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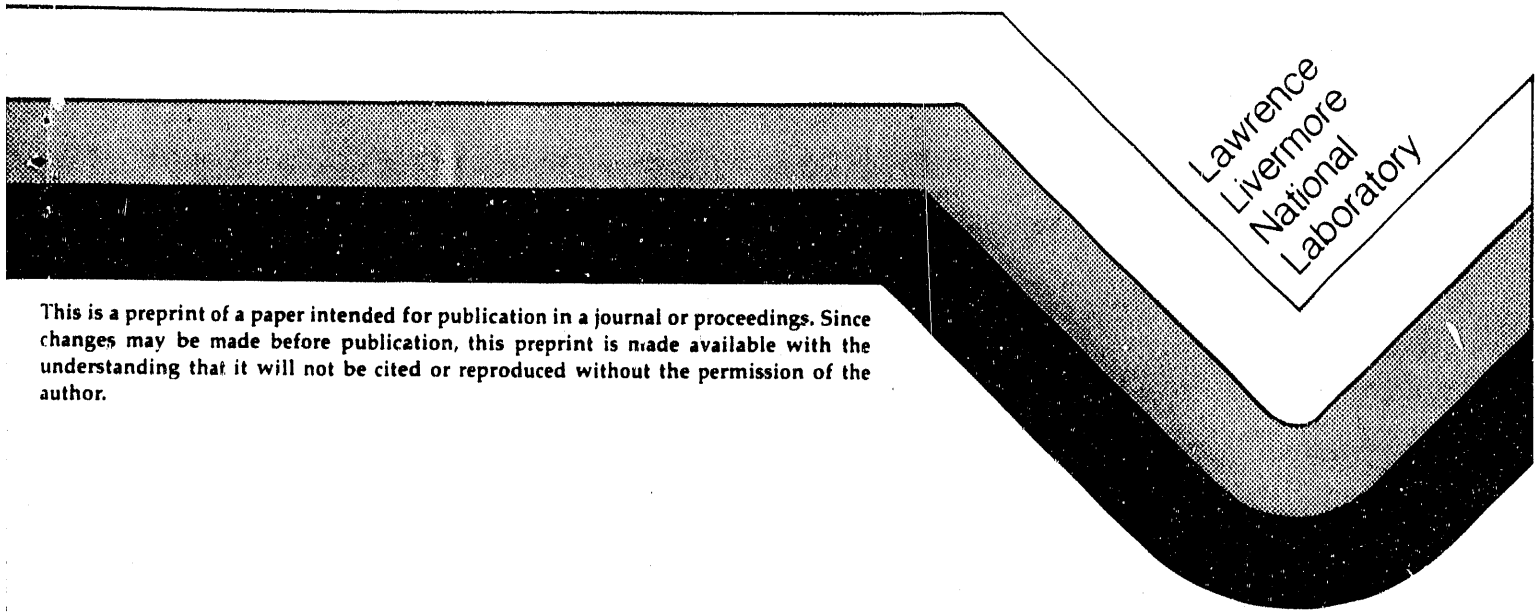

A RISK-BASED CLEANUP CRITERION FOR PCE IN SOIL*

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ABSTRACT

The most important attribute of a chemical contaminant at a hazardous-waste site for decision makers to consider with regard to its cleanup is the potential risk associated with human exposure. For this reason we have developed a strategy for establishing a risk-based cleanup criterion for chemicals in soil. We describe this strategy by presenting a cleanup criterion for tetrachloroethylene (PCE) in soil associated with a representative California landscape. We begin by discussing the environmental fate and transport model, developed at the Lawrence Livermore National Laboratory (LLNL), that we used to predict the equilibrium concentration of PCE in five environmental media from a steady-state source in soil. Next, we explain the concept and application of pathway-exposure factors (PEFs), the hazard index, and cancer-potency factors (CPFs) for translating the predicted concentrations of PCE into estimated potential hazard or risk for hypothetically exposed individuals. Finally, the relationship between concentration and an allowable level of risk is defined and the societal and financial implications are discussed.

INTRODUCTION

Currently, there are no Federal or State of California regulatory limits for concentrations of chemical contaminants in soil analogous to the maximum contaminant levels, MCLs, designed to protect against adverse health effects from the ingestion of drinking water. Therefore, government agencies and responsible parties accountable for the mitigation of hazardous-waste sites in California cannot compare the concentration of a chemical detected in soil at a praticular site to a regulatory limit for that chemical in soil to rapidly ascertain whether the measured soil concentration might pose an unacceptable level of risk to human health. To overcome this predicament confronting risk managers, we have developed a strategy consisting of a six-step procedure for establishing a risk-based cleanup criterion for chemicals introduced into soil. Because this strategy accounts for the multimedia and multiple-pathway exposure characteristics of many soil contaminants, the resulting cleanup criterion for a soil contaminant is consistent with a complete assessment of potential risk to exposed populations.

We illustrate the six-step procedure for developing risk-based cleanup criterion for chemicals in soil with the example of tetrachloroethylene (perchloroethylene, PCE) in the soil of a 100-km^2 landscape representing a typical region of California. This organic chemical is a common contaminant of soil and groundwater in many urban areas of California because of its widespread use as an industrial solvent. Additionally, the U.S. Environmental Protection Agency (U.S. EPA, 1985) classified PCE as a Possible Human Carcinogen (Group C), and the State of California includes PCE in a list of "Chemicals Known to the State to Cause Cancer or Reproductive Toxicity" (California Department of Health Services [CDHS], 1990).

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MULTIMEDIA PARTITIONING OF PCE INTRODUCED INTO SOIL

Once introduced into soil, chemicals such as PCE will distribute into other environmental media including air, surface water, and groundwater. To estimate the concentrations of PCE in air, water, and soil resulting from the steady-state input of PCE to the soil layer of a 100-km^2 landscape representing a typical region in California, we use the multimedia transport and transformation model GEOTOX (McKone, 1981; McKone et al., 1983; McKone and Kastenberg, 1986; McKone and Layton, 1986; McKone et al., 1987; and Layton et al., 1986), which is a computer program developed at the Lawrence Livermore National Laboratory (LLNL). Table 1 summarizes the processes by which contaminants are exchanged and lost among compartments of the GEOTOX model.

The specific landscape parameters (e.g., meteorology, hydrogeology, and soil properties) used in GEOTOX are based on the national land-classification system for describing ecoregions of the United States developed for the U.S. Department of the Interior and the U.S. Department of Agriculture (Bailey, 1980). We estimated or obtained from the literature the physicochemical properties of PCE (e.g., solid/liquid and air/liquid partition coefficients and the diffusion coefficients of PCE in air and water) that also are essential input to the GEOTOX model. Table 2 shows the equilibrium concentrations of PCE in different environmental media predicted by GEOTOX for a 100-km^2 California landscape assuming a soil-based steady-state source of contamination. The predicted concentrations appearing in Table 2 are scaled to a soil concentration of PCE of 1 ppm (mg/kg).

DEVELOPMENT OF PATHWAY-EXPOSURE FACTORS

Pathway-exposure factors (PEFs) incorporate information on human physiology, human-behavior patterns, and environmental transport to link environmental concentrations of a chemical to potential exposure pathways and lifetime accumulations. Accordingly, we use PEFs developed by McKone and Daniels (1990) to translate environmental concentrations of PCE into quantitative estimates of the amount of PCE that passes into the lungs and gastrointestinal tract, and across the surface of the skin and we express such intakes in units of mg/kg-d. For example, Equation 1 is the PEF for ingestion of drinking water (F_{ww}).

$$F_{ww} = \left(\frac{I_w}{BW}\right) , \qquad (1)$$

where

I_w = the lifetime-equivalent fluid intake (2 L/d); and
BW = the lifetime-equivalent body weight (58 kg).

Table 3 contains the numerical values and units of the PEFs for the nine exposure pathways and five environmental media applicable to PCE.

EXPOSURE MATRIX

We construct a matrix containing medium- and pathway-specific human exposures by multiplying the PEFs in Table 3 by the corresponding medium-specific concentrations of PCE shown in Table 2. Then, we total multimedia exposure for a particular pathway by summing the exposure values for that pathway across all media. Table 4 shows the matrix of medium- and pathway-specific exposures to PCE, the totals for multimedia pathway-specific exposures (e.g., inhalation [E_h], ingestion [E_g], and dermal contact [E_d]), and the total multimedia multiple-pathway exposure (i.e., $E_h + E_g + E_d$), which is a chronic daily intake (CDI).

Table 1. Summary of the processes by which contaminants are exchanged and lost among the seven compartments of the GEOTOX model.

Compartment	Gains	Losses
Air (gas phase of the troposphere)	Diffusion from soil and surface water.	Diffusion to soil; diffusion to surface water; washout by rainfall; and convection losses
Air Particles (atmospheric dust)	Resuspension of deposited soil particles.	Deposition from atmosphere to soil; deposition from air to surface water; and convective losses.
Upper-Soil Layer (surface-soil layer)	Diffusion from air; washout by rainfall; deposition of air particles; and irrigation from groundwater.	Diffusion to air; infiltration (leaching); resuspension of deposited soil particles; soil-solution runoff; and erosion.
Lower-Soil Layer (vadose zone)	Infiltration from upper-soil layer.	Infiltration to groundwater zone.
Ground-Water Zone	Infiltration from lower-soil.	Discharge to surface-water irrigation.
Surface Water	Diffusion from air; washout by rainfall; deposition of atmospheric particles; soil-solution runoff; erosion (mineral runoff); diffusion from sediment; and sediment resuspension.	Sediment deposition; diffusion to sediment; and surface-water outflow.
Sediment Layer	Diffusion from surface water; and sediment deposition (from surface water).	Diffusion to surface water; and sediment resuspension.

Table 2. Concentrations of PCE in the applicable environmental media of a 100-km^2 landscape assuming a soil-based steady-state source of contamination. Concentrations are predicted using the GEOTOX model, and are scaled to a soil concentration of PCE of 1.0 mg/kg.

Media (abbreviation;concentration units)	PCE concentration
Soil (C_s; mg/kg)	1.0
Air gases (C_a; mg/m^3)	1.4
Air particles (C_p; mg/m^3)	~ 0.0[a]
Potable water (C_w; mg/L)	0.21
Surface water (C_r; mg/L)	0.27

[a] There is some partitioning of PCE to particulates in air; however, the majority of PCE in the air compartment is in the gaseous phase and the very small amount present on particles is insignificant.

Table 3. Matrix of pathway-exposure factors (PEFs) for PCE in specific environmental media.

	PEFs associated with environmental medium concentration, C_i, where i is the type of medium[a]				
Pathway	Air (gas phase; m^3/kg-d), C_a	Air (particles; m^3/kg-d), C_p	Soil (kg/kg-d), C_s	Drinking water[b] (L/kg-d), C_w	Surface water (L/kg-d), C_r
Inhalation	F_{ah} (0.39)	F_{ph} (0.31)	F_{sh} (9.2×10^{-9})	F_{wh} (0.11)	—
Ingestion					
Water	—	—	—	F_{ww} (3.4×10^{-2})	—
Fruits and vegetables	F_{av} (1.6×10^{-4})	F_{pv} (14.0)	F_{sv} (1.1×10^{-3})	—	—
Grains	F_{ag} (2.5×10^{-4})	F_{pg} (22.0)	F_{sg} (8.0×10^{-4})	—	—
Meat	F_{at} (5.7×10^{6})	F_{pt} (2.8×10^{-2})	F_{st} (5.4×10^{-7})	F_{wt} (1.9×10^{-6})	—
Milk	F_{ak} (4.0×10^{-6})	F_{pk} (2.9×10^{-2})	F_{sk} (5.2×10^{-7})	F_{wk} (1.2×10^{-6})	—
Fish	—	—	—	—	F_{rf} (2.1×10^{-2})
Soil	—	—	F_{ss} (1.5×10^{-6})	—	—
Dermal contact	—	F_{sd} (2.6×10^{-6})	F_{wd} (3.8×10^{-2})	—	—

[a] Subscripts refer to the source media (a = air gases, p = air particles, s = soil, w = drinking water, and r = surface water) and pathways (h = inhalation, w = water ingestion, v = vegetables, g = grain, t = meat, k = milk, f = fish, s = soil ingestion, and d = dermal contact).

[b] Drinking-water concentrations are obtained by arithmetically averaging the concentrations in surface and groundwater so as to reflect the mix in a local-water supply. For calculation of a cleanup criterion, we assume half of the drinking water comes from groundwater and the other half from surface water.

Table 4. Summary of estimated exposures[a] to PCE (mg/kg-d). Values were calculated using the environmental concentrations of PCE predicted by GEOTOX for a 100 km^2 area in a typical California region (Table 2) and the PEFs listed in Table 3.

	Exposure					
Pathway	Air (gases)[b]	Air (particles)[c]	Soil[d]	Potable water[e]	Surface water[f]	Totals
Inhalation	5.5×10^{-1}	0.00	9.0×10^{-9}	2.3×10^{-2}	--	5.7×10^{-1} (E_h)[g]
Ingestion						
Water	--	--	--	7.1×10^{-3}	--	
Vegetables	2.2×10^{-4}	0.00	1.1×10^{-3}	--	--	
Grains	3.5×10^{-4}	0.00	8.0×10^{-4}	--	--	
Milk	5.6×10^{-6}	0.00	5.2×10^{-7}	2.5×10^{-7}	--	
Meat	8.0×10^{-6}	0.00	5.4×10^{-7}	4.0×10^{-7}	--	
Fish	--	--	--	--	5.7×10^{-3}	
Soil	--	--	1.5×10^{-6}	--	--	
Ingestion total	5.9×10^{-4}	0.00	1.9×10^{-3}	7.1×10^{-3}[h]	5.7×10^{-3}[h]	1.5×10^{-2} (E_g)[i]
Dermal uptake	--	--	2.6×10^{-6}	8.0×10^{-3}	--	8.0×10^{-3} (E_d)[j]
						5.9×10^{-1}(CDI)[k]

[a] Exposure = PEF (from Table 3) x Concentration (from Table 2).

[b] Concentration of PCE in the gaseous phase of air, $C_a = 1.4$ mg/m^3.

[c] Concentration of PCE in the particulate phase of air, $C_p = 0.00$ mg/m^3.

[d] Concentration of PCE in soil, $C_s = 1.0$ mg/kg.

[e] Concentration of PCE in potable water, $C_w = 0.21$ mg/L, which is the average of the concentrations of PCE in both surface and groundwater.

[f] Concentration of PCE in surface water, $C_r = 0.27$ mg/L.

[g] E_h = Total multimedia exposure to PCE from the inhalation exposure pathway.

[h] Total ingestion exposure due to PCE in potable and surface water = 1.3×10^{-2} mg/kg-d.

[i] E_g = Total multimedia exposure to PCE from the ingestion exposure pathway.

[j] E_d = Total multimedia exposure to PCE from dermal contact.

[k] CDI = Chronic daily intake associated with multimedia, mulitple-pathway exposure (i.e., $E_h + E_g + E_d$).

CARCINOGENIC POTENCY AND NONCARCINOGENIC THRESHOLD

The term carcinogenic "potency" is used here to refer to the quantitative expression of increased tumorigenic response per unit dose rate at very low doses. The noncarcinogenic threshold applies to the daily exposure over the course of a lifetime that is likely to be without appreciable risk of noncarcinogenic deleterious effects; this safe level is referred to as the reference dose (RfD).

Carcinogenic Potency

We used the range of PCE cancer-potency estimates calculated by Bogen et al. (1987) from rodent tumor-incidence data published by the NCI (1977) and NTP (1986). These potency estimates were calculated as a function of metabolized dose of PCE because there is evidence that it is a product of PCE's metabolism that is responsible for PCE's carcinogenicity (U.S. EPA, 1985; Bogen et al., 1987). We relied on these values instead of those derived by the U.S. EPA (1985 and 1986), because the potencies derived by Bogen et al. (1987) were adjusted to include data on animal body weight, metabolite elimination, and other factors not addressed in the calculations by the U.S. EPA (1985 and 1986). Bogen et al. (1987) used the term $q_1^*(M)$ to specify the 95% upper-confidence limit (UCL) estimate of potency. This term relates low levels of metabolized dose, M, to predicted upper-bound increased tumor risk, R*, by the linear approximation $R^* \sim q_1^*(M) \times (M)$.

To estimate the carcinogenic potency of PCE in terms of human-applied dose, A, which is necessary for calculation of an acceptable-exposure limit (because exposure is expressed in terms of applied and not metabolized dose), the term $q_1^*(M)$ is multiplied by f, the fraction of the applied dose that is metabolized (i.e., $q_1^*(A) = q_1^*(M) \times f$). We calculated the arithmetic means of the fraction metabolized of an orally-acquired dose of PCE, f^*_{mo}, or a respired or dermally-acquired dose of PCE, f^*_{mr}, using the respective ranges of values for these parameters developed by Bogen and McKone (1988) from the data of Ikeda et al. (1972) and Ohtsuki et al. (1983). Table 5 presents the highest and lowest estimates of the pathway-specific values of $q_1^*(A)$ based on the lowest and highest values of $q_1^*(M)$ calculated by Bogen et al. (1987) and based on the mean values of f^*_{mo} (26%) and f^*_{mr} (20%).

Table 5. Range of carcinogenic-potency estimates for PCE for ingestion, respiration, or dermal exposure.

Metabolized potency, $q_1^*(M)$[b] (mg M/kg-d)$^{-1}$	Applied potency, $q_1^*(A)$[a] (mg A/kg-d)$^{-1}$	
	Ingestion[c]	Inhalation or dermal uptake[d]
0.095	0.025	0.019
0.42	0.11	0.084

[a] 95%-UCL potency of human applied dose, A, based on surface-area, interspecies dose-extrapolation method. Note that $q_1^*(A) = q_1^*(M) \times f$, where $f = f^*_{mo}$ or f^*_{mr}.

[b] Range of 95%-UCL potency of human metabolized dose, M, based on surface-area, interspecies dose-extrapolation method. Values from Bogen et al. (1987).

[c] Calculated using the arithmetic mean of $f^*_{mo} = 0.26$.

[d] Calculated using the arithmetic mean of $f^*_{mr} = 0.20$. It is assumed that the fraction of applied dose that is metabolized is the same for inhalation and dermal contact (see Bogen, 1988).

Noncarcinogenic Hazard Index

According to the U.S. EPA (1989) the ratio of the total calculated multimedia exposure, expressed as a CDI, to the RfD is the hazard index. If this ratio exceeds one, then there is a potential for the exposure to yield a noncarcinogenic adverse health effect. Because only an oral RfD for PCE of 1×10^{-2} mg/kg-d is available (U.S. EPA, 1990a), we use this value in our calculations.

DERIVATION OF RISK-BASED SOIL CONCENTRATIONS FOR PCE

To derive a range of risk-based concentrations of PCE in soil as possible cleanup criterion, we first calculate the total carcinogenic risk and noncarcinogenic hazard index associated with 1 ppm of PCE in soil. We then use these values in the derivation of alternatives as cleanup criterion for PCE in soil. The alternatives we derive correspond to each of three levels of excess individual-lifetime cancer risk–1 per 10,000 (i.e., 10^{-4}), 1 per 100,000 (i.e., 10^{-5}), and 1 per 1,000,000 (i.e., 10^{-6}), and to a hazard index that does not exceed one. The three levels of excess individual-lifetime cancer risk were selected because they are cited by regulatory agencies (see U.S. EPA, 1990b and CDHS, 1989) as being acceptable levels of risk associated with exposure to a carcinogen present in environmental media.

Calculated Total Risk for 1 PPM of PCE in Soil

For a 100-km^2 landscape in a typical California region, the calculated total risk associated with 1 ppm of PCE in soil [R_{TC}(1 ppm)] is determined using Equation 2:

$$R_{TC}(1ppm) = (E_h \cdot CPF_h) + (E_g \cdot CPF_g) + (E_d \cdot CPF_d), \quad (2)$$

where

$R_{TC}(1ppm)$ = the *calculated* total risk associated with 1 ppm of PCE in soil;

E_h = inhalation intake related to multimedia exposure resulting from a soil-based PCE concentration of 1 ppm (mg/kg-d);

E_g = ingestion intake related to multimedia exposure resulting from a soil-based PCE concentration of 1 ppm (mg/kg-d);

E_d = dermal contact related to multimedia exposure resulting from a soil-based PCE concentration of 1 ppm (mg/kg-d);

CPF_h = maximum or minimum cancer-potency factor for the inhalation pathway (1/[mg/kg-d]);

CPF_g = maximum or minimum cancer-potency factor for the ingestion pathway (1/[mg/kg-d]); and

CPF_d = maximum or minimum cancer-potency factor for the dermal-contact pathway (1/[mg/kg-d]).

Cleanup Alternatives

We calculate a cleanup concentration of PCE in soil, $C_s(cleanup)_i$, corresponding to a specified level of total acceptable carcinogenic risk, R_{TA} (e.g., 10^{-6}), and to a noncarcinogenic hazard index of one

by scaling the steady-state concentration of PCE in soil, C_S(1 ppm), by the ratio of R_{TA}(cleanup) to R_{TC}(1 ppm) as shown in Eq. 3 (where i = c), or by the ratio of 1 to CDI/RfD as shown in Eq. 4 (where i = h).

$$C_S(\text{cleanup})_c = C_S(1\text{ppm}) \times [R_{TA}(\text{cleanup})/R_{TC}(1\text{ppm})], \text{ and} \quad (3)$$

$$C_S(\text{cleanup})_h = C_S(1\text{ ppm}) \times \left[\frac{1}{\frac{CDI}{RfD}}\right] . \quad (4)$$

Table 6 shows alternative cleanup concentrations for PCE corresponding to low and high cancer-potency estimates and acceptable levels of carcinogenic risk that range from 10^{-4} to 10^{-6}, and corresponding to a hazard index of one.

CONCLUSION

The PCE concentrations presented in Table 6 constitute a range of risk-based cleanup levels for consideration by risk managers for a 100-km^2 California landscape. However, the methodology described here can be used to develop other more or less conservative values for consideration. Nevertheless, it is apparent that the concentrations of PCE in Table 6 that correspond to carcinogenic risk ranging from 10^{-4} to 10^{-6} are all lower than the concentration of PCE associated with a noncarcinogenic hazard index equal to one. Consequently, the concentration of PCE corresponding to a hazard index of one may be rejected from consideration because it is associated with a carcinogenic risk above the range of acceptability (i.e., 10^{-4} to 10^{-6}), although it is likely to be without appreciable risk of noncarcinogenic deleterious effects. Nothwithstanding, selection of a landscape-specific cleanup criterion for any chemical in soil should also include consideration of criteria such as likelihood of public exposure, size and overall susceptibility of the population at risk, and the principal exposure pathway, as well as uncertainties in the input parameters and final results.

Finally, adopting a scientifically credible landscape-specific risk-based cleanup criterion for a chemical in soil will benefit society by serving as a mechanism to protect public health while justifying decisions about cleaning up or not cleaning up contaminated sites. Such decisions are important to our society because they will reduce the enormous overall cost that is now contemplated for cleaning up every contaminated site completely and that threatens to cripple the nation's economic competitiveness.

Table 6. Risk-based concentrations of PCE in soil for consideration as a cleanup criterion for a 100-km^2 landscape representing a typical California region.

Acceptable level of risk[a] or noncarcinogenic hazard	Soil Concentration (C_S[cleanup]; mg/kg)	
	Low Potency[b]	High Potency[b]
10^{-4}	9×10^{-3}	2×10^{-3}
10^{-5}	9×10^{-4}	2×10^{-4}
10^{-6}	9×10^{-5}	2×10^{-5}
Hazard index (CDI/RfD) = 1	2×10^{-2}	

[a] Risk is the incremental probability of an individual developing cancer over a 70-y lifetime as a consequence of multipathway exposure (i.e., from ingestion, inhalation, and dermal contact) to soil-based concentrations of PCE in multiple enviromental media (i.e., soil, air, and water).

[b] The terms "low" and "high" refer to the range of cancer-potency estimates for PCE listed in Table 5.

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