

Using an uncertainty analysis of direct and indirect exposure to contaminated groundwater to evaluate EPA's MCLs and health-based cleanup goals*

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Abstract

Groundwater which has been contaminated by industrial chemicals has been of significant concern in the U.S. since about 1975. Since then, dozens of regulatory decisions have been made to remediate many of these contaminated aquifers. The selected groundwater clean-up levels will dictate the cost and time frame of the remediation. Most clean-up decisions have been based either on EPA's Maximum Contaminant Levels (MCL) or so-called 'risk-based' levels. For many chemicals, risk-based levels are much lower than the corresponding MCLs. This paper uses an uncertainty analysis of probability density functions (PDF) to assess whether MCLs are sufficient to provide health protection for human populations using remediated groundwater as the sole tap-water source. A case-study involving tetrachloroethylene and chloroform and all the potentially direct and indirect routes of exposure to contaminated water is presented. The results suggest that groundwater need not be cleaned-up to concentrations less than drinking water standards (i.e., MCLs) to achieve health protection.

Introduction

Maximum Contaminant Levels (MCLs) are maximally acceptable concentrations of chemicals in a public water system that are set by U.S. EPA (Environmental Protection Agency) in accordance with the 1972 Safe Drinking Water Act. The MCLs are derived from health-based criteria in conjunction with technologic and economic factors relating to the feasibility of achieving and monitoring for these concentrations in water supply systems. Due to the balancing of health effect considerations with technologic and economic factors, the MCLs for many chemicals are much higher than their respective 'de minimis risk' concentrations (defined here as the concentration associated with

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one excess cancer case in a million individuals drinking two liters of water per day for 70 years). For example, the primary MCLs for chloroform ($100 \mu\text{g/L}$), and tetrachloroethylene ($5 \mu\text{g/L}$) are one to two orders of magnitude higher than their corresponding *de minimis* concentrations ($5.7 \mu\text{g/L}$ and $0.69 \mu\text{g/L}$, respectively). This dichotomy is analogous to many of the chemical-specific workplace ambient air standards (Threshold Limit Values, etc.) which are set partially on technical considerations and can be significantly higher than purely 'risk-based' standards [1].

One of the outcomes of such technical and economical concessions is that contaminated groundwater remedial goals, which are often strictly health-based, are sometimes set at concentrations far below drinking water standards. For example, the State of California's 'Recommended Public Health Levels' (RPHLs) for contaminants in drinking water, which are essentially *de minimis* concentrations, have been implemented as cleanup goals at several state and federal Superfund sites in California. The existence of such goals can create technical, financial, and legal dilemmas for the parties faced with cleaning groundwater to concentrations far below the levels that are generally considered safe for daily human consumption. On a more global scale, this issue begs the question as to whether such measures are truly necessary and whether limited resources might be better devoted to other environmental problems.

To date, health-based soil and groundwater remediation goals have largely been driven by the use of fairly conservative point estimates of exposure. For example, current U.S. EPA guidance for risk assessment at Superfund sites [2] suggests using either the maximum or the 95% upper confidence limit of the arithmetic mean of the measured contaminant concentrations and the 90th or 95th percentile of the available estimates of exposure rate and duration (e.g., soil ingestion rates, breathing rates, etc.) to assess contaminant uptake. As has been discussed extensively in the literature, the major shortcoming inherent in this approach is that repeated use of upper-bound values throughout an exposure assessment is likely to result in unrealistic estimates of risk and unreasonable cleanup goals [5,6]. As an alternative, it has been suggested that the exposure assessment process could be refined if probability density functions (PDFs), rather than point exposure estimates, were incorporated into the exposure analysis [3-5]. Specifically, instead of using a single value to represent the exposure parameter, each exposure variable takes on a range of values with a known probability. These PDFs are then analyzed statistically to develop a range of estimated risks and associated probabilities. This 'probabilistic' approach places the point estimate into a full and proper context, and provides more information to the risk managers and the public.

A review of the recent scientific literature indicates that Monte Carlo analysis of exposure parameter PDFs appears to be the current probabilistic approach of choice [4-8]. In the initial step of a Monte Carlo analysis, the available data for each exposure parameter are evaluated with respect to distribution

type (e.g., normal, log-normal, etc.) and the mean, maximum, and minimum values are identified. Commercially available software programs (e.g., @Risk™, Crystal Ball™) then simulate a full distribution frequency for the parameter based on these descriptors. If only the range of values is known, a uniform distribution may be assigned to the exposure parameter. If only the range and mode are known, a triangular distribution may be most appropriate. In addition, if the data set is not accurately described by a standard distribution (normal, lognormal, etc.) it is considered appropriate to use only the empirical data points themselves ('bootstrapping') rather than attempting to simulate a PDF [6]. In the next step, the risk calculation equation is solved several thousand times using a Monte Carlo program which draws values from each exposure PDF. This results in a distribution of risk values and associated probabilities.

In this paper, we examine the health risks associated with daily exposure to MCL concentrations of tetrachloroethylene and chloroform using a Monte Carlo analysis of exposure PDFs. The exposure pathways evaluated are: direct ingestion, dermal contact while showering, indoor inhalation, and garden vegetable ingestion. This scenario comprises all of the likely pathways of exposure to tapwater contaminants in the household. The purpose of this examination is to: (1) assess the degree of conservatism associated with the MCLs, (2) determine whether in fact the MCLs are protective for a vast majority of the population via *all* exposure pathways, and (3) provide a preliminary assessment as to whether groundwater remedial goals should be limited to drinking water standards.

Methods

This section describes the dose equations and data sources for each exposure variable. For the purposes of this evaluation, a single adult age group is considered. Tables 1 through 3 summarize the data distribution characteristics for each exposure variable.

Tapwater ingestion

Contaminant uptake via tapwater ingestion is described by the following equation:

$$Dose = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (1)$$

where *Dose* is in mg/kg day, *C* denotes the chemical concentration in water (mg/L), *IR* is the ingestion rate (L/day), *EF* the exposure frequency (day/year), *ED* the exposure duration (years), *BW* the body weight (kg), and *AT* the averaging time (days).

For the purposes of this evaluation, the rate of tapwater intake (the sum of water drunk directly as a beverage and water added to foods and beverages

TABLE 1

Probability density functions for exposure duration and frequency, averaging time, and body weight

Parameter	Distribution	Arithmetic mean	Minimum	Maximum	Source
Exposure duration (years)	Empirical	12.9	—	—	Census Bureau [11]
Exposure frequency (days/year)	Constant	350	—	—	USEPA [10]
Averaging time (days)	Constant	25,550	—	—	USEPA [2]
Body weight (kg)	Uniform	—	46.8	101.7	USEPA [12]

during preparation) is taken to be uniformly distributed between 0.4 and 2.2 L/day, as described by data presented by the International Commission on Radiologic Protection in the Report of the Task Group on Reference Man [9]. Exposure frequency is set at a point estimate of 350 days/year, per U.S. EPA guidance [10]. Exposure duration is taken to be an empirical distribution of data collected by the U.S. Census Bureau in 1980 [11]. The arithmetic mean of this distribution, which describes the period of residential tenure of a single household, is 12.9 years. Averaging time is 25,550 days per U.S. EPA guidance [2]. A uniform PDF for body weight is constructed from data collected in the Second National Health and Nutrition Examination Survey (NHANES II), in which adult body weights ranged from 46.8 kg (5th percentile, female) to 101.7 kg (95th percentile, males) [12]. The chemical concentration in water is the chemical-specific MCL.

Dermal contact

Dermal uptake of contaminants in tapwater during showering and bathing is described by the following equation:

$$Dose = \frac{C \times SA \times PC \times F \times CF \times ET \times ED}{BW \times AT} \quad (2)$$

where *Dose* is in mg/kg day, *C* denotes the chemical concentration in water (mg/L), *SA* the surface area of exposed skin (cm²), *PC* the STCT coefficient (cm/h), *F* the fraction of skin in contact with water (unitless), *CF* the conversion factor (10⁻³L/cm³), *ET* the exposure time (h/day), *EF* the exposure frequency (days/year), *ED* the exposure duration (year), *BW* the body weight (kg), and *AT* the averaging time (days).

TABLE 2

Pathway-specific probability density functions

Pathway	Distribution	Mean	Standard Deviation	Minimum	Maximum	Source
<i>Tapwater ingestion</i>						
Rate (L/day)	Uniform	—	—	0.4	2.2	[9]
<i>Dermal contact</i>						
Skin Surface (cm ²)	Normal	17,000 ^a	1,000 ^a	—	—	[7]
Shower Exposure Time (h/day)	Log-normal	0.11 ^b	1.8 ^b	—	—	[14]
Fraction of exposed skin (unitless)	Uniform	—	—	0.4	0.9	[6]
<i>Inhalation</i>						
<i>Water Use Rates (L/h)</i>						
Shower	Log-normal	460 ^b	1.4 ^b	—	—	[14]
House	Log-normal	37 ^b	1.4 ^b	—	—	[6]
<i>Air Exchange Rates (m³/h)</i>						
Shower	Uniform	—	—	4	20	[6]
Bath	Uniform	—	—	10	100	[6]
House	Uniform	—	—	300	1200	[6]
<i>Exposure Time (h/day)</i>						
Shower	Log-normal	0.11 ^b	1.8 ^b	—	—	[6]
Bath	Log-normal	0.27 ^b	1.8 ^b	—	—	[6]
House	Uniform	—	—	8	20	[6]
Inhalation rate (m ³ /h)	Uniform	—	—	0.21	0.74	[17]
<i>Vegetable ingestion</i>						
Rate (kg/day)	Log-normal	0.062 ^b	1.8 ^b	—	—	[7]
Fraction homegrown (%)	Empirical	—	—	18%	47%	[17]

^a Arithmetic mean.^b Geometric mean.

The surface area of adult skin is taken to be normally distributed with a mean of 17,000 cm² and an arithmetic standard deviation of 1,000 cm², as reported in McKone's and Bogen study of household exposure models [7]. The permeability coefficient for each chemical is assigned a uniform distribution between 0.4 and 1.0 cm/h, based on the data reported by Brown et al. [13] for volatile contaminants. Based on data compiled by James and Knuiman [14], the duration of contact during showering is lognormally distributed with a geometric mean of 0.11 hour/day and a generic standard deviation of 1.8 hours/day [6]. The fraction of skin surface area in contact with water is taken to be

TABLE 3

Chemical-specific distributions

Parameter	Distribution	Geometric mean	Mode	Geometric standard deviation	Minimum	Maximum	Source
<i>Dermal permeability coefficient (cm/h)</i>							
PCE ^a , Chloroform	Uniform	—	—	—	0.4	1.0	[13]
<i>Transfer efficiency from water to shower air (unitless)</i>							
PCE	Triangular	—	0.6	—	0.1	0.9	[6]
Chloroform	Triangular	—	0.6	—	0.1	0.9	[6]
<i>Transfer efficiency from water to household air (unitless)</i>							
PCE	Triangular	—	0.3	—	0.1	0.9	[6]
Chloroform	Triangular	—	0.3	—	0.1	0.9	[6]
<i>Soil-water partition coefficient (L/kg)</i>							
PCE	Triangular	—	2.4	—	0.0	34	[7]
Chloroform	Triangular	—	0.58	—	0.0	3.0	[7]
<i>Plant-soil partition factor (unitless)</i>							
PCE	Log-normal	0.59	—	4.0	—	—	[7]
Chloroform	Log-normal	2.8	—	4.0	—	—	[7]
<i>Cancer potency factors (mg/kg-day)⁻¹</i>							
PCE							
- oral and dermal	Empirical	5.1×10^{-2}	—	—	—	—	[18]
- inhalation	Empirical	1.8×10^{-3}	—	—	—	—	[18]
Chloroform							
- oral and dermal	Empirical	6.1×10^{-3}	—	—	—	—	[18]
- inhalation	Empirical	8.1×10^{-2}	—	—	—	—	[18]

^a PCE = Tetrachloroethylene.

uniformly distributed between 0.4 and 0.9 [6]. All other factors are the same as described for tapwater ingestion.

Indoor inhalation from tapwater

Tapwater related sources of indoor air contaminants include baths, showers, toilets, dishwashers, cooking, and washing machines. It has been suggested

that contaminant uptake via inhalation of VOCs in tapwater may exceed exposure via direct ingestion [6-15]. In this evaluation, we apply the assumptions of McKone and Bogen [6] and Fisk et al. [16] to estimate indoor air concentrations of tapwater contaminants using the following general equation:

$$C_{\text{air}} = \frac{W_x \times \phi_x \times C_w}{VR_x} \quad (3)$$

where C_{air} is the contaminant concentrations in air (mg/m^3), W_x the water use rate (L/h), ϕ_x the transfer efficiency from water to air (unitless), C_w the contaminant concentration in water (mg/L), and VR_x the air exchange rate (m^3/h).

Following the example of McKone and Bogen [6], the contaminant concentrations in shower air, bathroom air, and household air are estimated using activity-specific estimates of water use rates and air exchange rates and chemical-specific estimates of water to air transfer efficiencies. Based on McKone and Bogen's [6] interpretation of the domestic water use data collected by James and Knuiman [14], the amount of water used during a shower (W_{shower}) is described by a log-normal distribution with a geometric mean of 460 L/h and a geometric standard deviation of 1.4 L/h; total household water use (W_{house}) is represented by a lognormal distribution with a geometric mean of 37 L/h and geometric standard deviation of 1.4 L/h. Air exchange rates in the shower, bathroom, and house (VR_S , VR_B , and VR_H , respectively) are based on the assumption that the volumes of these compartments are 2, 10, and 600 m^3 , respectively, and that the number of air changes per hour ranges uniformly from 2-10 in the shower, 1-10 in the bathroom, and 0.5-2 in the house [6].

The dose associated with inhalation of indoor air is then calculated as follows:

$$Dose = \frac{[(C_S \times ET_S) + (C_B \times ET_B) + (C_H \times ET_H)] \times IR \times EF \times ED}{BW \times AT} \quad (4)$$

where $Dose$ is in mg/kg day, C_S, C_B, C_H are the contaminant concentrations in shower air, bathroom air, and household air, respectively (mg/m^3), ET_S, ET_B, ET_H are the exposure time in the shower, bathroom, and the house respectively (h/day), IR is the inhalation rate (m^3/h), EF the exposure frequency (days/year), ED the exposure duration (years), BW the body weight (kg), and AT the averaging time (days).

The amount of time an individual spends in the bathroom (ET_B) is represented by a log-normal distribution with a geometric mean of 0.27 h/day and a geometric standard deviation of 1.8 h/day; the amount of time an individual spends in the house (ET_H) is taken to be a uniform distribution ranging from 8 to 20 h [6].

The transfer efficiency of tetrachloroethylene from tapwater to shower air has been estimated to be best represented by a triangular distribution with a

range of 0.1–0.9 and mode of 0.6; the transfer efficiency for all other tapwater uses has been estimated to be described by a triangular distribution of 0.1–0.9 and a mode of 0.3 [6]. Transfer efficiencies for chloroform have not been similarly examined. However, given the similar physico-chemical characteristics of the VOCs in general, we assign the triangular distributions developed for tetrachloroethylene to chloroform (Table 3).

Inhalation rates during resting and light, moderate, and heavy activities have been measured in adults [17]. For the purposes of this evaluation, inhalation rates are taken to be uniformly distributed from 0.21–0.74 m³/h. This range is based on the weighted arithmetic means of resting and light activity reported for adult males and females [17], and assumes a 14-h duration (mid-point of ET_H range) during which 8 h are spent sleeping and 6 h are engaged in light activity.

Garden vegetable ingestion

Contaminant uptake via garden vegetable ingestion, which can occur as a result of vegetable irrigation with tapwater, can be described by the following equation:

$$Dose = \frac{C \times SW \times PS \times IR \times FH \times CF \times EF \times ED}{BW \times AT} \quad (5)$$

where *Dose* is in mg/kg day, *C* the chemical concentration in water (mg/L), *SW* the soil–water partition coefficient (L/kg), *PS* the plant–soil partition factor (unitless), *IR* the vegetable ingestion rate (kg/day), *FH* the fraction of ingested vegetables that are homegrown (unitless), *CF* the conversion factor (10⁻⁶ kg/mg), *EF* the exposure frequency (days/year), *ED* the exposure duration (years), *BW* the body weight (kg), *AT* the averaging time (days).

The soil–water partition coefficients for tetrachloroethylene and chloroform can be described as triangular distributions, based on the information presented in Salhotra et al. [7]. Table 3 summarizes the mode, minimum, and maximum values for each distribution. The plant–soil partition factors, which are lognormally distributed, are calculated from data presented by Salhotra et al. [7]. Adult vegetable ingestion rates, log-normally distributed with a geometric mean of 0.062 kg/day and a geometric standard deviation of 1.8 kg/day, are also calculated from presented in the 1991 Salhotra et al. report. Data collected by the U.S. EPA have estimated the fraction of consumed vegetables that are from homegrown sources in rural (47%), city (18%), suburban (29%), and all other areas (34%). For the purposes of this evaluation, these data are bootstrapped into the Monte Carlo analysis.

Cancer potency factors

The inhalation and oral cancer potency factors (*CPF*) are constants taken from U.S. EPA's Health Effects Assessment Summary Table [18]. For the

purposes of this evaluation, the oral potency factor is used to estimate the cancer risk for the absorbed dermal dose. It is important to note that a great deal of uncertainty and conservatism are present in most potency factors, as they represent the upper 95th percent confidence limit (UCL) of the slope of the dose response curve generated by the cancer bioassay. Indeed, for some chemicals the conservatism in the potency factor may "drown out" or overwhelm the uncertainty and conservatism in the estimates of exposure and uptake. For the purposes of simplification, we use the *CPF*s provided by U.S. EPA.

Results and discussion

Using the @Risk™ computer program, the risk estimate equation ($Dose \times CPF = \text{individual increased cancer risk}$) for each pathway was solved for 5,000 iterations. Figure 1 illustrates the distribution of total risk versus probability for tetrachloroethylene. Table 4 contains the 50th and 95th percentile values of risk for each pathway for each chemical. Total chemical-specific risks (all pathways summed for each chemical) are also presented. Total increased risks at the 50th percentile are 2.6×10^{-6} and 5.9×10^{-6} for tetrachloroethylene and chloroform, respectively; at the 95th percentile, increased cancer risks are 9.3×10^{-6} and 2.0×10^{-5} , respectively. As suggested by McKone and Bogen [6], the inhalation pathway "drives" the risk for both chemicals.

These estimated risks are well within the range of 'acceptable' risks typically established for Superfund sites (10^{-4} to 10^{-7}) [2]. Based on these results, it would seem that MCL concentrations of tetrachloroethylene or chloroform in drinking water are unlikely to pose a significant risk to a resident who uses

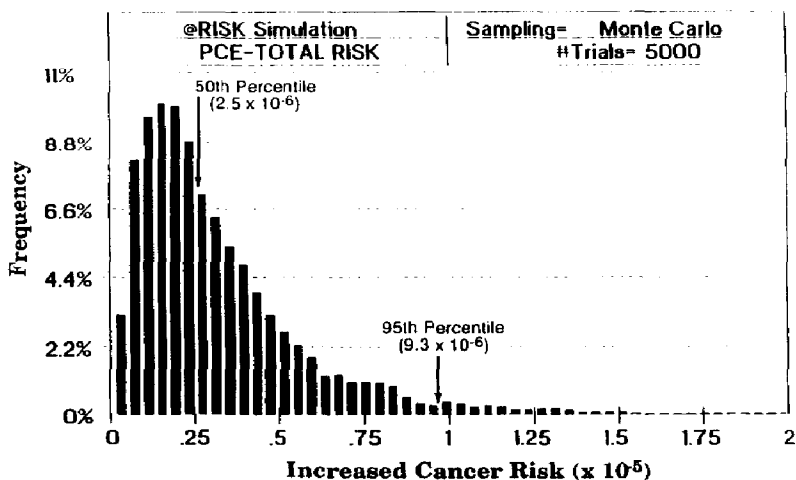


Fig. 1. Distribution of Total PCE risk.

TABLE 4

Estimated individual increased cancer risk

Chemical	Percentile	
	50th	95th
<i>Tetrachloroethylene</i>		
Tapwater ingestion	5.1×10^{-7}	2.8×10^{-6}
Dermal contact	3.4×10^{-7}	2.2×10^{-6}
Inhalation (indoor)	8.5×10^{-7}	6.7×10^{-6}
Vegetable ingestion	4.2×10^{-8}	9.9×10^{-7}
Total risk	2.6×10^{-6}	9.3×10^{-6}
<i>Chloroform</i>		
Tapwater ingestion	1.2×10^{-6}	6.5×10^{-6}
Dermal contact	7.9×10^{-7}	5.7×10^{-6}
Inhalation (indoor)	2.0×10^{-6}	1.6×10^{-5}
Vegetable ingestion	4.5×10^{-8}	1.2×10^{-6}
Total risk	5.9×10^{-6}	2.1×10^{-5}

tapwater as a source of drinking water and for showering, bathing, and garden irrigation. This limited analysis suggests that the "risk-based" remedial goals often implemented at hazardous waste sites might be unnecessarily low and that, at least in the case of tetrachloroethylene and chloroform, MCLs should be considered amply health-protective at most sites. It should be noted that these risk estimates are based on a refinement of exposure assessment *only*; using the probabilistic approach to address pharmacokinetics and cancer potency would likely result in lower estimates of health risk. An important issue in this analysis is how one defines compliance with a clean-up level. For example, is compliance achieved when 95% of the samples are less than the MCL or must each of them be less than that value? Perhaps the best approach to define compliance is to input each sample value into the PDF of water concentrations and routinely check to be sure that the estimated risk remains acceptable.

In this analysis, we specifically address cleanup goals for contaminated groundwater at hazardous waste sites wherein the groundwater may be used as a drinking water source. This implies a finite duration of exposure, as Bureau of the Census studies have shown that most individuals spend far less than a full lifetime in a single residence [11]. For the purposes of evaluating acceptable levels in a public drinking water system, which may be a lifetime drinking water source, it would be inappropriate to assume a less than lifetime exposure.

Case examples such as these are useful for illustrating the conservatism and uncertainty inherent in most risk assessments being performed today. The

Monte Carlo analysis provides the assessor a way to examine the conservatism present in the default point estimate approach and provides a full distribution of risk estimates to risk managers and the public. As stated by Burmaster and Lehr [5]. "We see a way to re-introduce science and fact into risk assessment calculations. The Monte Carlo method gives us a way to distinguish once again risk assessment from risk management." We concur and suggest that the probabilistic approach to exposure and risk assessment be implemented to the fullest extent possible in the coming years. We believe that this refinement will help ensure that our financial resources are properly apportioned to the most pressing environmental and social problems which America faces.

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