

Health risk assessment of biodegradable volatile organic chemicals: A case study of PCE, TCE, DCE and VC

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Abstract

A long-term health risk assessment based on a multi-media, multi-pathway approach for biodegradable volatile organic compounds is presented in this paper. In particular, health risk assessment of perchloroethylene (PCE), trichloroethylene (TCE), 1,1-, *cis*-1,2-, *trans*-1,2 dichloroethylene (DCE) and vinyl chloride (VC) in Los Angeles county is considered. The chemicals PCE, TCE, DCE and VC are commonly used and have been known to undergo anaerobic and/or aerobic transformation in the sub-surface environment. Vinyl chloride has been identified as a carcinogenic agent and PCE, TCE, and DCE have been identified as possible carcinogenic agents. In order to simulate the fate of these chemicals in the environment, subsequent human exposure through various pathways, and finally public health risk, multi-component, multi-media transport and multi-pathway exposure models are employed. The health risk assessments are estimated using various dose-response models with and without biodegradation effects.

Introduction

Toxic substances, when released into the biosphere, are distributed among environmental media (the air, water, and soil), and subsequently can enter the human body through various exposure pathways. Because of the complexity of environmental transport/transformation mechanisms, the assessment of public health risks for some chemicals is generally difficult and highly uncertain. For both organic and inorganic chemicals, long-term public health risk depends on the time scale considered (from previous decades, well into the future), the source strength and environmental fate of the chemicals the exposure pathway, and the toxicological properties and dose via each pathway.

The long-term public health risk of a family of volatile organic compounds, in this case, perchloroethylene (PCE), trichloroethylene (TCE), 1,1-, *cis*-1,2-, *trans*-1,2-dichloroethylene (DCE) and vinyl chloride (VC) is

studied in this paper¹. This family is chosen because it is composed of major industrial chemicals which are among the most common volatile organic compounds (VOCs) detected in air and groundwater, and reported world-wide [1-3].

The assessment presented in this paper illustrates a number of important considerations in estimating the risks for a class of toxic chemicals, i.e. VOCs: (1) the importance of biodegradation and its relation to Henry's law constant, (2) the importance of multi-media, multi-component modeling in determining the dominant exposure pathways and hence the risk, and (3) the sensitivity of the exposure and the estimated risks to the important parameters.

This work can provide critical views on the effect of multi-media, multi-pathway exposure on health impact assessment, and can be used as a screening basis for the selection of available alternatives in risk management for VOCs.

The multi-component, multi-media transport model

In general, the following tasks are required in order to assess the health risks of a pollutant in the environment:

- (a) Characterization of the pollutant source term: Determine its chemical and physical state, its rate of release and its magnitude.
- (b) Quantify the dispersion of the pollutant from the source to the various air, water, and soil media, given the meteorologic and hydrogeologic conditions.
- (c) Quantify each human exposure pathway, human uptake, and assess the dose.
- (d) Estimate the adverse health effects based on appropriate dose-response relationships.

In this paper, a multi-component, multi-media transport model called GEO-TOX [4] is employed in predicting the distribution of contaminants among the environmental media. It is based on a compartmental view of the environment, i.e. a series of distinct but interconnected compartments consisting of terrestrial, atmospheric, and aquatic compartments. A compartment can be described by its total mass, total volume, solid-phase mass, liquid-phase mass or gas-phase mass. Various mass flows among compartments and their boundaries can occur. A chemical species in the environment can undergo various transport mechanisms, e.g. diffusion and advection, as well as transformation (chemical reactions and biodegradation).

The mathematical mass-balance equations are given by:

¹Chemical Abstracts Service Registry No. perchloroethylene, 127-18-4; trichloroethylene, 79-01-6; 1,1-dichloroethylene, 75-35-4; *cis*-1,2-dichloroethylene, 156-59-2; *trans*-1,2-dichloroethylene, 156-60-5; vinyl chloride (VC), 75-01-4.

$$\begin{aligned} \frac{d}{dt}N_i^k(t) = & - \sum_{j=1}^m T_{ij}^k N_i^k(t) - T_{io}^k N_i^k(t) - T_{io}^k N_i^k(t) + \sum_{\substack{i=1 \\ j \neq i}}^n (\lambda_i^j N_i^j - \lambda_i^k N_i^k) \\ & + \sum_{j=1}^m T_{ji}^k N_j^k(t) + S_i^k(t) \end{aligned} \quad (1)$$

$N_i^k(t)$ is the time-varying mole of species k in compartment i ; λ_i^k is the first-order rate constant for removal of species k from compartment i by radioactive decay, chemical decomposition, biodegradation, etc.; T_{ij}^k is the rate constant for the transfer of species k from compartment i to compartment j ; T_{io}^k is the rate constant for the transfer of species k from compartment i to a compartment outside of the landscape system; S_i^k is the source term for the introduction of species k into compartment i ; m is the total number of compartments within the landscape system; n is the total number of species within the landscape system.

When only considering the transformation of species k to species $k-1$ within compartment i , the mass-balance equations can be simplified to the following matrix equations such as that used by the GEOTOX code [4]:

$$\dot{\mathbf{n}}(t) = [\mathbf{T}]\mathbf{n}(t) + \mathbf{s}(t) \quad (2)$$

in which \mathbf{n} and \mathbf{s} are vectors of concentration (in moles) and source with length $M = m \times n$, and $[\mathbf{T}]$ is a matrix of size M by M . Note that the matrix $[\mathbf{T}]$ is sparse. In the case study presented, a linear biodecay relationship is used so that the GEOTOX code can calculate the environmental concentrations preparatory to the exposure pathway analysis. There are eight compartments used in GEOTOX to model the environment (air, air particle, upper soil, lower soil, groundwater, surface water, biota, and sediment). The calculated environmental concentrations will be used to determine human exposure through different pathways. To assess long-term health risk, for simplicity, the steady-state concentrations for each of the VOCs are used to estimate long-term average exposures for each of the different pathways.

The VOCs PCE, TCE, DCE and VC are known to undergo anaerobic and/or aerobic transformation [5–8] in the sub-surface environment and produce daughter products through various biodegradation chains. The biodegradation products will depend on soil conditions (acidity and alkalinity) and other factors such as microbial population, temperature, moisture content in the soil, etc. The biodegradation products are usually neglected in long-term health risk assessment, even though they are known as possible carcinogenic agents or may produce other health effects. In this paper, PCE, TCE, DCE (including all the isomers, i.e. 1,1-, *trans*-1,2-, *cis*-1,2 and VC are considered to undergo both aerobic and anaerobic transformation. Aerobic transformation occurs in the upper soil because there is sufficient oxygen available, and anaerobic trans-

formation occurs in the lower soil, groundwater and sediment due to a lack of sufficient oxygen. The possible biodegradation relationships and biodegradation half-lives under aerobic and anaerobic conditions are shown in Fig. 1. The important physical and chemical properties of PCE, TCE, DCE, and VC are listed in Tables 1 and 2. Note that for convenience DCE is considered to be a compound with an average property of its isomers.

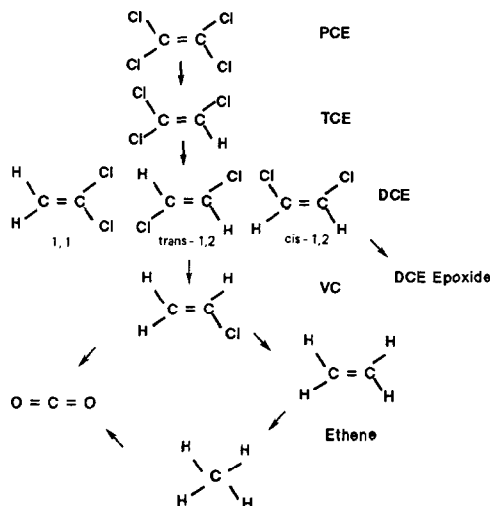


Fig.1. Biodegradation relationship of PCE, TCE, DCE and VC.

TABLE 1

Physical properties of PCE, TCE, DCE and VC

Physical properties	Chemicals			
	PCE	TCE	DCE	VC
Molecular weight (g/mol)	165.83	131.4	96.94	62.5
Henry's law const. (torr/mol l) ^a	17240	8837	99840	17610
Organic carbon part. coef. (k_{oc}) ^a	364	126	58	57
Diffusion coef. in air (m^2/s) ^b	7.0×10^{-6}	7.78×10^{-6}	9.94×10^{-6}	1.07×10^{-5}
Diffusion coef. in water (m^2/s) ^c	8.834×10^{-10}	9.71×10^{-10}	1.09×10^{-9}	1.25×10^{-9}
Bioconcentration factor ^{a,e}	31	10.6	2.9	1.17
Unit risk for inhalation (UR) ^d	5.8×10^{-7}	1.3×10^{-6}	1.37×10^{-4}	2.07×10^{-6}
Cancer potency for ingestion (q^*)	5.1×10^{-2}	1.1×10^{-2}	5.8×10^{-1}	2.3

^aHenry's law constant, k_{oc} and bioconcentration factor are based on reference 13.

^bAir diffusion coefficient is calculated by Hirschelder formula at 20°C [12,35].

^cWater diffusion coefficient is obtained from Wilke and Chang's work [12,36].

^d UR , in $(\mu g/m^3)^{-1}$, q^* , in $(mg/kg \cdot day)^{-1}$.

^eBioconcentration factor is defined as: $bcf = (\text{ppm in fish meat}) / (\text{ppm water})$.

TABLE 2

Half-lives and partition coefficients of PCE, TCE, DCE, and VC at 293K

I. Half-life (days)

Chemical	Air	Upper soil	Lower soil	Surface water	Ground water	Sediment
PCE	47	54	108	30	108	54
TCE	3.7	43	86	90	86	43
DCE	1.8	53	106	6	106	53
VC	1.2	80	160	5	160	80

II. Partition coefficient

	PCE	TCE	DCE	VC
Air/Water	0.096	0.039	0.87	0.98
Upper soil/Water	8.74	3.02	1.46	1.37
Lower soil/Water	0.36	0.13	0.061	0.057
Rock/Ground water	0.105	0.036	0.017	0.016
Sediment/Surface water	9.83	5.79	1.64	1.54
Biota/Upper soil	0.57	1.65	3.42	3.66
Meat fat/Diet	0.0094	0.0051	0.0032	0.0085
Milk fat/Diet	0.0094	0.0051	0.0032	0.0085
Fish/Water	31.0	10.6	3.6	1.17

When VOCs (such as PCE, TCE, DCE, and VC) are introduced into the environment, they tend to diffuse from phase in a direction towards establishing equilibrium between all phases. In determining the steady state concentrations among each environmental compartment, Henry's law constant (air-water partition coefficient) for VOCs is known to be a dictating factor. Frequently, Henry law's constant has been reported for VOCs with very different values either from experimental measurement or calculation by empirical formula. To investigate the importance of the Henry law constant, a sensitivity study has been conducted. The results are presented, along with the other results of this study in the next section.

Distribution of PCE, TCE, DCE and VC among different environmental media are simulated in the area of Los Angeles county by using the estimated emission rates of each chemical. Perchloroethane, TCE, DCE, and VC are treated in a coupled sense. This means that they are coupled through biodegradation relationships in the simulation. In order to determine this biodecay effect on sub-surface concentrations and corresponding impact on final risk assessment, PCE, TCE, DCE, and VC are also treated as separate chemicals, i.e. with no biodecay relation in the sensitivity study.

The atmospheric concentrations of PCE and TCE were estimated by the

TABLE 3

Comparison of air and water concentrations ($\mu\text{g}/\text{m}^3$) of TCE and PCE
Air

Chemical	GEOTOX	AQMD	ARB	TEAM ^a
PCE	170	17	6.8	6.0
TCE	2.0	1.5	1.7	0.45
DCE	2.2	na	na	na
VC	0.0029	na	na	na

Water		GEOTOX ^b	TEAM
PCE	18	55	
TCE	1.1	75	
DCE	0.2	na	
VC	0.0088	na	

^aThe environmental concentrations are average values [10].

^bThe environmental concentrations are based on concentration in the surface water compartment.

South Coast Air Quality Management District (SCAQMD) and the Air Resource Board (ARB) [9]. They are used to compare the results in the air compartment obtained with the GEOTOX simulation in Table 3. The air and water concentration of PCE, TCE, and VC estimated by the “TEAM” study [10] are also used to compare the results predicted by GEOTOX in Table 3.

Data collection and compilation

As noted above, VOCs when introduced into the environment tend to diffuse from phase to phase in a direction towards establishing equilibrium between all phases. In order to predict concentrations among each environmental compartment, the partition coefficient K_d and Henry's law constant H (air–water partition coefficient) are needed. They are obtained from either field measurement, experiment, or estimated by correlation formulae, if field data are not available. If no measurement is available, the K_d is then estimated by the product of the organic carbon–water partition coefficient, K_{oc} , and the organic carbon fraction, f , in the soil, which are among the required input values in GEOTOX. There are four partition coefficients used in GEOTOX in order to estimate the transfer of a pollutant to foods. The first is K_{sp} , the plant–soil partition coefficient, and is defined as the ratio of pollutant concentration in vegetation (dry weight) to concentration in soil. The second is B_k , the feed to dairy biotransfer coefficient. The third is B_t , the feed to meat biotransfer coefficient. Both B_k and B_t are estimated based on the early work by Travis and Arms [11] which uses geometric mean regressions to correlate B_t and B_k with K_{oc} , respectively. The fourth is the bioconcentration factor (BCF) and is cal-

culated from the ratio of the pollutant concentration in fish (at steady state) to its concentration in water [12]. The K_{oc} and BCF are also listed in Table 1 which are based on EPA's (U.S. Environmental Protection Agency) recommended values [13]. The bioconcentration factor is used for the calculation of concentrations due to contaminants transferring to leafy vegetables.

Landscape setting

The landscape chosen for this study is based on the characteristics of Los Angeles county. The Los Angeles region is approximately 1.05×10^4 km², including land and water surface, and has 8.6 million population. The atmospheric height is estimated to be 400 meters which represents the average mixing height over the South Coast Air Basin of California [14]. The depth of the water body is estimated to be 5 meters which is the average depths of natural and manmade water bodies in Los Angeles county [15]. The average temperature is taken as 15°C, the average relative humidity is 65%, and the yearly average wind speed is 2.7 m/s [14]. The dust load of the air particles is 61.5 µg/m². The average deposition velocity of the particles is 334 m/y. The upper and lower soil layers are 0.26 and 2.4 meters thick respectively. The surface soil layer supports an annual average standing dry biomass of 3.1×10^7 kg/km² respectively. Erosion of surface soil is estimated at 3.06×10^5 kg/km²-y. The groundwater inventory is estimated to be 2.1×10^9 kg/km², porosity 0.1, and average rock density 2.33 kg/L in groundwater zone. The recharge rate to the aquifer is 5 cm/y and irrigation withdrawals from ground water are estimated to be equal to 0.12 cm/y. The area of the surface water is about 1.9% of the total land surface and the underlying sediments are about 0.05 m. Surface runoff amounts to 35 cm/y, evaporation from surface water is 1.1 cm/y, and the "evapotranspiration" rate from soil is 57.9 cm/y. The average precipitation onto land and surface water are estimated to be 36 cm/y [15].

The source terms

The source terms for PCE, TCE, and VC were estimated from emission rates of point sources and area sources into the Los Angeles atmosphere by the SCAQMD [16]. Since there is no information on DCE, it is estimated by using the total emission of solvents in California, and multiplying the ratio of the average annual consumption for 1986, 1984, and 1973 in Los Angeles to those in California [17–19].

Health risk assessment

When potentially toxic substances are dispersed throughout the environment, the population in the area can then become exposed. Several modes of exposure are possible, such as inhalation of emitted gases and particulates,

ingestion of contaminated drinking water, and uptake through dermal exposure to agent-containing rain, lake, or ocean water. To determine whether these exposures constitute a significant hazard to the population, the dose for each toxicant over time and for each exposure pathway needs to be evaluated for each geographically and biologically distinct portion of the population (for simplicity, two demographic groups were used in this study). This is then used to estimate the increased risk over background exposure of a particular toxic substance. In order to estimate risk, statistical or statistical-biological models (dose-response models) are needed. In the first approach, long-term health risks are determined by using the simple point-estimator dose-response model known as the unit risk/potency factor. In the sensitivity study, other point-estimators such as the one-hit, the multi-stage, the multi-hit, and the log-probit are used in order to provide information on possible uncertainty involved when different models are chosen.

Exposure pathways analysis

Depending on the mode and amount of exposure, there are several different possible human health effects. Chronic exposures leading to long-term health risks are considered here. Exposure levels are determined from the various pathways based on daily intake per unit body weight, averaged over the population. The average exposure for the i th pathway in time interval T is expressed as:

$$E_i = 1/T \int_{T_0}^{T_0+T} I_i(t) dt \quad (3)$$

where I_i is the intake by pathway i . The intake rate I_i is defined by the behaviour of the receptor BR_i and the concentration of the i th VOC i.e. C_i . Clearly, the value of BR_i is characteristic of human activity. For example, the inhalation exposure I_h considered in GEOTOX is due to the air I_a and particle I_p inhalation pathways, and I_a can be expressed as:

$$I_a = BR_a C_a = \left[\frac{15}{70} f_{\text{child}} + \frac{55}{70} f_{\text{adult}} \right] C_a \quad (4)$$

where $f_{\text{child}} = 16I_h_a + 8I_h_r$, $f_{\text{adult}} = 16I_h_a + 8I_h_r$, the number of hours per day that an adult or child is active is taken as 16 h, the number of hours per day that an adult or child is resting is taken as 8 h, I_h_a is the breathing rate per unit body weight for adult or child while active, I_h_r is the breathing rate per unit body weight for adult or child while resting.

For simplicity, the various population activity in different micro-environment (indoor, outdoor, in-transit) which would affect exposure level will not be considered. Considering the long-term effect, which allows simplifying assumptions to be made in estimating exposure for PCE, TCE, DCE, and VC. They are summarized in the following:

- (1) Uniform distribution for the consumption of drinking water, aquatic organisms, and animal and vegetable products among two age groups, i.e. children (new-born to fifteen years old) and adults (fifteen to seventy years old).
- (2) Local consumption of the products.
- (3) The synergistic or antagonistic effects due to multiple exposures of more than one chemical is not considered.

In general, because a large number of model parameters are required but are often unavailable, a best-estimate method has been employed. The exposure pathways considered are inhalation, drinking water ingestion, biota ingestion, meat and dairy product ingestion, fish ingestion, soil ingestion, and dermal absorption. These pathways are linked to environmental concentrations in ambient air, air particulates, surface water, groundwater, and soil through pathway exposure factor models [20]. In order to address the volatile organic compounds, the models developed by McKone were used as basis for calculating the pathway exposure factor characterizing the exposure of inhalation attributable to volatilization of the VOC contaminated potable water [21].

More detail regarding population activity pattern studies, influences of time spent in micro-environments to person's exposure, and exposure modeling can be found in Wallace et al. [10] and Ott [22]. The most recent study on criteria pollutant (e.g. O₃) exposure analysis considering nine demographic groups and their corresponding exercise states for various activity patterns in different micro-environment has also been reported [23,24].

Cancer risk

The dose rate for pathway j , D_j , can be constant over the exposure period or be time dependent. It is related to exposure by an absorption factor f_j , i.e. $D_j = f_j E_j$. The absorption factor can be obtained from experimental data or from pharmacokinetic models. More detailed evaluation of f_j is beyond the scope of this paper.

As a first step, the oncogenic potency factor (q^*) and unit risk factor (UR) for PCE, TCE, DCE, and VC are used. Their values are listed in Table 1. The factor q^* or UR is defined as the 95% upper bound confidence limit that tumor induction is likely to occur at a given dose through the ingestion or inhalation pathway, and has units of tumors/(mg of VOC \times kg of body weight \times day) [13]. Assuming an average body weight of 60 kg (allowing for women in the population and the average inhalation rate being 18 m³/day) the conversion factor for $UR = (3.0 \times 10^{-4}) \times q^*$ and $q^* = (3.3 \times 10^3) \times UR$.

The total risk model for a VOC is described as follows:

$$R_{\text{total}} = R_j = \{1.0 - \exp[-D_j q^*(i)]\} \quad \text{for } j=1, p \quad (5)$$

where R_j is the risk of exposure to VOC i for pathway j ; p is the total number of pathways.

$$R_{\text{total}} = D_j q^*(i) \text{ for } j=1, p \quad (6)$$

where the products $D_j q^*(i)$ are small ($< 10^{-3}$). The actual cancer risk can be less than that predicted by the calculational method because the use of q^* or UR should lead to a conservative upper bound.

Three major assumptions were made in estimating the long-term health risks for the family of chemicals treated in this paper:

- (1) Two modes of exposure are dominant: inhalation and ingestion [25].
- (2) Steady-state concentrations are used.
- (3) The dose is constant over the exposure period.

The risk results presented in Table 4 are based on using the unit risk factor for the inhalation pathway and the potency factor for ingestion and other pathways [1,2,13,26–30]. To be conservative, the potency factor can also be used for other pathways such as dermal exposure. However, dermal exposure is not a major risk contributor for VOC's [25].

Sensitivity analysis

Sensitivity analyses were used to identify the relative effect of important parameters on the estimate of human exposure, dose–response, and health risk. They are described in the following paragraphs.

We have studied the sensitivity of exposure rate to different values of Henry's law constant. Using the same source term in the upper soil, groundwater, and air compartment for TCE, we varied Henry's law constant (ranging from 10^{-3} to 10^3). The results of this sensitivity study are plotted in Fig. 2. When the source is placed in the ground water compartment, the exposure rate is almost constant. However, it has one order of magnitude difference with and without consideration of biodecay for a given Henry's law constant. When the source term is placed in the air or upper soil compartment, the exposure rate

TABLE 4

Results of the health risk assessment

Chemicals	Cancer risk			
	Inhalation exposure (w/Bio)	(/wo Bio)	Ingestion Exposure ^a (w/Bio)	(/wo Bio)
PCE	1.9×10^{-5}	1.9×10^{-5}	5.3×10^{-7}	5.3×10^{-7}
TCE	1.4×10^{-6}	1.4×10^{-7}	5.0×10^{-9}	5.0×10^{-9}
DCE	7.8×10^{-5}	7.8×10^{-5}	6.6×10^{-8}	4.3×10^{-8}
VC	1.8×10^{-8}	1.7×10^{-8}	7.4×10^{-9}	2.1×10^{-10}

^aIngestion = Drinking water + Biota ingestion + Meat/dairy ingestion + Fish ingestion + Soil ingestion.

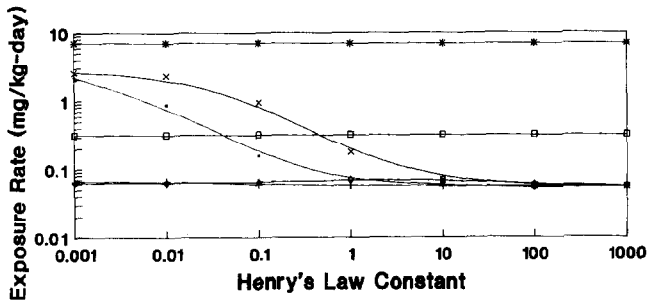


Fig. 2. Effect of Henry's law constant on the exposure rate (bilogarithmic) to TCE. Source/biodecay: (●) air/w bio, (□) groundwater/w bio, (+) air/w bio, (×) upper soil/wo bio, (*) groundwater/wo bio, and (◇) upper soil/w bio. Solid lines are best fits from the GEOTOX model.

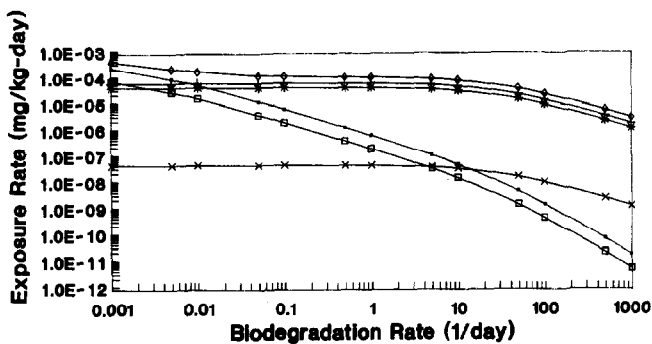


Fig. 3. Biohalf-life sensitivity study of PCE ingestion exposure. Bilogarithmic plot of exposure rate vs. biodegradation rate. (●) Drinking water, (□) fish ingestion, (+) biota ingestion, (×) soil ingestion, (*) meat or dairy ingestion, and (◇) cumulative ingestion. Solid lines are best fits from the GEOTOX model.

decreases with higher Henry's law constant but is essentially the same when Henry's law constant is greater than one. The exposure rate has two orders of magnitude difference (with a decreasing trend when Henry's law constant increases) with and without consideration of biodecay.

The variations of biodegradation half-lives (from days to months depending on soil chemistry) can possibly lead to order of magnitude differences in the calculation of health risk. We thus investigated the sensitivity of the environmental concentrations, and hence the exposure rates for different pathways, to the biodegradation half-life of PCE. The source term is placed in the air, upper soil, and groundwater, and the rate constant varied between 10^{-3} to 10^3 day $^{-1}$ (or equivalently, the biodegradation half-life varied ranging from 6.9×10^{-4} to 6.9×10^2 day). The results are plotted in Fig. 3. The environmental concentrations with and without biodecay in the upper and lower soil, groundwater, and sediment are found to be influenced the most with the vari-

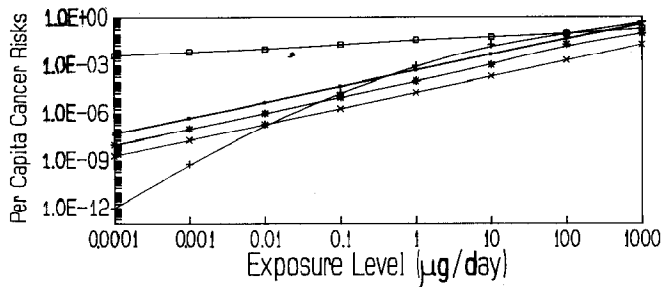


Fig. 4. Dose-response uncertainty inhalation of vinyl chloride monomer. Dose-response models: (■) one-hit, (□) multi-hit, (+) log probit, (×) unit risk, and (*) multi stage.

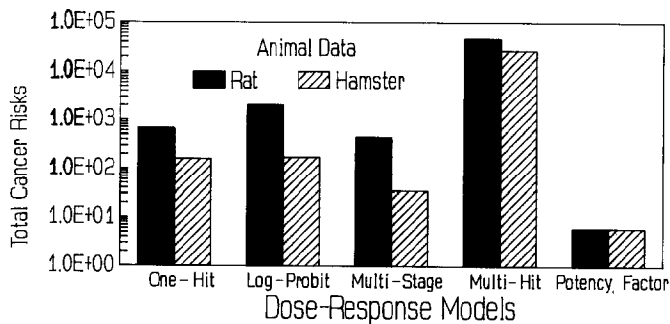


Fig. 5. Comparison of total risks. Hamster vs. rat data.

ation of biodegradation half-life. As can be seen in Fig. 3, the exposure rate due to drinking water and fish ingestion differs by seven orders of magnitude when the source is placed in the upper soil compartment, and the total ingestion exposure rate varies by two orders of magnitude with variation of the biodegradation half-life.

As noted above, there are many dose-response models available, mainly point estimators in a sense, including the one-hit, multi-hit, multi-stage, probit, and Weibull models. For the case of carcinogenic compounds, individual cells are susceptible to genetic level transformation through a sequence of stochastic events induced by those compounds. Most of the dose-response data available have been obtained from different test animals (such as hamsters and rats) exposed to a carcinogen over their lifetime. The effects of using different dose-response models, and different animal data are shown in Figs. 4 and 5. In order to estimate the cancer risk to humans, animal to human scaling is considered. The techniques used for the calculation of equivalent human dose from animal dose data has been discussed in the literature [31].

For the purpose of uncertainty and sensitivity analyses, various dose-response models can be used to assess the health risk due to inhalation of VC predicted at different exposure levels. The prediction of excess health risk for

the different models should approach one another at higher exposure levels. The unit risk and potency factor approach is considered as conservative in the estimation of low level exposure risk.

Discussion

When PCE, TCE, DCE, and VC undergo biodegradation they produce daughter products. These daughter products have been known as carcinogenic or possible carcinogenic agents [1,2,13]. In this paper a simplified biodegradation chain is assumed (i.e. considering PCE→TCE→DCE→VC only) and its behavior in the environment is simulated by the GEOTOX code. Also, the health risk assessment is based on a series of simplified assumptions.

The environmental concentrations of PCE, TCE, DCE, and VC predicted by GEOTOX, with consideration of the bio-degradation effect, are shown in Table 3. Analysis, as indicated by Yeh and Kastenbergl [25], inhalation is the most important exposure pathway and ingestion is the next most important pathway. Hence the total exposure with and without biodegradation due to inhalation and ingestion is not very different. Table 3 also compares the PCE and TCE atmosphere concentrations to those obtained by the SCAQMD and ARB studies [9]. The air concentrations measured are comparable to concentration projected by GEOTOX. Also, the risk estimated by the SCAQMD and ARB are very close to the risks estimated by our approach. However, if one considers the contribution from the other pathways, our approach will be more complete and useful as a screening tool.

There is a great deal of uncertainty involved in the biodegradation half-lives due to the complexity of the soil environment [32]. The biodegradation half-lives used in this paper are based on Vogel and McCarty [5] and Wood et al.'s work [33]. In the sensitivity analysis, the variation of biodegradation half-life has great influence when the source is in the soil or groundwater compartments. Because of a common characteristic of VOCs (a strong tendency to move towards the air compartment), inhalation pathways are dominant. Therefore, the estimated exposure rate and therefore the risk is not significantly different when calculated by any one of the dose-response models.

In reality it is possible for PCE, TCE, and DCE to metabolize in biota and transform to other toxic products [34]. This may lead to a higher removal rate in biota and a compounding effect on exposure and health impact. Such a metabolic enhancement of contaminant removal can be modeled if the metabolic rate constants are estimated correctly. Even though scavenging may also have a significant effect on the concentration in the upper soil and surface water compartments in the event of rain, especially when a significant emission of PCE, TCE, DCE, or VC occurs, it is not so important in Los Angeles county because of little rainfall. Further investigation of these phenomena will be needed to quantify their effect on risk.

The major difficulty encountered in this study is to find true multi-media field data in order to benchmark the GEOTOX modeling results. In this paper only atmospheric data based on inhalation can be found, and used for comparison (keeping in mind the assumptions, of course).

Concluding remarks

Most current studies of the transport and sorption of toxic contaminants have focused on chemical, physical, and biological activity in the saturated zone of groundwater systems. However, volatile pollutants that can readily move between the aqueous and vapor phases in saturated and unsaturated zones has made the prediction of their fate and transport very difficult. In the soil environment, some categories of groundwater pollutant are recognized as biotransformable and/or biodegradable. Risk assessment based on a multi-media, multi-pathway approach for such a family of chemicals has been studied and presented in this paper. The atmospheric concentration, as calculated by the GEOTOX code is in good agreement with the SCAQMD, ARB and TEAM field measurements as shown in Table 3. As for other media, confidence with respect to prediction is still unknown due to a lack of monitoring data. Clearly, further refinement of each compartmental model will require more studies of inter-media mass transport processes and multi-media field data.

Inhalation of contaminated air, and ingestion of contaminated drinking water and food, are the principal exposure routes which partially coincides with a previous study by EPA [1,2]. Due to simplifications made in the calculations (simple biodegradation relationship and screening dose-response models), the estimated risks are not significantly different. In general, the biodecay phenomenon was found to have a secondary effect on exposure rate and health risk for medium and high Henry's law constant chemicals. To properly assess the long-term health risk due to a biodegradable, low Henry's law constant chemical (or chemical family), one should include this commonly ignored effect.

Throughout the process of estimating risk, various sources of uncertainty have been encountered such as selection of biodegradation half-lives, selection of dose-response model, size of the exposed population, Henry's law constant, etc. It is the purpose of this paper to investigate these sources of uncertainty and provide useful suggestions for health risk assessment. Also, the need for multi-component, multi-media models in conducting overall risk assessment has been demonstrated for members of a close family of chemicals. The methods developed in this paper can be applied to other chemical families for assessing health risk.

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