

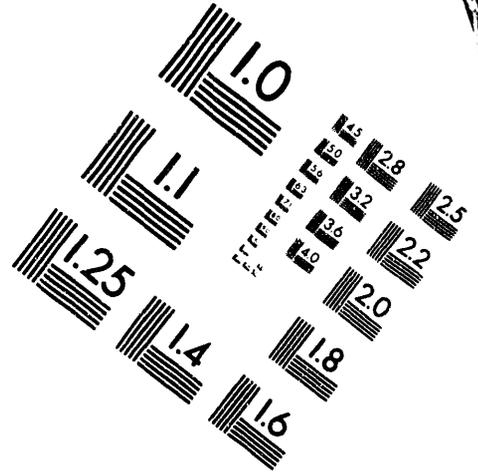
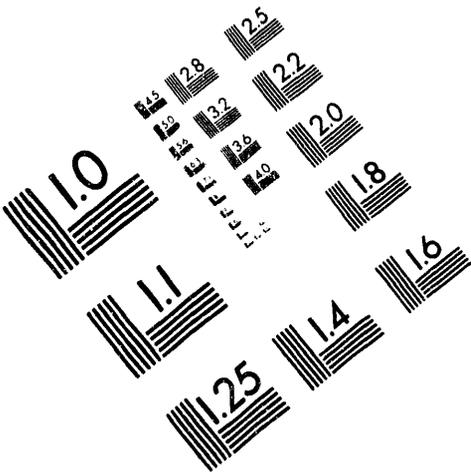


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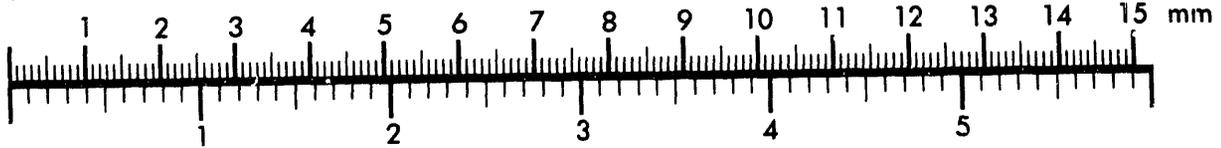
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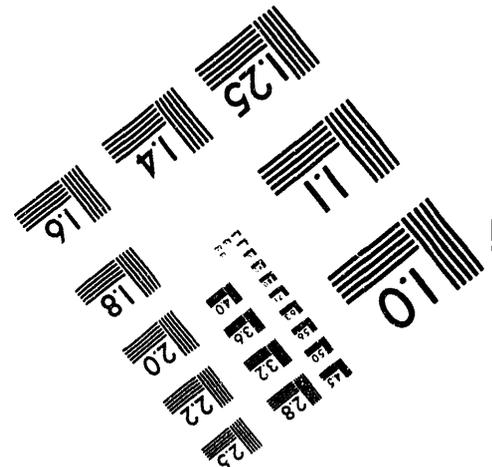
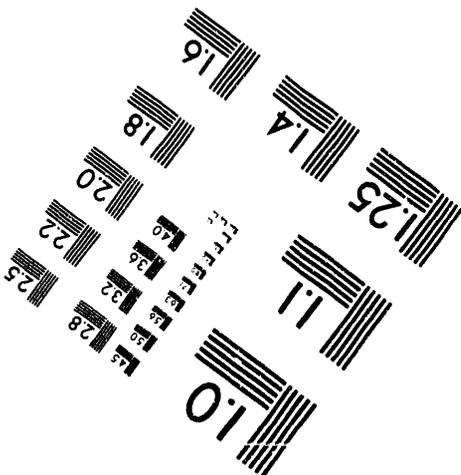
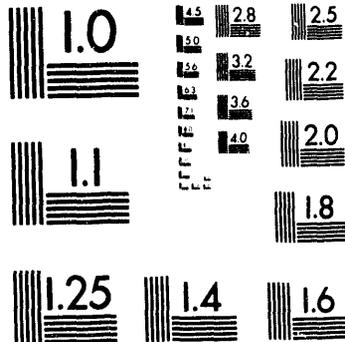
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RISK ASSESSMENT FRAMEWORK OF FATE
AND TRANSPORT MODELS APPLIED TO
HAZARDOUS WASTE SITES

S. T. Hwang

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INTRODUCTION

Risk assessment is an increasingly important part of the decision-making process in the cleanup of hazardous waste sites. Despite guidelines from regulatory agencies and considerable research efforts to reduce uncertainties in risk assessments, there are still many issues unanswered.

An integral part of risk assessment involves estimating pollutant concentrations at exposure points when monitoring data are not available or when predictions are required to assess future migration potentials. Impacts of releases of waste contaminants to the environment and on human health are generally assessed by estimating concentration levels in environmental media using fate and transport models.

This paper presents new research results pertaining to fate and transport models, which will be useful in estimating exposure concentrations and will help reduce uncertainties in risk assessment. These developments include an approach for 1) estimating the degree of emissions and concentration levels of volatile pollutants during the use of contaminated water, 2) absorption of organic chemicals in the soil matrix through the skin, and 3) steady state, near-field, contaminant concentrations in the aquifer within a waste boundary.

These are not new areas of concern and have been the subject of many research publications. The models presented here are intended to enhance estimation accuracy while keeping the approach relatively simple. Some simplifications were made to facilitate model uses and to present them in an analytical form. Because the process in the ground-water media is an unsteady-state process that can last for many years, the model presented here is a steady-state solution that will provide conservative estimations of concentrations in ground water within the waste boundary. This approach should be particularly useful in the regulatory process because a steady-state condition could be attained in a relatively short time period for the near-field concentrations being considered. These concentrations in ground water should represent the maximum contaminant concentrations in ground water averaged over plume depths affected by the contaminant migration.

VOLATILIZATION DURING USE OF CONTAMINATED WATER

The U.S. Environmental Protection Agency recommends Andelman's¹ methodology for estimating the concentrations of toxic organics volatilized from the contaminated water during showering. Andelman's, and Little's⁽¹⁰⁾ methods for estimating air concentrations of toxic volatiles within the shower chamber, in the bathroom subsequent to showering, or within the indoor air affected by appliances using the contaminated water is based on the two-resistant theory of mass transfer. Although a methodology based on the two-resistance theory is a scientifically valid approach, the mass transfer coefficients needed to predict the mass transfer rate of volatile organics released from the shower

water-drops to the air phase were not readily available. In the numerical example, Andelman assumed a range of values for the fractional volatilization of volatiles from the shower water based on empirical results. This paper analyzes the phenomenon of liquid flashing during spraying of water droplets being generated and the liquid phase control of mass transfer common in such a system, and incorporates chemical and physical characteristics important in the volatilization process in the model presented. This analysis avoids the use of mass transfer coefficients but instead allowed the use of parameters readily available for prediction of tendency to volatilize. We will later use an example demonstrating the use of the method and compare the results with experimental results published in the literature.

DERIVATION OF A MODEL FOR AIR CONCENTRATIONS WITHIN A VOLATILIZATION CHAMBER

Suppose that volatilized organics are mixed with air streams entering a volatilization chamber and with the water vapors evaporated from the spraying droplets. The volatilization chamber could be a shower chamber, bathroom, or the enclosure of an appliance using contaminated water. Assuming well-mixed conditions within the containment system under consideration, a mass balance of a volatile component can be presented as:

$$V \frac{dc_i}{dt} = MW G_i - Q_t C_i \quad (1)$$

where V = volume of the volatilization chamber, m^3 ; C_i = indoor air concentration of contaminant i within the chamber, g/m^3 ; MW = molecular weight of component i , g/mol ; G_i = release rate of volatile contaminant i , mol/h ; Q_t = ventilation flow rate leaving the chamber, m^3/h ; and t = time, h . In the above equation and in subsequent nomenclatures, mole refers to gram mole.

An initial condition of $C_i = 0$ when $t = 0$ means that at the moment of turning on the water (i.e., showering), there is no volatile contaminant present within the chamber compartment, solution to Eq. (1) can be written as

$$C_i = \frac{MW G_i}{Q_t} \left(1 - e^{-\frac{Q_t}{V} t} \right) \quad (2)$$

As will be discussed later, when the term $Q_t t/V$ is less than one (1), Eq. (2) can be simplified to

$$C_i = \frac{MW G_i}{V} t \quad (3)$$

The concentration expressed by Eq. (3) represents an instantaneous concentration within the chamber at any time (t) during

the exposure period. When t is the use or showering period, the air concentration estimated by Eq. (3) represents the maximum concentration occurring near the end of this period. The average contaminant concentration within the volatilization chamber during time period t will be

$$\bar{C}_i = \frac{t}{2} \frac{MW G_i}{V} \quad (4)$$

where \bar{C}_i = average contaminant concentration within the shower chamber during the showering period t .

The amount of a contaminant being volatilized from the water droplets within the volatilization chamber can be estimated from a material balance or

$$F_i = d_i + y_i A \quad (5)$$

where F_i = feed rate of volatile contaminant i to shower in mol/h; d_i = flow rate of volatile contaminant i in the water dropped to the bottom of the volatilization chamber and being drained from it in mol/h; y_i = mole fraction of contaminant i in the gas leaving the shower room; and A = total flow rate of gas leaving the volatilization compartment in mol/h. Upon some rearrangement, and assuming an equilibrium relationship between the water phase and the vapor phase within the chamber, Eq. (5) can be modified to

$$d_i = \frac{F_i}{1 + \frac{A}{L} K_i} \quad (6)$$

where K_i = equilibrium constant of contaminant i between the water phase and the vapor phase; and L = total flow rate of liquid (including water and contaminants) being drained out of the volatilization chamber in mol/h. The assumption of an equilibrium between the liquid water phase and the vapor phase should be valid for an aqueous system with a low concentration of volatiles because the liquid phase is a mass transfer controlling phase in the system being considered (or $y_i = K_i d_i/L$).

From Eq. (6), one can obtain the molar flow rate of contaminant i volatilized to the vapor phase by taking the difference between flow rate in the water feed and the flow rate drained out of the volatilization chamber ($G_i = F_i - d_i$) or

$$G_i = F_i \left(\frac{1}{1 + \frac{A}{L} K_i} \right) \quad (7)$$

The total flow rate of gas leaving the volatilization room will consist of air entering the volatilization chamber and being exchanged with the air within the volatilization chamber gas, and can be obtained from

$$A = (\text{ACH})(V)/0.0224 + w + \Sigma G_i \quad (8)$$

where ACH = air exchange rate, h^{-1} ; w = water vapor volatilization rate within the shower chamber; and ΣG_i = total amount of contaminants volatilized in mol/h. The conversion factor $0.0224 \text{ m}^3/\text{mole}$ was applied in Eq. (8) to convert the air flow rate from m^3/h to mol/h.

Equation (4) along with Eqs. (7) and (8) can be used to estimate the concentrations of volatiles released within the air space of the volatilization chamber. In Eq. (8), the term ΣG_i on the right side of the equation is small when compared to the other two terms in the practical situation, as will be demonstrated later. Also the term L needed in Eq. (7) can be approximated by $F - w$ where F is the total shower water flow rate in moles per hr.

EXAMPLE CALCULATIONS FOR AIR CONCENTRATIONS WITHIN A SHOWER CHAMBER

Assume that ground water contaminated with chloroform at a concentration of 50 ppb is used for showering. We wish to estimate an average concentration of chloroform in the air expected to be during a showering period. To proceed with the estimation, it will be assumed that the shower water flow rate is 500 L/h, the shower water temperature is 40°C , the air exchange rate within a shower chamber with a volume of 10,000 L is 0.53 h^{-1} , and the shower duration is 6 minutes. These default values are those recommended by EPA. Note that the molecular weight of chloroform is 119.

The water vapor flow rate flashed from the shower water can be estimated from consideration of a physical process. The concentration of water vapor in the shower chamber is

$$y_w = \frac{w}{\frac{(\text{ACH})(V)}{0.0224} + w + \Sigma G} \quad (9)$$

where y_w is the mole fraction of the water vapor in the shower chamber air. Rearrangement of Eq. (9), noting that the term ΣG is small compared to other two terms in the denominator, yields

$$w = \frac{y_w (\text{ACH})(V)/0.0224}{1 - y_w} \quad (10)$$

Eq. (10) can be further simplified if the term $1 - y_w$ is approximated by unity (1).

At 40°C the vapor pressure of water is 55.3 mmHg. Hence $y_w = 0.073$. Substituting this value into Eq. (10) we get $w = 0.073(0.53 \text{ h}^{-1})(10 \text{ m}^3) / [(0.0224 \text{ m}^3/\text{mol})(1-0.073)] = 18.7 \text{ mol/h}$. From Eq. (8), $A = 255.7 \text{ mol/h}$. The Henry's Law constant for chloroform is $3.4 \text{ atm m}^3/\text{mol}$ at 25°C. Hence $K_i = 180$ at 40°C for chloroform. The amount of chloroform in the feed shower water at the 50 ppb concentration is $F_i = 2.1 \times 10^{-4} \text{ mol/h}$. Also $L = F - w = 27,760 \text{ mol/h}$. Substituting these values into Eq. (7) will give $G_i = 1.3 \times 10^{-4} \text{ mol/h}$. From Eq. (4) the average concentration of chloroform in the shower chamber during the shower period of 0.1 h^{-1} is 0.078 g/m^3 , which is equal to $0.078 \text{ } \mu\text{g/L}$.

Equation (7) can be used to estimate the fraction of chloroform in the shower water volatilized, or the fraction of chloroform volatilized = $G_i/F_i = 0.62$. The magnitudes of the terms in Eqs. (8) and (9) can be tabulated as follows

$$\text{The first term} \quad \frac{(\text{ACH})(V)}{0.0224} = 236.6 \text{ mol/h}$$

$$\text{The second term} \quad w = 18.7 \text{ mol/h}$$

$$\text{The third term} \quad G_i = 1.3 \times 10^{-4} \text{ mol/h.}$$

The above comparison shows that the G_i term is much smaller than the other two terms and hence approximations made for Eqs. (8) and (9) are valid. Even at chloroform concentration of 100 ppm the volatilization rate $G_i = 0.3 \text{ mol/h}$. This value is still much smaller than the other two terms even though this high concentration of chloroform in shower water is unlikely in practical situations. The magnitude of the ΣG_i term will be dependent on the number of volatile contaminants present in the water and their concentrations, but it is still unlikely that their total concentration will reach a range of 100 ppm in water.

It is difficult to compare the air concentrations estimated above with those recommended in EPA guidelines (Andelman¹), because Andelman approached the problem using the mass transfer coefficient for estimating the volatilization rate but in the illustrative problem he assumed the fraction volatilized was in the range of 0.5 to 0.9. Although his approach reflected laboratory experiments, it cannot be translated to a specific chemical and other shower conditions. The fraction estimated is within the experimental range of fractions volatilized but applies only to chloroform at the given conditions. This method is chemical-specific and accounts for variables important in the volatilization process. For other chemicals, the Henry's Law constant should be changed to exhibit different volatility. The water volatilization rate will be affected by temperature.

Figure 1 is a plot of the fraction of a volatile vaporized versus Henry's Law constant at the operating condition used in the example. In preparing this figure, only Henry's Law constant is varied, keeping other conditions the same as those used for

Fig. 1. Fraction Vaporized vs Henry's Law Constant at a Selected Condition

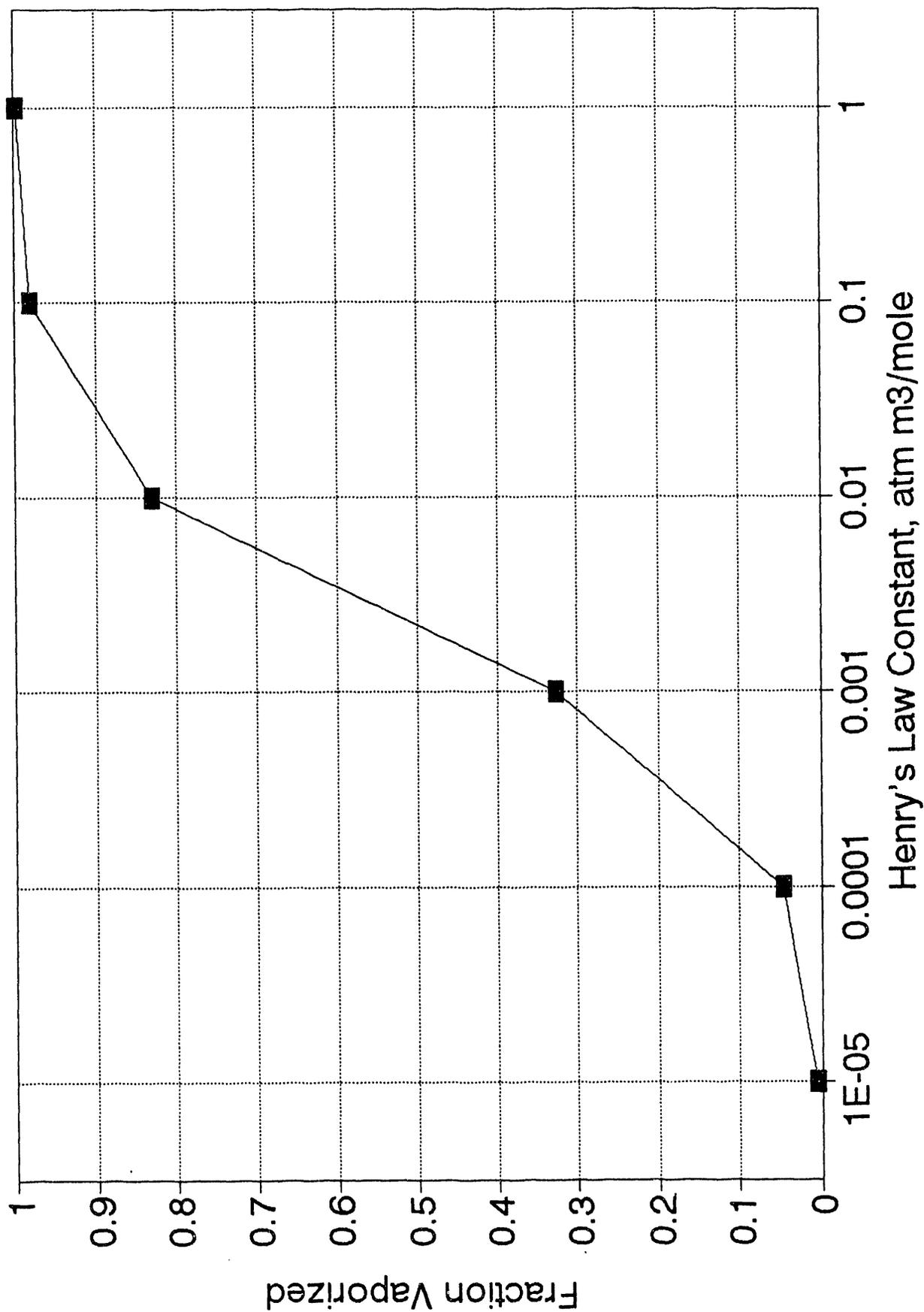
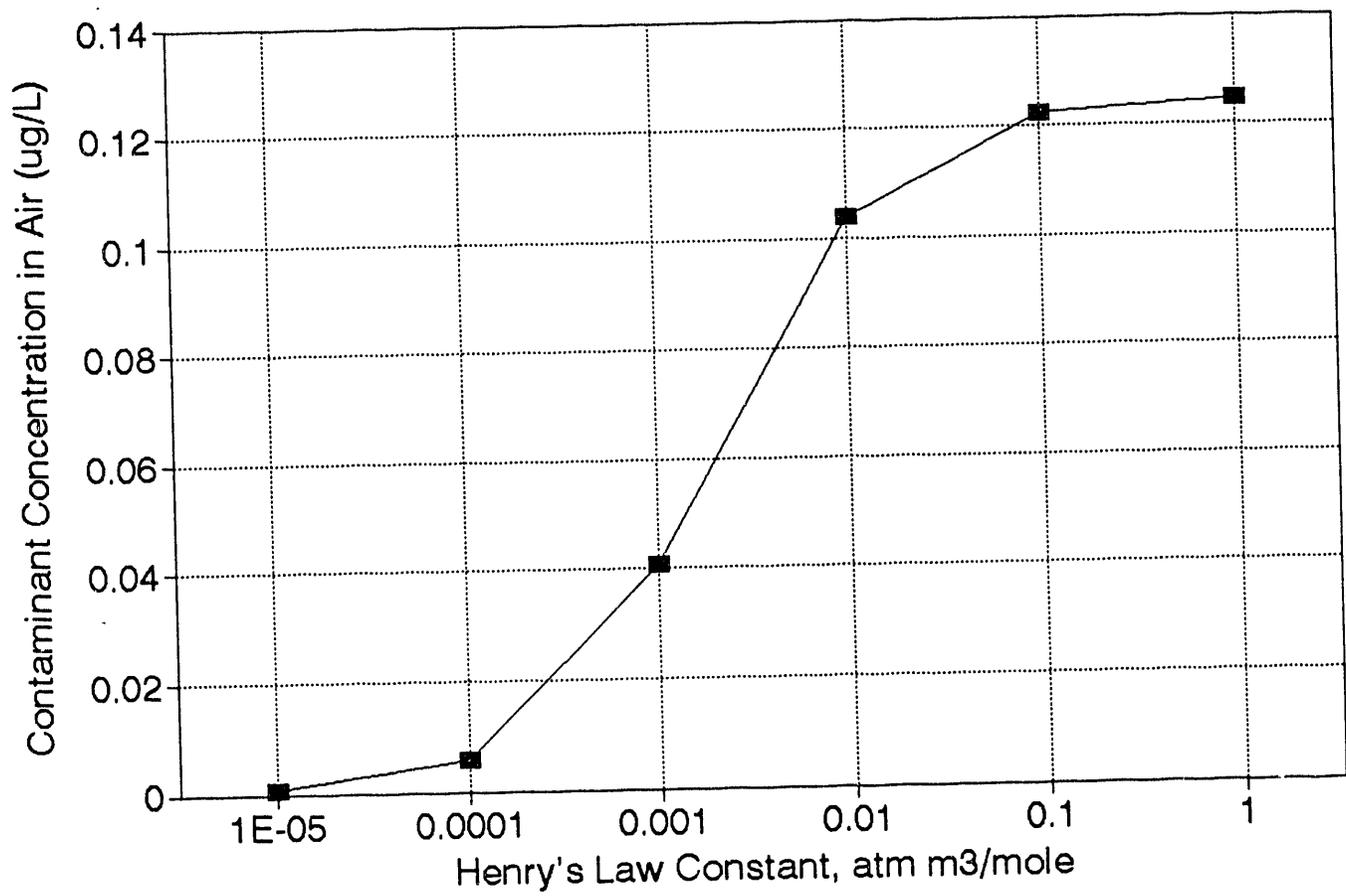


Fig. 2 Air Concentrations vs. Henry's Law Constant at a selected condition



the example. The fraction vaporized is within the experimental range reported in Little.⁽¹⁰⁾ Figure 2 shows a variation of contaminant concentrations in air depending upon the values of Henry's Law constant. This figure was also prepared using the same conditions as the example.

DERMAL ABSORPTION FROM SOIL MATRIX

The U.S. Environmental Protection Agency² issued an interim final guidance for treating dermal absorption of contaminants in aqueous solution. No treatment was presented in this guidance regarding the dermal absorption of contaminants on soil. McKone³ and McKone and Howd⁴ presented models for describing the penetration of contaminants from soil through a series of skin layers based on fugacity and mass transfer concepts. The authors pointed out that there are many assumptions pertaining to the diffusion coefficients through the various layers of the skin and the soil types, and cautioned that a number of questions need to be answered before the models can be used for reliable prediction.

Hwang⁵ presented an alternative model for predicting the fraction of uptake of contaminants in soil through the skin. The method is based on the concept of external and internal bioavailability. The former phenomena refer to the amount of the contaminants removed from the soil matrix to become available for absorption through the skin surface. The latter phenomena refer to the rate of absorption of externally bioavailable contaminants through the skin layer. The overall fraction of contaminants that have penetrated the skin is a combination of both bioavailabilities. Models for these processes, as presented by Hwang⁵ are external bioavailability (B_e), represented by

$$B_e = 1/(1+K_{OC} (OC)) \quad (11)$$

where K_{OC} = organic matter partition coefficient, OC = fraction of organic matter carbon in soil. Internal bioavailability (B_i) in terms of the fraction of the contaminants externally bioavailable that have absorbed through the skin is presented as

$$B_i = 1 - e^{-Qt} \quad (12)$$

where $Q = \rho_b P B_e / S$, ρ_b = bulk density of soil, P = permeability constant, S = amount of soil on the skin, and t = time of contact between soil and skin. An example calculation is shown below to manifest all terms used in the estimation.

EXAMPLE CALCULATION FOR ESTIMATING THE ABSORPTION FRACTION CONTAMINANTS FROM SOIL MATRIX

Assume that we wish to estimate the amount of toluene absorbed through the skin from soil adhered to the hands during normal activities at home. This type of problem occurs in assessing the baseline or remediation risk under various land use scenarios. The following data can be obtained:

$$\begin{aligned} \text{Permeability constant of toluene}^2 &= 0.0009 \text{ cm/h} \\ K_{OC} &= 339 \text{ L/kg} \\ \rho_b &= 1.5 \text{ g/cm}^3 \\ S &= 1 \times 10^{-3} \text{ g/cm}^2 \end{aligned}$$

We get $B_e = 0.13$ from Eq. (11). This represents that the external bioavailability, the fraction of toluene on soil that becomes available for absorption, is about 13%. From these values one can get $Q = 0.18 \text{ h}^{-1}$. If it is assumed that soil stays on the skin for about 4 h before it is washed off, the internal bioavailability can be obtained from Eq. (12) as $B_i = 0.51$. The overall fraction of absorption of toluene in the soil matrix through the skin is $(0.13)(0.51) = 0.07$. This estimation shows that about 7% of toluene in soil is absorbed through the skin in 4 h.

GROUND-WATER FATE AND TRANSPORT

Analyzing the fate and transport of contaminants in the ground-water environment has been extensively researched by hydrologists and engineers. Environmental concerns about ground-water contamination has placed emphasis on the predictive capabilities of the ground-water fate and transport models to forecast potential contamination problems in the future and in wells external to monitoring locations. Despite this emphasis, many ground-water fate and transport models are very complicated to use, and their results leave many unanswered questions.

Codel et al.⁶, Whelan et al.⁹, and Hwang⁷ presented a semi-analytical model to predict potential migration of ground-water contaminants. Although this model is much simpler and requires less data than many numerical models, time integrals requiring a convolution process make its use in the risk assessment process inefficient. The interpretation of its results can be accomplished only by a few experts. But when the time-dependent concentration values are desired, this approach requiring a convolution integral with respect to time is one approach that can be used.

Regulatory agencies are concerned with protecting public health from contaminated ground water at any point, in location and time, impacted by contaminant migration⁸. It is generally assumed that the highest contamination in ground water will be attained at some point within the waste site boundary and under a steady state condition. Regulatory agencies tend to compare these predicted concentration levels to some reference concentrations such as MCLs or risk-based concentration levels.

As leachate enters ground water, contaminant plumes develop in the aquifer. The plumes will penetrate deeper into the aquifer as the ground water travels from the upgradient side of the waste boundary to the downgradient side. The vertical distribution of the plumes should follow a Gaussian distribution with a standard deviation of

$$\sigma_z = (2D_z * t)^{1/2} \quad (13)$$

where σ_{z^*} = standard deviation in the vertical direction,
 D_z^* = retarded effective diffusion coefficient in the
 vertical direction defined as D_z/R_d , where
 D_z = effective diffusion coefficient in the vertical
 direction,
 R_d = retardation factor defined as $R_d = 1 + K_d \rho_b/\epsilon$,
 K_d = soil water partition coefficient
 ρ_b = bulk density of soil
 ϵ = effective porosity of ground water medium
 t = time of travel.

When a steady-state condition is established, the time of travel represents the ground-water travel time below the waste area within the waste boundary. The near-field concentration is defined as the maximum concentration that can occur under the steady-state condition because the pollutant migration and its plume concentration will penetrate vertically into the aquifer as it travels downgradient as described by Eq. (13).

If it is assumed that the pollutant is completely mixed with ground water within its volume of the plume, it is possible to derive an expected concentration level within this near-field volume of contamination. It is expected that the distribution of the near-field contaminant concentrations would be established in a relatively short period of time because the contaminants do not travel beyond the waste boundary. The time required to attain the steady-state concentrations will depend upon the type of chemical, media, and the flow conditions.

The cross-sectional area of a pollutant plume at some point in the near-field aquifer through which the ground water flow occurs is $W \sigma_z$ where W is the width of a waste site under consideration. It is assumed that the depth represented by the standard deviation encompasses most of the contaminant plume. The volume of the contaminated plume within an infinitesimal distance of dx at a downgradient distance x is $W \sigma_z dx$, or

$$\text{The plume volume in } dx = W (2 D_z^* t)^{1/2} dx \quad (14)$$

Eq. (14) can be rearranged after substituting the relationship $t = x/v$ where v is the pore velocity of ground water.

$$\text{The plume volume in } dx = W \left(\frac{2 D_z^* x}{v} \right)^{1/2} dx \quad (15)$$

The total plume volume within the waste boundary can be obtained by integrating Eq. (15) from $x = 0$ to $x = L$ where L is the length of the waste site.

$$\text{The total volume of the plume (within the waste boundary)} = 3 W \left(\frac{D_z^*}{2 v} \right)^{1/2} L^{3/2} \quad (16)$$

Hence the average mixing depth, h_m , for the near field contaminant plume under the steady state condition is the contaminant flow volume divided by the length of ground water travel (L), or

$$h_m = 3 \left(\frac{D_z^*}{2 v} \right)^{1/2} L^{1/2} \quad (17)$$

The average near-field concentration of the contaminant within this mixing depth, C_w , will be

$$C_w = \frac{(C_L) (L) (I)}{(h_m) (v) (\epsilon) + (L) (I)} \quad (18)$$

where C_L = contaminant concentration in leachate, I = precipitation infiltration rate, and ϵ = porosity of ground-water medium.

In Eq. (18) the denominator represents the volumetric flow rate of ground water passing through the average cross-sectional area of the plume represented by the mixing depth being mixed with the water in the infiltrating leachate, and the numerator is the contaminant flux rate in the leachate entering the ground water within the waste boundary. The concentration estimated by Eq. (18) should represent the average near-field contaminant concentration within the plume standard deviation established under the steady-state condition.

An Example Estimating the Steady State Contaminant Concentration within the Near-Field Contaminant Mixing Depth.

Assume that one of the components in a spill site approximately 600 feet long and 300 feet wide is benzene. A sampling shows that the benzene concentration in the contaminated soil is about 250 ppb. The infiltration rate of precipitation is evaluated using EPA's HELP model, based on the precipitation rate of about 0.07 cm/d in the area, is estimated at 0.014 cm/d. Additional site-specific and chemical specific data include

$$v = 12.2 \text{ cm/d}; K_d \text{ for benzene} = 1.5; \epsilon = 0.25; R_d = 10; \alpha_z = 10 \text{ cm}$$

where α_z is the longitudinal dispersivity. The retardation diffusion coefficient can be obtained as $D_z^* = \alpha_z v / R_d$.

The steady-state benzene concentration within the near-field aquifer estimated by Eqs. (17) and (18) is

$$C_w = 8 \text{ ppb}$$

The concentration of benzene within the plume in the aquifer beneath the waste site is estimated to be about 8 ppb when a steady-state condition is reached.

RESEARCH NEEDS

Models presented above are theoretical and have not been tested by experiments. However, the degree of volatilization estimated by the flash vaporization principles is well within the range of stripping provided in various papers, including those of Andelman and McKone. This type of comparison is not sufficient to compare the estimation with experimental results. Additional research will be needed to test the validity of the equilibrium flash principles in the shower situation depending upon the type of compounds and the shower contaminant configuration. Because the liquid phase is generally the controlling phase for volatilization of volatile organics at low concentrations from water, it is believed that the mass transfer rate reaches a steady state condition in the shower room and that the assumption of equilibrium flashing is sound.

The model for predicting the absorption fraction of organics through the skin will require experimental verification. No general procedure is available for testing the absorption process through the skin. Human skin behaves differently from that of animals that are commonly used in skin absorption experiments for contaminants in soil. Diffusion experiments based on human cadaver skin could also provide a different absorption fraction from living skin because it is dryer and less porous. Until an experimental procedure is established, it will be difficult to experimentally test the model predictions.

The prediction of contaminant concentrations in ground water present a challenge. Despite past efforts by numerous researchers, this area still requires expert knowledge in ground-water flow and contaminant transport phenomena.

Many of the transport models used for risk assessment were derived on the basis of describing releases into a single media compartment. As a result, it has been difficult to assess the multimedia partitioning of releases occurring to the various media from a single source of contamination. The future multimedia exposure assessment models, including ground-water fate and transport models, should account for releases to the multimedia environment and the mass balances within the source.

There are other research areas that should be addressed to meet some of the regulatory requirements for risk assessment pertaining to hazardous waste sites. Regulatory agencies should identify these areas and set priorities for research to expedite their regulatory procedures. Enforcing regulations without providing proper tools and guidelines for the potentially responsible parties (PRPs) is a costly process for regulatory agencies and PRPs. There are many areas requiring model development and experimental research. Enumerating these areas is beyond the scope of this paper. Regulatory agencies and PRPs should cooperate to resolve technical and budgetary issues so that public health and the environment can be protected as early as possible but without undue economic burden.

DISCUSSION

In deriving the formula for estimating the amount of absorption of a compound in soil through the skin, no consideration was given regarding the disappearance of the compound as a result of volatilization. This assumption would be particularly pertinent to volatile organic compounds or in situations in which the exposure period is extensive. In these cases, the extent of absorption predicted by Eq. (12) should represent the conservative values.

Equation (18) can be used to estimate the steady state contaminant concentration within the near-field aquifer. This equation is based on the assumption that a continuous leachate flux exists to contaminate the ground water. In many spill situations only finite amounts of contaminants are spilled. As a result, the leachate concentration will tend to decrease as the leaching process progresses and the contaminant inventory in the waste source decreases. Because this transient decrease in the mass inventory was not considered in deriving Eq. (18), the estimated concentration should represent a conservative value. In addition, other multimedia migration processes such as volatilization may need to be incorporated for depleting the original mass inventory of the contaminant.

In the example for estimating the near-field benzene concentration in the ground water plume, it can be noted that the estimated concentration would be dependent upon the site-specific value for the vertical diffusivity coefficient. At present there is no reliable method for estimating this parameter. One method involves estimating the scale-dependent longitudinal dispersivity (such as 0.1 of the travel distance), taking one half or one third of the longitudinal dispersivity for the lateral and vertical dispersivities. The vertical effective diffusion coefficient is obtained by multiplying the vertical dispersivity by groundwater velocity. Uncertainty associated with the vertical dispersivity estimated by this method should be assessed to determine the extent of uncertainty associated with the estimated concentration value in ground water.

CONCLUSION

This paper presented some of the models that can be used in the process of exposure assessment for hazardous waste sites. The models presented pertain to estimating air concentrations of volatiles emitted from contaminated water during use, dermal absorption of organics in soil, and steady-state near-field contaminant concentrations in the ground water plumes. Use of these models should help risk assessors better characterize the levels of contaminant concentration in the environmental media for evaluating pertinent pathways during the process of risk assessment.

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