wind speed,  $P$  is the vapour pressure,  $M$  is the molecular weight and  $t$  is the time. For oil slicks, the vapour pressure,  $P$ , was said to decrease sharply as evaporation of volatile components takes place causing a changing oil composition.

Weathering curves were used to give values of the vapour pressure,  $P$ , and the molecular weight as a function of the evaporation fraction,  $M(\gamma)$ . The weathering curves for crude oil and gasoline were approximated by the following exponential functions:

$$PM(\gamma) = 1900 \exp\{-8\gamma - 200\gamma^3\} \quad \text{crude oil (20 }^\circ\text{C)}, \quad (34)$$

$$PM(\gamma) = 4000 \exp\{-1.2\gamma - 2.5\gamma^2\} \quad \text{gasoline (2 }^\circ\text{C)}, \quad (35)$$

where  $PM$  is the change in mass of the oil and  $\gamma$  denotes the application of the gamma probability function. Fallah and Stark applied this probabilistic approach to some literature data to demonstrate the technique.

In 1979 Grose [18] used the Mackay and Matsugu [5] equations with some modification:

$$L = (CU^{0.78}D_0^{-0.11})/(RK)P_i \text{Sk} MW_i, \quad (36)$$

where  $L$  is the mass of oil evaporated with time ( $\text{kg s}^{-1}$ ),  $C$  is the environmental transfer constant,  $U$  is the wind speed at the surface ( $\text{m h}^{-1}$ ),  $D_0$  is the diameter of the oiled area (m),  $R$  is the universal gas constant,  $K$  is the oil temperature (K),  $P_i$  is the vapour pressure of the particular component,  $\text{Sk}$  is the skin factor and  $MW_i$  is the molecular weight equivalent of the particular component.

The factor  $\text{Sk}$  ranges from 0.1 to 8 and accounts for the effect of skinning (the formation of a semi-permeable surface layer). Yang and Wang [8] suggest a value of  $\text{Sk} = 0.2$  after the density of their test oils had increased by 0.78%. A value of 1.0 was used in testing the model. In addition, mass loss rate depends on the vapour pressure,  $P_i$ , and the molecular weight,  $MW_i$ , of each fraction.  $C$  is an environmental transfer constant which is dimensionless, but depends on the units used. The value used (0.00024) for  $C$  includes the constant 0.015 after Mackay and Matsugu [5].

Mackay and co-workers developed an extensive oil spill model incorporating a number of process equations including evaporation [19]. The earlier work of Leinonen and Mackay [20] was used with the modification proposed by Yang and

Wang [8]. The process is essentially that of dividing the oil in a number of different fractions and analysing the loss of each fraction by evaporation. The mass transfer function used is the familiar one proposed by Mackay and Matsugu [5].

In 1981 Aravamudan and co-workers [21, 22] developed an oil spill model incorporating evaporation equations of their own development. The rate of evaporation of the different components in crude oil can be represented by the equation

$$\frac{d}{dt}[c_i(t)V(t)] = -k_0 u_w p_i A(t), \quad (37)$$

where  $c_i$  is the mass concentration of the  $i$ th species (mass in unit volume of the oil),  $V$  is the total volume of oil floating on the water surface,  $k_0$  is an empirical evaporation constant,  $u_w$  is the wind speed,  $p_i$  is partial pressure of the  $i$ th species and  $A$  is the total horizontal surface area of the oil slick. Aravamudan and co-workers show that, using various volumetric relationships,

$$\frac{dV}{dt} = -k_0 u_w A(t) \sum_{i=1}^n \frac{p_i}{\rho_i}, \quad (38)$$

and

$$V \frac{dV}{dt} (\ln C_i) = -k_0 u_w A(t) \left( \frac{p_i}{C_i} - \sum_{i=1}^n \frac{p_i}{\rho_i} \right), \quad (39)$$

where  $\rho_i$  is the density of the  $i$ th component and all others are as in Eq. (37). The partial pressure of each component is related to the saturated vapour pressure  $P_i$  of the  $i$ th component at the temperature  $T$  of the oil by

$$\frac{p_i}{P_i(T)} = \frac{C_i/\mu_i}{\sum_{i=1}^n C_i/\mu_i}, \quad (40)$$

where  $\rho_i$  is the density of the  $i$ th component,  $C_i$  is the concentration of component  $i$ ,  $\mu_i$  is the activity of the component  $i$  and  $P_i$  is the partial pressure of component  $i$ . These equations can be solved to obtain  $V$  and  $C_i$  as functions of time. Solutions were developed by assuming a five-component crude oil that spreads on the water surface according to the correlations for the area.

In 1983 Huang [23] reviewed oil spill models and noted the state-of-the-art up to that time. Huang notes that many of the approaches are similar and can be generalized into the following:

- (1) The oil is assumed to be composed of a number of hydrocarbon groups, the mixture of which has physical-chemical characteristics similar to the parent oil.
- (2) The evaporative loss of a given hydrocarbon component is assumed to follow a logarithmic decay, or first-order kinetics.
- (3) The evaporation rate is assumed to be a function of the following key physical parameters: (a) spill area, (b) wind speed, (c) vapour pressure, (d) slick thickness and (e) temperature.

Huang notes that the main difference among models of the second type seems to be the level of detail and sophistication by which various hydrocarbon components and various physical-chemical parameters affecting evaporation are incorporated in the model.

In 1984 Payne and co-workers [24-26] developed an oil spill model using the pseudo-component approach. Given the boiling point (1 atm) and API gravity of each cut (or pseudo-component), the vapour pressure of the cut as a function of temperature was calculated. First, the molecular weight and critical temperature of the cut were calculated according to the following correlation:

$$y = C_1 + C_2X_1 + C_3X_2 + C_4X_1X_2 + C_5X_1^2 + C_6X_2^2, \quad (41)$$

where  $X_1$  is the boiling point ( $^{\circ}\text{F}$ ) at 1 atm,  $X_2$  is the API gravity and  $C_{1-6}$  are constants whose values are shown in Table 2. Similarly, the critical temperature was calculated from the same equation form using the indicated constant values in Table 2.

Next the equivalent paraffin carbon number was calculated according to

$$N_c = (\text{MW} - 2)/14, \quad (42)$$

where  $N_c$  is the equivalent carbon number and MW is the molecular weight assigned to the particular cut. The critical volume was then calculated according to

$$V_c = (1.88 + 2.44N_c)/0.044, \quad (43)$$

where  $V_c$  is the overall critical volume, and the critical pressure was calculated from

$$P_c = \frac{20.8T_c}{(V_c - 8)} + P'_c, \quad (44)$$

where  $P_c$  is the critical pressure,  $T_c$  is the critical temperature,  $V_c$  is the critical volume,  $P'_c$  is the correction factor for critical pressure.  $P'_c = 10$  to correct the critical pressure correlation from a strictly paraffinic mixture to a naphtha-aromatic-paraffin mixture. Next a parameter  $b$  was calculated according to

$$b = b' - 0.02, \quad (45)$$

where

$$b' = C_1 + C_2N_c + C_3N_c^2 + C_4N_c^3, \quad (46)$$

and the values of the constants  $C_1$  to  $C_4$  are indicated in Table 2.

A final parameter designated as  $A$  is then calculated according to

$$A = \frac{T_{rb}}{T_{rb} - 1} [\log_{10}(P_{rb}) + \exp(-20(T_{rb} - b)^2)], \quad (47)$$

where  $A$  is an intermediary parameter,  $T_{rb}$  is the reduced temperature at the normal boiling point,  $P_{rb}$  is the reduced pressure at the normal boiling point and  $b$  is an

Table 2  
Correlation equation constants for the characterization of narrow boiling petroleum fractions [24]

Property	$C_1$	$C_2$	$C_3$	$C_4$	$C_5$	$C_6$
Molecular weight ( $T_b \leq 500^\circ\text{F}$ )	6.241E + 01	- 4.595E - 02	- 2.836E - 01	3.256E - 03	4.578E - 04	5.279E - 04
Molecular weight ( $T_b \leq 500^\circ\text{F}$ )	4.268E + 02	- 1.007	- 7.491	1.380E - 02	1.047E - 03	2.621E - 02
Critical temperature ( $T_b \leq 500$ )	4.055E + 02	1.337	- 2.662	- 2.169E - 03	- 4.943E - 04	1.454E - 02
Critical temperature ( $T_b > 500$ )	4.122E + 02	1.276	- 2.865	- 2.888E - 03	- 3.707E - 04	2.288E - 02
$b'$	1.237E - 02	2.513E - 01	4.039E - 02	- 4.024E - 02	—	—
Kinematic viscosity (cS at 122 °F, API $\leq$ 35)	- 4.488E - 01	- 9.344E - 04	1.583E - 02	- 5.219E - 05	5.268E - 06	1.536E - 04
Kinematic viscosity (cS at 122 °F, API $\leq$ 35)	6.019E - 01	1.793E - 03	- 3.159E - 03	- 5.1E - 06	9.067E - 07	3.522E - 05

intermediary parameter determined in Eq. (45). The vapour pressure equation which can be used down to 10 mmHg is

$$\log_{10} P_r = \frac{-A(1 - T_r)}{T_r} - \exp[-20(T_r - b)^2], \quad (48)$$

where  $P_r$  is the reduced pressure,  $T_r$  is the reduced temperature,  $A$  is an intermediary parameter determined in Eq. (47) and  $b$  is a constant determined in Eq. (45).

$A$ ,  $b$ ,  $T_c$  and  $P_c$  were determined from the normal boiling point and API gravity of the cut. The temperature at which the vapour pressure is 10 mmHg was obtained by the root-finding algorithm of Newton-Raphson.

Below 10 mmHg, the vapour pressure was calculated according to the Clausius-Clapeyron equation as follows,

$$\ln \frac{P_2}{P_1} = \frac{\lambda_0}{RT_c} \int_{T_{r,1}}^{T_{r,2}} \frac{(1 - T_r)^{0.38}}{T_r^2} dT_r, \quad (49)$$

where  $P_1$  is the vapour pressure at temperature 1,  $P_2$  is the vapour pressure at temperature 2,  $\lambda_0$  is the heat of vaporization at 0 K,  $R$  is the universal gas constant,  $T_c$  is the critical temperature and  $T_r$  is the reduced temperature and was based on the law which states that the ratio of the heat of vaporization,  $\lambda$ , to  $(1 - T)^{0.38}$  is a constant at any temperature. The latent heat of vaporization was calculated from the slope of the natural log of the vapour pressure equation with respect to the temperature where the vapour pressure is 10 mmHg. Thus, in the above equation,  $P_2$  is the 10 mmHg vapour pressure at the temperature,  $T_r$ , previously determined.

Rasmusen [27] developed an oil spill model for Danish waters and proposed an equation to describe the evaporative mass flux of a single hydrocarbon:

$$N_i = k_{mi} \frac{P_i^{\text{SAT}} - P_{i\text{air}}}{RT} X_i^{\text{surface}}, \quad (50)$$

where  $N_i$  is the evaporative mass flux,  $k_{mi}$  is the mass transfer coefficient of component  $i$  ( $\text{m s}^{-1}$ ),  $P_i^{\text{sat}}$  is the vapour pressure of component  $i$  at ambient temperature  $T$ ,  $P_{i\text{air}}$  is the partial pressure of component  $i$  in the air,  $R$  is the universal gas constant and  $X_i^{\text{surface}}$  is the mole fraction of component  $i$  at the surface.

Rasmusen chose an equation by Mackay and Matsugu [5] to estimate the mass transfer coefficient:

$$k_{mi} = 16.076 U^{0.78} R^{-0.11} Sc_i^{-0.67}, \quad (51)$$

where  $k_{mi}$  is the mass transfer coefficient,  $U$  is the wind speed ( $\text{m s}^{-1}$ ) and  $Sc_i$  is the Schmidt number for component  $i$ .

In 1988 Ross and Dickins [28] used empirical data to model the evaporation of oil under snow. The evaporative exposure approach of Stiver and Mackay [1] was used:

$$F_v = (T/10.3T_G) \ln(1 + (10.3T_G/T)) \theta \exp(6.3 - 10.3T_0/T), \quad (52)$$

and

$$\theta = kAt/V = kt/x, \quad (53)$$

where  $F_v$  is the volume fraction evaporated,  $T$  is the environmental temperature (K),  $T_G$  is the slope of the modified ASTM distillation curve (K) (539 K for medium crude),  $T_0$  is the intercept of the modified ASTM distillation curve (K) (385 K for medium crude),  $\theta$  is the evaporative exposure coefficient,  $k$  is the mass transfer coefficient ( $\text{m s}^{-1}$ ),  $A$  is the spill area ( $\text{m}^2$ ),  $V$  is the spill volume ( $\text{m}^3$ ),  $x$  is the slick thickness (m) and  $t$  is the time (s).

The following relationships were defined:

$$A = (T/10.3T_G), \quad (54)$$

$$B = (10.3T_G/T) \exp(6.3 - 10.3T_0/T), \quad (55)$$

$$F_v = A \ln(1 + B\theta). \quad (56)$$

Rearranging,

$$\theta = (\exp(F_v/A) - 1)/B, \quad (57)$$

and after substituting for  $\theta$ ,

$$x(\exp(F_v/A) - 1)/B = kt, \quad (58)$$

A plot of  $x(\exp(F_v/A) - 1)/B$  vs.  $t$  yielded a slope of  $k$ , the overall mass transfer coefficient. The resistance-in-series approach to mass transfer was used:

$$1/k = 1/k_w + H/k_o + L/D_s, \quad (59)$$

where  $k_w$  is the air-side mass transfer coefficient ( $\text{m s}^{-1}$ ) ( $0.002U^{0.78}$  [5]),  $k_o$  is the oil internal mass transfer coefficient ( $\text{m s}^{-1}$ ),  $H$  is Henry's law constant for the oil,  $D_s$  is the diffusivity of oil vapours in snow ( $\text{m}^2 \text{s}^{-1}$ ) and  $L$  is the depth of oil below the snow surface (m).

A plot of  $1/k$  against snow depth ( $L$ ) has a slope of  $1/D_s$  and an intercept of  $1/k_w + H/k_o$ . The least-squares fit to the small-scale data from the trays with uncompacted snow gives a slope of  $5.5 \times 10^4 \text{ s m}^{-2}$  or  $D_s = 1.8 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ .

In 1989 Reed [29] reported on the development of an evaporation equation. He used the familiar Mackay and Matsugu [5] approach to estimate the mass transfer coefficient:

$$K_2 = 0.029W^{0.78}D^{-0.11}Sc^{-0.67}\sqrt{(\text{MW} + 29)/\text{MW}}, \quad (60)$$

where  $K_2$  is the mass transfer coefficient,  $W$  is the wind speed ( $\text{m h}^{-1}$ ),  $D$  is the slick diameter (m),  $Sc$  is the Schmidt number (Reed used 2.7, that of cumene) and  $\text{MW}$  is the molecular weight of the volatile portion of the spill ( $\text{g mol}^{-1}$ ). The mass transfer rate of the surface slick was then stated as

$$dm/dt = (K_2P_{vp}A/RT)f\text{MW}, \quad (61)$$

where  $\overline{dm}/dt$  is the mass transfer rate ( $\text{g h}^{-1}$ ),  $K_2$  is the mass transfer coefficient,  $P_{\text{vp}}$  is vapour pressure (atm),  $A$  is the slick area ( $\text{m}^2$ ),  $R$  is the gas constant ( $8.206 \times 10^{-5} \text{ atm m}^3 \text{ mol}^{-1} \text{ K}^{-1}$ ),  $T$  is the temperature (K),  $f$  is the fraction of the remaining slick that is composed of volatile substances and  $MW$  is the molecular weight of the volatile portion of the spill ( $\text{g mol}^{-1}$ ).

In 1991 Lunel [30] combined the mass transfer rates of evaporation and dissolution to deal with these competing processes simultaneously. The mass transfer rate of the evaporative portion was expressed as

$$dM_{\text{E}}/dt = (k_{\text{E}}MPA)/RT, \quad (62)$$

where  $dM/dt$  is the mass transfer rate,  $k_{\text{E}}$  is the evaporative mass transfer coefficient,  $M$  is the relative molecular mass,  $P$  is the vapour pressure,  $A$  is the slick area,  $R$  is the gas constant  $T$  is the temperature. The evaporative mass transfer coefficient was solved using the work of Mackay and Matsugu [5]:

$$k_{\text{E}} = 0.029U^{0.78}X^{-0.11}Sc_G^{-0.67}, \quad (63)$$

where  $k_{\text{E}}$  is the evaporative mass transfer coefficient,  $U$  is the wind speed at a height of 10 m,  $X$  is the pool diameter and  $Sc_G$  is the gas phase Schmidt number.

Estimates of  $k_{\text{E}}$  and  $k_{\text{s}}$  were derived from work on dissolved gases. For a dissolved gas to pass into the atmosphere across the air-sea interface it has to overcome two resistances (the resistance being the reciprocal of the mass transfer coefficient): one from the water dissolved in ( $1/k_{\text{s}}$ ) and one from the air above the interface ( $1/k_{\text{E}}$ ). The two resistances were combined to yield an overall mass transfer coefficient according to the formula

$$\frac{1}{k_{\text{Overall}}} = \frac{1}{k_{\text{s}}} + \frac{RT}{Hk_{\text{E}}}, \quad (64)$$

where  $k_{\text{Overall}}$  is the overall mass transfer coefficient,  $H$  is Henry's law constant (the vapour pressure divided by the solubility),  $k_{\text{s}}$  is the mass transfer coefficient from water,  $k_{\text{a}}$  is the mass transfer coefficient from air,  $R$  is the universal gas constant and  $T$  is the ambient temperature. Once the overall mass transfer coefficient was calculated, the workers obtained information on both  $k_{\text{E}}$  and  $k_{\text{s}}$ .

Luk and Kuan [31] describe an oil spill model which incorporates an evaporative equation nearly identical to that of Reed above. Spaulding and co-workers [32] similarly use the same equations for the model OILMAP.

Lehr and co-workers [33] developed an oil spill model (ADIOS) using the evaporative algorithm developed by Stiver and Mackay [1], expressed as

$$\frac{df}{d\theta} = \exp \left[ 6.3 - \frac{10.3}{T}(T_0 + T_G f) \right], \quad (65)$$

where  $f$  is the volume fraction of oil evaporated,  $\theta$  is the evaporative exposure,  $T$  is the temperature of the oil and  $T_0$  and  $T_G$  are oil-dependent parameters derived from the



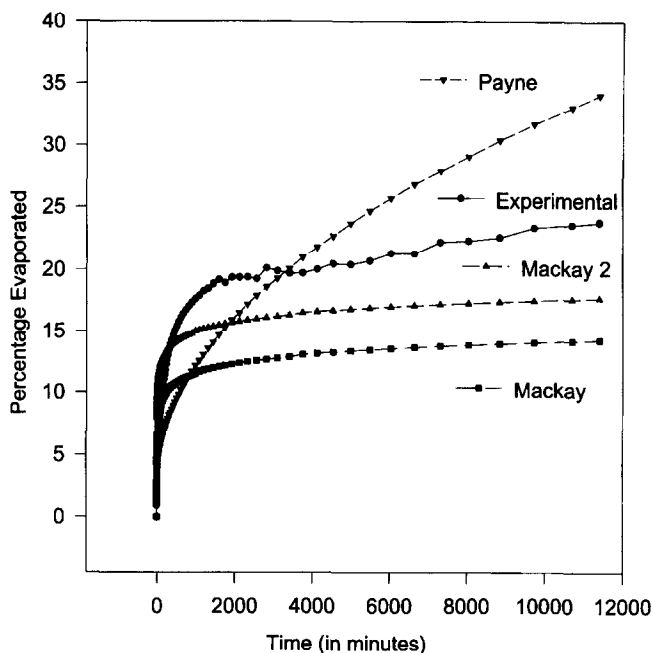


Fig. 1. Comparison of evaporation equations.

fractional distillation data. The evaporative exposure is a dimensionless variable related to time:

$$\theta = \frac{K_m A t}{V_0}, \quad (66)$$

where  $\theta$  is the evaporative exposure,  $K_m$  is the mass transfer coefficient,  $A$  is the area of the spill,  $t$  is the time of exposure and  $V_0$  is the initial spill volume. An adjustment was made to account for the decrease in the evaporation rate as the water content increases to account for oil emulsification. The mass transport coefficient was scaled linearly with the oil fraction in the emulsion.

#### 4. Discussion

In conclusion, many models exist incorporating evaporative equations. The most recent models (after 1980) use one of three approaches to model oil spill evaporation: the Mackay and Matsugu [5], the use of distillation cut data to simulate each fraction, and the Stiver and Mackay [1] approach. The equations developed by Mackay and co-workers can be implemented in a variety of ways. Often the difference in models is

the manner in which the models are applied. The comparison by Bobra [16] found that the Stiver and Mackay equation predicts the evaporation of most oils relatively well until time exceeds about 8 h, after that it over-predicts the evaporation that occurs. This is especially true for very light oils. The comparison by Brown and Nicholson [15] found that for the heavy and mixed oils used, the Mackay and Matsugu approach was better than the distillation cut approach; however the mass transfer coefficient required adjustment. One important point to note is that there exists a large amount of actual evaporation data gathered from laboratories (eg. Refs. [16, 34]). These data are known to be relatively accurate and are data of the type used to develop the earlier models. Another, perhaps more accurate, means to model oil spill evaporation is to use these data directly.

Fig. 1 shows a comparison of the Payne approach [24], the Mackay approach [1, 16] and evaporation experiments conducted in the laboratory [34]. The latter data were gathered by methods presented by Fingas [34] and also similar to that of Bobra [16]. Fig. 1 shows that prediction by both modelling methods can vary from actual evaporation data. Further work is necessary to develop equations that are both simple and accurate.

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