CONF-910508--1

UCRL-JC-105085
PREPRINT

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This paper was prepared for submittal to
Joint International Symposium on
Environmental Consequences of
Hazardous Waste Disposal
May 27-31, 1991
Stockholm, Sweden

June 1991

Lawrence
Livermore
National
Laboratory

MODELING HUMAN EXPOSURE TO HAZARDOUS-WASTE SITES: A QUESTION OF COMPLETENESS

UCRL-JC--105085

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ABSTRACT

In risk analysis, we use human-exposure assessments to translate contaminant sources into quantitative estimates of the amount of contaminant that comes in contact with human-environment boundaries, that is, the lungs, the gastrointestinal tract, and the skin surface of individuals within a specified population. An assessment of intake requires that we determine how much crosses these boundaries. Exposure assessments often rely implicitly on the assumption that exposure can be linked by simple parameters to ambient concentrations in air, water, and soil. However, more realistic exposure models require that we abandon such simple assumptions. To link contaminant concentrations in water, air, or soil with potential human intakes, we construct pathway-exposure factors (PEFs). For each PEF we combine information on environmental partitioning as well as human anatomy, physiology, and behavior patterns into an algebraic term that converts concentrations of contaminants (in mg/L water, mg/m^3 air, and mg/kg soil) into a daily intake per unit body weight in mg/kg-d for a specific route of exposure such as inhalation, ingestion, or dermal uptake. Using examples involving human exposure to either a radionuclide (tritium, ^{3}H) or a toxic organic chemical (tetrachloroethylene, PCE) in soil, water, and air, we illustrate the use of PEFs and consider the implications for risk assessment.

INTRODUCTION

When contaminants enter the environment, they contact people through air, water, food, and soil in varying amounts throughout a lifetime. Therefore, managing the health and environmental risks associated with environmental contaminants requires an integrated and comprehensive assessment of multimedia transport and human exposure. In response to this need, a number of multimedia monitoring and modeling efforts have emerged (Allen *et al.*, 1989). However, the need still remains to integrate multimedia-concentration data with more realistic models of human exposure. Our approach to achieving this goal is to develop credible pathway-exposure factors (PEFs) to link contaminant concentrations in multiple environmental media with potential chronic daily intake (CDI).

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The process of estimating exposure often involves using limited data and extrapolating to a large diverse population, which means that many assumptions, inferences, and simplifications are required. Accordingly, there are many sources of uncertainty, and many of these uncertainties defy quantification. However, the principal product of our approach is a matrix of pathway-specific exposure values, and developing this matrix not only facilitates investigating the sufficiency of the data and the efficacy of the exposure model(s) used for each pathway, but also permits addressing and comparing simultaneously the magnitudes of the potential exposures by each of the possible routes.

We begin by assuming that exposure is determined by measuring or estimating the amount of an agent available at the environment→human-exchange boundaries (i.e., lungs, gut, and/or skin) during some specified time. Next, we introduce the concept of the PEF, where we combine information on environmental partitioning as well as human anatomy, physiology, and behavior patterns into an algebraic term that converts concentrations of contaminants (in mg/L water, mg/m^3 air, and mg/kg soil) into a daily intake per unit body weight in mg/kg-d for a specific route of exposure. We also mention how the PEF approach compares to other methods used for exposure assessment. We then review the most credible exposure pathways linking human populations to environmental contaminants in air, water, and soil. Finally, we use examples for exposures to a radionuclide (tritium, ^{3}H) and to a toxic organic chemical (tetrachloroethylene, PCE) in soil, water, and air to illustrate the use of PEFs and to consider the implications for risk assessment.

PATHWAY-EXPOSURE FACTORS (PEFs)

Exposure assessments often rely implicitly on the assumption that exposure to environmental contaminants can be linked by simple parameters directly to concentrations of such contaminants in ambient environmental media (e.g., air, water, and soil). For example, 2 L/d is used commonly for humans as the total contact rate with water and 20 m^3/d is used typically for humans as the total contact rate with ambient air. However, exposure assessments such as the TEAM (Wallace, 1989) and Harvard Health (six-cities) studies (Spengler *et al.*, 1981) reveal the inaccuracy of these parameters given more detailed information concerning realistic human-activity patterns and microenvironmental data. Consequently, research on exposure should be based on the premise that an exposure assessment is most valuable when it provides a comprehensive view of exposure routes and identifies major sources of uncertainty.

To link contaminant concentrations in multiple environmental media to potential human exposure comprehensively and completely, we derive and employ pathway-exposure factors (PEFs) (McKone, 1987; McKone, 1990; McKone and Daniels, 1991). As mentioned above, these PEFs account for environmental partitioning and for human anatomy, physiology, and behavior patterns. The construction of a PEF usually involves many details and can be quite complicated. Here, for illustration purposes only, a PEF is derived for the potential intake by a 70-kg adult of a contaminant contained in drinking water, which is consumed at a rate of 2 L/d. Accordingly, the PEF in this illustration is calculated as 2 L/d divided by 70 kg or 0.029 L/kg-d. When this PEF value is multiplied by the contaminant concentration in the water in units of mg/L, we obtain an estimate for intake that is expressed in units of mg/kg-d.

Generally, when environmental concentrations of a contaminant are constant over time, the population-average exposure, expressed as the chronic daily intake (CDI), to a contaminant concentration C_k (measured or modeled) in environmental medium "k" (i.e., air, water, or soil) is expressed as the product of the concentration and the pathway exposure factor, PEF(k→i):

$$\mathrm{CDI} = \left[\frac{CR_i}{BW}\right] \times \frac{C_i}{C_k} \times \frac{EF \times ED}{AT} \times C_k = \mathrm{PEF}(k \rightarrow i) \times C_k \;, \tag{1}$$

where $[CR_i/BW]$ is the contact rate per kg body weight, such as kg(soil)/kg-d, L(milk)/kg-d, m^3(air)/kg-d, etc.; C_i/C_k is the ratio of concentration in the contact medium *i* (i.e., air, tap water, milk, soil, etc.) to the concentration in environmental medium *k* (air, water, soil), units depend on the two media; EF is the exposure frequency, days per year; ED is the exposure duration, years; AT is the averaging time, days; and PEF(k→i) is the pathway exposure factor relating the concentration C_k in medium *k* to the chronic daily intake, mg/kg-d, during the period ED. This expression makes the PEF consistent with U.S. Environmental Protection Agency (U.S. EPA) guidelines for estimating chronic daily intakes (U.S. EPA, 1989a).

Table 1 contains examples of the linkages between environmental concentrations of contaminants in air, water (surface and/or ground water), and soil and the potential multiple pathways of exposure for humans. The models and assumptions used to develop PEFs for each linkage are covered in numerous publications, but a useful overview of the information needed and how to apply it is available in two U.S. EPA reports—*Risk Assessment Guidance for Superfund, Volume 1. Human Health Evaluation Manual (Part A), Interim Final* (U.S. EPA, 1989a) and the *Exposure Factors Handbook* (U.S. EPA, 1989b)—as well as in an overview paper by McKone and Daniels (1991). Table 2 contains the equations used to derive PEFs for ^3H and for PCE. The definitions and numerical values (applicable to

Table 1. Matrix containing examples of the relationships between contaminant concentrations in environmental media and potential exposure pathways.

Pathways	Media		
	Air	**Soil**	**Water**
Inhalation	• Inhalation of ambient outdoor air • Inhalation of indoor air	• Inhalation of soil vapors that migrate to indoor air • Inhalation of soil particles transferred to indoor air	• Inhalation of contaminants transferred from tap water to indoor air
Ingestion	• Ingestion of fruits, vegetables, and grains contaminated by atmospheric particle deposition • Ingestion of fruits, vegetables, and grains contaminated by foliar uptake from the gas-phase of the atmosphere • Ingestion of meat, milk, and eggs contaminated through inhalation by animals	• Human soil ingestion • Ingestion of meat, milk, and eggs contaminated through soil ingestion by animals • Ingestion of fruits, vegetables, and grains contaminated by transfer from soil • Ingestion of meat, milk, and eggs contaminated by transfer from soil to plants to animals	• Ingestion of tap water • Ingestion of irrigated fruits, vegetables, and grains • Ingestion of meat, milk, and eggs from animals consuming contaminated water • Ingestion of fish and sea food
Dermal contact	• Dermal contact with vapor-phase contaminants	Dermal contact with soil	Dermal contact in baths and showers

^3H and PCE) for the algebraic terms shown in Table 2 are described in the notes at the bottom of that table. The plant/air partition coefficient, K_{pa}, is used to relate the concentration of a substance in the fresh mass of a plant to that in air at equilibrium. Similarly, the soil/plant partition coefficient, K_{sp}, is used to relate the concentration of

Table 2. Summary matrix of the mathematical expressions used to derive pathway-exposure factors (PEFs).

	Environmental media			
Pathway	Air (gas phase) (m^3/kg-d)	Soil (kg/kg-d)	Potable water (L/kg-d)	Surface water (L/kg-d)
Inhalation	0.39	9.0×10^{-9}	$3.2 \times 10^5 \times \left[2.5/D_\ell^{2/3} + RT/(D_a^{2/3} H)\right]^{-1}$	—
Ingestion				
Water	—	—	0.034	—
Fruits and vegetables	$0.0025 \times K_{pa}$	$0.0011 \times K_{sp}$	—	—
Grains	$0.0040 \times K_{pa}$	$0.00079 \times K_{sp}$	—	—
Meat	$[0.38 + (0.186 \times K_{pa})] \times B_t$	$[0.0012 + (0.038 \times K_{sp})] \times B_t$	$0.14 \times B_t$	—
Milk	$[0.68 + (0.476 \times K_{pa})] \times B_k$	$[0.0022 + (0.10 \times K_{sp})] \times B_k$	$0.27 \times B_k$	—
Fish	—	—	—	$0.00032 \times BCF$
Soil	—	1.5×10^{-6}	—	—
Dermal uptake	—	2.6×10^{-6}	0.037 (only for organic compounds; see Table 3 for ^3H)	

Notes: D_ℓ = diffusion coefficient in water (m^2/sec)—we use values for this parameter of 2.4×10^{-9} m^2/sec for ^3H and 7.6×10^{-10} m^2/sec for PCE; D_a = diffusion coefficient in air (m^2/sec)—we use values for this parameter of 2.4×10^{-5} m^2/sec for ^3H and 7.4×10^{-6} m^2/sec for PCE; R = the gas constant, 8.31 Pa-m^3/mol-K; T = temperature, 293 K; H = Henry's law constant (Pa-m^3/mol)—we use values for this parameter of 6.3×10^{-2} Pa-m^3/mol for ^3H and 2.2×10^3 Pa-m^3/mol for PCE; K_{pa} = plant/air partition coefficient [m^3 (air)/kg (plant fresh mass)]—we use values for this parameter of 45 m^3/kg for ^3H and 4.4×10^{-2} m^3/kg for PCE; K_{sp} = soil/plant (dry mass) partition coefficient (dimensionless)—we use values for this parameter of 40 for ^3H and 2.0 for PCE; B_t = meat/diet biotransfer factor (days/kg)—we use values for this parameter of 9.5×10^{-3} d/kg for ^3H and 1.0×10^{-5} d/kg for PCE; B_k = milk/diet biotransfer factor (days/L)—we use values for this parameter of 7.5×10^{-3} d/L for ^3H and 3.2×10^{-6} d/L for PCE; and BCF = bioconcentration factor (dimensionless)—we use values for this parameter of 1.0 for ^3H and 39 for PCE.

a substance in the dry mass of a plant to that in the soil, at equilibrium, and for purposes of our examples this term can be considered dimensionless.

USING PEFS FOR ASSESSING EXPOSURE TO TRITIUM AND PCE

Tritium (^3H) is the heaviest and only radioactive isotope of hydrogen. Natural production due to cosmic rays is approximately 1.8×10^{17} Bq/y (NCRP, 1979), corresponding to 1 Bq/km^2-d globally. Large amounts of tritium have also been produced by atomic energy programs worldwide. Typically, tritium appears in the form of tritiated water, which is transferred from the atmosphere to the surface of the earth by precipitation and by vapor exchange (NCRP, 1979). Tetrachloroethylene (PCE) is a volatile organic compound (VOC) that is ubiquitous in the environment of industrial nations because of its usefulness as a solvent, cleaner, or degreasing agent and is suspected to be a human carcinogen (U.S. EPA, 1990).

The pathway-exposure factors for ^3H and PCE corresponding to specific environmental media (derived using the mathematical expressions appearing in Table 2) are shown in Tables 3 and 4, respectively. For each substance, the product of a specific PEF and a corresponding modeled or monitored media-specific concentration yields a pathway- and media-specific CDI. However, even without calculating medium- and pathway-specific CDIs, the matrices of PEF values presented in Tables 3 and 4 identify the relative magnitudes of the pathway(s) for exposure associated with each environmental medium. For example, the results in Table 3 reveal that for ^3H some pathways are indeed trivial, that is, soil inhalation, direct soil ingestion, and dermal uptake from soil contribute little to total exposure from soil. Similarly, dermal uptake of ^3H from water constitutes a trivial contribution to total exposure from water. However, among the medium-specific pathways that remain, none clearly dominates in its respective environmental media. This is to be expected because ^3H partitions as water—a constituent of all environmental media—and PEFs account for this partitioning, as well as the human physiology and human-behavior patterns that determine the degree of human contact (e.g., skin is a good barrier to water). Alternatively, PCE, as a consequence of its volatility and partitioning, has dominant exposure pathways in specific environmental media—inhalation for air (gases) and ingestion of food crops for soil. Additionally, three pathways—inhalation, direct ingestion, and dermal uptake are significant for exposure to PCE in potable water. In fact, the PEF for PCE in potable water is greater for inhalation than

Table 3. Matrix of environmental media and pathway-exposure factors (PEFs) for tritium (^3H).

	Environmental media			
Pathway	Air (gas phase) (m^3/kg-d)	Soil (kg/kg-d)	Potable water (L/kg-d)	Surface water (L/kg-d)
Inhalation	3.9×10^{-1}	9.0×10^{-9}	6.7×10^{-3}	—
Ingestion				
Water	—	—	3.4×10^{-2}	—
Fruits and vegetables	1.1×10^{-1}	4.4×10^{-2}	—	—
Grains	1.8×10^{-1}	3.2×10^{-2}	—	—
Meat	8.3×10^{-2}	1.4×10^{-2}	1.3×10^{-3}	—
Milk	1.7×10^{-1}	3.0×10^{-2}	2.0×10^{-3}	—
Fish	—	—	—	3.2×10^{-4}
Soil	—	1.5×10^{-6}	—	—
Dermal uptake	—	2.6×10^{-6}	2.0×10^{-5}[a]	

[a] Corresponding expression in Table 2 has been adjusted to account for dermal uptake of water.

Table 4. Matrix of environmental media and pathway-exposure factors (PEFs) for tetrachloroethylene (PCE).

	Environmental media			
Pathway	Air (gas phase) (m^3/kg-d)	Soil (kg/kg-d)	Potable water (L/kg-d)	Surface water (L/kg-d)
Inhalation	3.9×10^{-1}	9.0×10^{-9}	1.1×10^{-1}	—
Ingestion				
Water	—	—	3.4×10^{-2}	—
Fruits and vegetables	1.1×10^{-4}	2.2×10^{-3}	—	—
Grains	1.8×10^{-4}	1.6×10^{-3}	—	—
Meat	3.9×10^{-6}	7.7×10^{-7}	1.4×10^{-6}	—
Milk	2.2×10^{-6}	6.5×10^{-7}	8.6×10^{-7}	—
Fish	—	—	—	1.2×10^{-2}
Soil	—	1.5×10^{-6}	—	—
Dermal uptake	—	2.6×10^{-6}	3.7×10^{-2}	

for ingestion because PCE in tap water is easily volatilized to indoor air, particularly in showers (see McKone, 1987). Finally, because the bioconcentration factor (BCF) for PCE from surface water to fish exceeds that for ^3H (assumed to be one), the associated PEF for PCE is substantially larger than that for ^3H.

UNCERTAINTIES

The mathematical models used to generate the PEFs only approximate real systems, and therefore their predictions are inherently uncertain. For example,

- The partitioning of contaminants between air and/or soil and vegetation (such as food and pasture crops) is not well known. Using what is known about organic chemicals, for which this parameter has been measured, provides a way of making order-of-magnitude estimates of plant/air and plant/soil partitioning factors. In fact, Travis and Arms (1988) have developed an estimating equation for plant/soil partitioning based on the octanol-water partition coefficient. But for many organic compounds, the uncertainty in such estimates can be orders of magnitude (McKone and Ryan, 1989). This has important implications for any compound for which the ingestion of meat, milk, fruits, vegetables, and grains is likely to be an important pathway.

- The biotransfer factors, which express contaminant partitioning between animal intake and animal-based food products (such as meat, dairy products, and eggs), probably can only be estimated within about two orders of magnitude for many organic chemical species (McKone and Ryan, 1989). This limitation has important implications for situations where these exposure pathways are potentially important.

- Finally, dermal uptake of chemicals can result from contact with soil and with bath, shower, and swimming water. The PEF for dermal contact, particularly for organic compounds, must be based on limited data defining the permeability of the skin to chemical transport. Until more data on skin permeability is available, these PEFs will also be highly uncertain.

Consequently, when possible such uncertainties are best addressed when exposure and risk are presented as probability distributions so that uncertainty in risk and exposure can be characterized by expectation (mean) and spread (variance).

CONCLUSION

The procedures described here and the results from the examples that have been presented indicate that decision makers confronting the health consequences of toxic substances released from hazardous-waste sites to the environment as a result of industrial economies should (1) consider the potential for multiple exposure pathways, (2) compare relative contributions to total exposure from multiple exposure pathways, (3) be aware of the uncertainty in risk estimates, and (4) incorporate these considerations and judgments into the decision-making process. Such informed decisions will be defensible scientifically and can help focus limited resources on the most important problems.

ACKNOWLEDGMENTS

This paper describes work being performed under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

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