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Impact of Surrogate Selection on Risk Assessment for Total Petroleum Hydrocarbons

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ABSTRACT: Environmental contamination involving total petroleum hydrocarbons (TPH) is being investigated and remediated at underground storage tanks, tank farms, pipelines, and refineries across the country. Human health and environmental risk play a significant role in decision making at these sites. However, risk assessment for sites contaminated with petroleum products typically is complicated by inadequate information about the composition of TPH present at the site and the physical and chemical properties and toxicity of the components. To address these data gaps, risk assessors can select surrogate compounds to represent the movement of TPH in the environment at the site and toxicity of TPH present at the site. This article illustrates the potential impact of choice of surrogates on risk estimates, which in turn affect remediation costs.

KEY WORDS: petroleum contamination, total petroleum hydrocarbons, risk assessment, remediation.

I. BACKGROUND

The composition of released petroleum products varies significantly, depending on composition of the source, weathering of the product over time, and differential movement of the components in the environment. For most release sites, detailed information about the composition of TPH will not be available. A previous article (Heath *et al.,* 1993) compiled information about the composition of various petroleum products, the chemical and physical properties of TPH components, and U.S. Environmental Protection Agency-derived (USEPA) values representing the tox-

icity of TPH components. This article applies that information to the process of selecting surrogate compounds to represent TPH and discusses the practical implications of surrogate selection. For simplicity, this article considers only human receptors; however, similar concepts would apply to evaluation of risk to non-human receptors of site-related TPH.

II. SELECTION OF SURROGATES

The first step in selection of surrogate compounds (or combinations of surrogate compounds) is to determine the likely composition of TPH present at the release site. For most release sites, detailed information about the composition of TPH will not be available. Information presented in Tables 1 and 2 of the previous article (Heath *et al.,* 1993) can be used to estimate the initial composition of the TPH that was released, thereby providing a starting point for evaluation of petroleum product releases. The next important step in the selection process is to consider the effects of weathering on the ultimate composition of TPH detected in the environment as a result of the release. Information describing chemical and physical properties of TPH components (Table 3 in Heath *et al.,* 1993) can contribute to an evaluation of the effects of weathering and to a consideration of the impact of fate and transport processes on the composition of TPH both close to and away from the original release point. Surrogate compounds can be selected to predict movement of TPH (or fractions of TPH) in the environment. Toxicity information (Table 4 of Heath *et al.,* 1993) finally can be used to identify one or more surrogate compounds to represent the toxicity of TPH associated with a particular release. When properly integrated, the information provided in the previous article can contribute to selection of surrogate compounds that represent the movement of site-specific TPH in the environment and the toxicity of TPH that reaches human receptors.

The chemical, physical, and toxicologic characteristics of TPH components should be considered in surrogate selection. For instance, highly volatile components of some TPH mixtures can represent a unique exposure pathway (inhalation of volatiles) that can be overlooked if the surrogates used are not volatile. On the other hand, volatiles may disperse rapidly upon release to the environment, so that older (weathered) product may no longer contain significant concentrations of volatiles. Similarly, TPH components that move quickly through soil and with ground water may pose risks through exposure pathways different from those that are relevant for relatively immobile components. Again, weathered product close to the source may not contain significant concentrations of very mobile components.

Perhaps the most common approach to selection of surrogate compounds for TPH is to identify one component for which ample information is avail-

able, and assume that it represents the chemical and physical and toxicologic properties of the TPH at the site. This approach, while common, does not consider the variety of components of TPH or their toxicity or movement in the environment. However, it can be a conservative approach for evaluating risk from TPH; thus it serves as an inexpensive and overly protective screening approach to sites. Another approach would be to use available information about the composition of the type of product that was released to the environment and select a combination of potential surrogates to represent the TPH.

A range of approaches can be used to select surrogate compounds (or combinations of surrogates) to represent TPH in the environment. Because benzene, toluene, ethylbenzene, and xylenes (BTEX) are typically quantified separately from TPH, risks can be quantified for the BTEX components of TPH. In this case, the concentrations of TPH could be reduced by the contribution of BTEX or, more conservatively, the total concentration of TPH can be evaluated. If BTEX are not quantified separately for TPH from gasoline, they should be included in selection of surrogates for TPH.

For this article, surrogate selection considered information about the composition of TPH from different sources, available information describing the toxicity of those components (specifically, verified toxicity values), and available information describing the chemical and physical properties of components. Although significant toxicologic information is available in the literature for a number of the TPH components, many regulatory agencies are reluctant to accept toxicity values derived on the basis of the literature if confirmatory information is not available through USEPA, on the Integrated Risk Information System (IRIS) or in the Health Effects Assessment Summary Tables (HEAST). Therefore, from the perspective of real-world applications for most petroleum release sites, the information provided on IRIS and in HEAST is most pertinent to selection of surrogate compounds for TPH. Because toxicity values are not available for most of the TPH components, toxicity information is the primary constraint on surrogate selection.

For illustrative purposes, six surrogates and surrogate combinations are considered in this paper as a percentage of 100 mg/kg TPH. The surrogates include TPH as 100% *n*-hexane; TPH as 100% benzo(*a*)pyrene; TPH as 100% pyrene; TPH (from gasoline) as 0.00028% benzo(*a*)pyrene, 35% *n*-hexane, and 0.49% pyrene; TPH (from diesel) as 7×10^{-9} benzo(*a*)pyrene and 1.04% pyrene; and TPH (from gasoline with BTEX) as 3.5% benzene, 0.00028% benzo(*a*)pyrene, 35% *n*-hexane, 0.49% pyrene, and 36.6% toluene (Table 1). This range of surrogates incorporates different approaches (i.e., a single surrogate representing the whole range of TPH or surrogate combinations representing a portion of the TPH) and incorporates straight-chain alkanes, monocyclic aromatic hydrocarbons, and PAHs. Relative composition is based on Heath *et al.,* (1993).

TABLE 1
Surrogate Combinations Selected for TPH in Soil

Surrogate combinations	Soil concentration (mg/kg)
Benzo(*a*)pyrene	100
n-Hexane	100
Pyrene	100
Gasoline	
Benzo(*a*)pyrene	0.00028
n-Hexane	35
Pyrene	0.49
Gasoline with BTEX	
Benzene	3.5
Benzo(*a*)pyrene	0.00028
n-Hexane	35
Pyrene	0.49
Toluene	36.5
Diesel	
Benzo(*a*)pyrene	7×10^{-9}
Pyrene	1.04

Note: BTEX, benzene, toluene, ethylbenzene, and xylenes. mg/kg, milligrams per kilogram.

Benzo(*a*)pyrene was selected to represent the carcinogenic PAHs, pyrene to represent noncarcinogenic PAHs, and *n*-hexane to represent alkanes. Selection of benzo(*a*)pyrene is very conservative because, even when appropriate analytical methods are used, it is seldom detected at hydrocarbon sites. Although pyrene is not specifically identified as a component of gasoline, it has the lowest reference dose of the PAHs, making it a conservative surrogate. Benzene and toluene were included in one surrogate combination to represent the BTEX portion of gasoline for sites where BTEX are not analyzed separately.

Although *n*-hexane was selected to represent the toxicologic properties of the straight-chain alkanes, *n*-octane was used to represent their chemical and physical properties. *n*-Pentane and isopentane are the predominant alkanes in gasoline. However, these components are very volatile and are likely lost to the atmosphere on release to the environment, leaving the longer chain alkanes in soil at the site. Because longer chain alkanes are less toxic and move more slowly in the environment than those with shorter chains, *n*-hexane and *n*-octane are conservative surrogates, likely overestimating toxicity and movement in the environment (e.g., leaching). Similarly, using *n*-hexane and *n*-octane as surrogates for alkanes in diesel is conservative because diesel contains longer-chain alkanes (Rumack and Lovejoy, 1991).

III. IMPLICATIONS OF SURROGATE SELECTION

Three scenarios were considered for the purpose of illustrating the importance of surrogate selection: leaching from soil to ground water, exposure of humans excavating in affected soil, and exposure of humans living on affected soil. All three scenarios assume that the concentration of TPH in soil is 100 mg/kg. This value is somewhat arbitrary, but was selected because many states use 100 mg/kg as a screening level for TPH in soil (Millner *et al.*, 1992).

A. Leaching

The potential for affected soil to leach site-related constituents into ground water (potentially causing constituent levels in ground water to exceed ground-water standards or to pose a risk to users) was evaluated using the USEPA's Organic Leachate Model (OLM) (Federal Register, 1986).

When a constituent is released to soil, a certain amount will be adsorbed by the soil; some will dissolve in soil pore water; and some will volatilize. These are equilibrium processes that are affected by factors such as the physical and chemical properties of the constituent, the quantity released, soil characteristics, depth of contamination, temperature, etc. The OLM estimates the leaching behavior of organic constituents using a concentration and solubility-based logarithmic regression equation. The OLM equation is presented below:

$$C_L = 0.00221\ C_s^{0.678} S^{0.373}$$

where C_L, predicted constituent concentration in the leachate (mg/l); C_s, constituent concentration in soil (mg/kg); S, constituent's water solubility at ambient temperature (25°C) (mg/l) from Table 2.

The OLM is a simplistic and conservative method for evaluating the leaching potential of organic compounds. It was chosen for use here because it illustrates the importance of a compound's physical and chemical characteristics (in this case solubility) to affect leaching and mobility in soil.

The predicted TPH concentrations in the leachate are presented in Table 2. The predicted leachate concentrations for the surrogates assuming a residual soil concentration of 100 mg/kg were 0.006 mg/l, 0.03 mg/l, and 0.04 mg/l for benzo(*a*)pyrene, pyrene, and *n*-hexane, respectively. For the three surrogates chosen to represent TPH components associated with gasoline, the predicted leachate concentrations ranged from 0.000001 mg/l for benzo(*a*)pyrene to 0.021 mg/l for *n*-hexane. The predicted leachate concentrations for the surrogates chosen to represent TPH associated with gasoline

TABLE 2
Predicted Leachate Concentrations of Surrogate Combinations

Surrogate combinations	Water solubility (mg/l)	C_s (mg/kg)	Predicted leachate concentration (mg/l)
Benzo(*a*)pyrene	0.004	100	0.004[a]
n-Hexane	0.66	100	0.04
Pyrene	0.171	100	0.03
Gasoline			
Benzo(*a*)pyrene	0.004	0.00028	0.000001
n-Hexane	0.66	35	0.021
Pyrene	0.171	0.49	0.0007
Gasoline with BTEX[b]			
Benzene	1780	3.5	0.08
Benzo(*a*)pyrene	0.004	0.00028	0.000001
n-Hexane	0.66	35	0.021
Pyrene	0.171	0.49	0.0007
Toluene	627	36.5	0.3
Diesel			
Benzo(*a*)pyrene	0.004	7×10^{-9}	8×10^{-9}
Pyrene	0.171	1.04	0.0012

Note: BTEX, benzene, toluene, ethylbenzene, xylene; C_s, concentration in soil; mg/l, milligrams per liter; mg/kg, milligrams per kilogram.

[a] OLM predicts a leachate concentration (0.006 mg/l) higher than the water solubility (0.004 mg/l).

[b] Assumes no analytical data specific to BTEX.

and BTEX components ranged from 0.000001 mg/l for benzo(a)pyrene to 0.3 mg/l for toluene.

The lowest leachate concentrations were predicted for the two compounds chosen to represent TPH components associated with diesel (benzo[*a*]pyrene and pyrene). Leachate concentrations were 0.0012 mg/l for pyrene and 8×10^{-10} mg/l for benzo(*a*)pyrene as a result of the low water solubility and assumed low soil concentrations for these two PAHs.

Using a more complex model tailored to a particular site would result in similar insights, although perhaps somewhat different specific results because additional constituent-specific and site-specific factors would be considered.

For many sites that have been impacted by releases of TPH, future (re)development of the site is likely, potentially resulting in human exposure to affected soil. Two plausible exposure scenarios, construction/excavation activities and residential use, were chosen to illustrate the importance of surrogate selection when evaluating risk associated with TPH contamination.

B. **Excavation**

Future construction at a site could result in exposure (via incidental ingestion, dermal contact, and inhalation) to residual concentrations of site-related TPH present in surface or near subsurface soil. Excavations for building foundations, basements, sewers, or utilities would, for the most part, not be expected to extend beyond 10 ft below land surface. Daily soil exposure doses were calculated using standardized equations and exposure assumptions that are consistent with USEPA risk assessment guidance for Superfund sites. Site-specific information should be incorporated into this equation when available. The equation and assumptions used to calculate the soil exposure doses for an excavation worker are shown in Table 3.

The exposure doses and risk estimates associated with exposure of a hypothetical excavation worker to soil containing TPH are presented in Table 4. Of the three surrogates selected to represent 100% TPH (benzo[*a*]pyrene, *n*-hexane, and pyrene), benzo(*a*)pyrene is the only carcinogen. The excess lifetime cancer risk calculated for exposure of an excavation worker to soil containing 100 mg/kg benzo(*a*)pyrene is 2×10^{-5} (Table 4). The hazard quotients (a measure of potential noncarcinogenic effects) were 0.002 for TPH as *n*-hexane and 0.003 for pyrene.

For an excavation worker exposed to soil containing the TPH combination selected to represent gasoline (without BTEX), the excess lifetime cancer risk is 4×10^{-11} (due solely to benzo[*a*]pyrene) and the hazard index is 0.0006 (rounded to one significant figure) (Table 4). For the TPH combination selected to represent gasoline with BTEX, the excess lifetime cancer risk was 9×10^{-8} and the hazard index was 0.01 (Table 4). The excess lifetime cancer risk and hazard index for an excavation worker exposed to soil containing TPH (from diesel) were 1×10^{-15} and 0.00004 (Table 4).

C. **Residential Use**

Alteration of a site for development of private residences also was evaluated. The scenario used here assumes that adult residents are exposed to soil containing the TPH surrogates and surrogate combinations described previously. (This scenario is illustrative, but child residents can also be considered.) Daily exposure doses were calculated using standardized equations and exposure assumptions that are consistent with USEPA risk assessment guidance for Superfund sites. (Again, site-specific factors may affect choice of equations or parameters.) The equation and assumptions used to calculate the soil exposure doses for an adult resident are shown in Table 3.

The exposure doses and risk estimates for a hypothetical adult residents exposed to soil containing TPH are shown in Table 5. The risk estimates (both excess

TABLE 3
Equations and Sample Calculations for Soil Exposure

Equation definitions:

$$SExD_o = \frac{C_s \times IR \times EF \times ED \times UC_1}{BW \times AP}$$

$$SExD_i = \frac{C_s \times SPM \times FIP \times BR \times ET \times EF \times ED \times UC_1}{BW \times AP} \text{ (particulates)}$$

or

$$\frac{C_s \times BR \times ET \times H \times PGV \times UC_2 \times UC_3 \times EF \times ED}{BW \times Kd \times W \times AP \times UC_4} \text{ (vapors)}$$

$$SExD_d = \frac{C_S \times SSA \times SAR \times ABS \times EF \times ED \times UC_1}{BW \times AP}$$

$$HQ = (SExD_o / RfD_o) + (SExD_i / RfD_i) + (SExD_d / RfD_S)$$

$$ELCR = (SExD_o \times CSF_o) + (SExD_i \times CSF_i) + (SExD_d \times CSF_a)$$

where ABS, dermal absorption efficiency, constituent-specific; AP, averaging period (25,550 d for cancer effects, and ED × 365 d for non-cancer effects) (USEPA, 1991, USEPA, 1989a); BR, breathing rate (2.5 m^3/h for an excavation worker; 0.83 m^3/h for a resident) (USEPA, 1991b); BW, body weight (70 kg) (USEPA, 1991); C_s, constituent concentration in the soil (mg/kg); CSF_a, cancer slope factor for dermal exposure, adjusted for absorbed dose $(mg/kg\text{-}day)^{-1}$; CSF_i, cancer slope factor for inhalation exposure $(mg/kg\text{-}day)^{-1}$; CSF_o, cancer slope factor for oral exposure $(mg/kg\text{-}day)^{-1}$; ED, exposure duration (6 days/week for an excavation worker; 30 years for a resident) (USEPA, 1991); EF, exposure frequency (13 weeks for an excavation worker; 350 days/year for a resident) (USEPA, 1991); ELCR, excess lifetime cancer risk (unitless); ET, exposure time (8 h/d for an excavation worker; 24 h/d for a resident) (USEPA, 1991); FIP, fraction inhaled particulates (0.125) (Hwang and Falco, 1986); H, Henry's Law Constant (atm-m^3/mol; constituent specific); HQ, hazard quotient (unitless); IR, incidental ingestion rate for soil (480 mg/d for an excavation worker; 100 mg/d for a resident) (USEPA, 1989b); Kd, soil-water partition coefficient (cm^3/g or ml/g) (constituent-specific); PGV, pore gas velocity (1.63×10^{-5} m/sec) (Hwang and Falco, 1986); RfD_a, reference dose adjusted to an absorbed dose (mg/kg-day); RfD_i, reference dose for inhalation exposure (mg/kg-day); RfD_o, reference dose for oral exposure (mg/kg-day); SAR, soil adherence rate (1 mg/cm^2-day) (USEPA, 1992b); $SExD_d$, soil exposure dose from dermal contact (mg/kg-day); $SExD_i$, soil exposure dose from inhalation of particulates or vapors from soil (mg/kg-day); $SExD_o$, soil exposure dose from incidental ingestion (mg/kg-day); SPM, suspended particulate matter (0.075 mg/m^3) (*Federal Register,* 1988); SSA, exposed skin surface area (2940 cm^2) (USEPA, 1989b); UC_1, unit conversion (10^{-6} kg/mg); UC_2, unit conversion 2 (41 mol/atm-m^3) (Hwang and Falco, 1986); UC_3, unit conversion 3 (10^6 cm^3/m^3); UC_4, unit conversion 4 (10^3 g/kg); W, wind speed, assumed (4 m/sec).

TABLE 4
Soil Exposure Risks for a Hypothetical Excavation Worker

Surrogate combinations	C_s (mg/kg)	Excess lifetime cancer risk	Hazard quotient
Benzo(*a*)pyrene	100	2×10^{-5}	—
n-Hexane	100	—	0.002
Pyrene	100	—	0.003
Gasoline			
Benzo(*a*)pyrene	0.00028	4×10^{-11}	—
n-Hexane	35	—	0.00056
Pyrene	0.49	—	0.000016
		ELCR = 4×10^{-11}	HI = 0.0006
Gasoline with BTEX[a]			
Benzene	3.5	8.7×10^{-8}	—
Benzo(*a*)pyrene	0.00028	4.3×10^{-11}	—
n-Hexane	35	—	0.00056
Pyrene	0.49	—	0.000016
Toluene	36.5	—	0.012
		ELCR = 9×10^{-8}	HI = 0.01
Diesel			
Benzo(*a*)pyrene	7×10^{-9}	1×10^{-15}	—
Pyrene	1.04	—	0.000035
		ELCR = 1×10^{-15}	HI = 0.00004

Note: BTEX, benzene, toluene, ethylbenzene, and xylenes; C_s, concentration in soil; ELCR, excess lifetime cancer risk; HI, hazard index (sum of the HQs); HQ, hazard quotient; mg/kg, milligrams per kilogram.

[a] Assumes no analytical data specific to BTEX.

lifetime cancer risks and hazard indices) for a resident are higher than those for an excavation worker. This is due primarily to the assumed greater exposure frequency and duration (see assumptions in Table 3). The excess lifetime cancer risk for an adult resident exposed to soil containing TPH as 100% benzo(*a*)pyrene were 4×10^{-4}. The hazard indices for TPH as *n*-hexane and TPH as pyrene were 0.009 and 0.02, respectively (Table 5).

Residential exposure to soil containing the TPH combination selected to represent gasoline (without BTEX) results in an excess lifetime cancer risk of 1×10^{-9} (due to benzo[*a*]pyrene) and a hazard index of 0.003 (Table 5). For the surrogate combination chosen to represent TPH as gasoline with BTEX, the cumulative excess lifetime cancer risk was 1×10^{-5} (attributable to benzene and benzo[*a*]pyrene), and the hazard index was 0.07 (Table 5). The excess lifetime cancer risk and hazard index for an adult resident exposed to soil containing TPH (from diesel) were 3×10^{-14} and 0.0002, respectively (Table 5).

TABLE 5
Soil Exposure Risks for a Hypothetical Resident

Surrogate combinations	C_s (mg/kg)	Excess lifetime cancer risk	Hazard quotient
Benzo(*a*)pyrene	100	4×10^{-4}	—
n-Hexane	100	—	0.009
Pyrene	100	—	0.02
Gasoline			
Benzo(*a*)pyrene	0.00028	1.2×10^{-9}	—
n-Hexane	35	—	0.0031
Pyrene	0.49	—	0.000095
		ELCR = 1×10^{-9}	HI = 0.003
Gasoline with BTEX[a]			
Benzene	3.5	1.1×10^{-5}	—
Benzo(*a*)pyrene	0.00028	1.2×10^{-9}	—
n-Hexane	35	—	0.0031
Pyrene	0.49	—	0.000095
Toluene	36.5	—	0.07
		ELCR = 1×10^{-5}	HI = 0.07
Diesel			
Benzo(*a*)pyrene	7×10^{-9}	3×10^{-14}	—
Pyrene	1.04	—	0.0002
		ELCR = 3×10^{-14}	HI = 0.0002

Note: BTEX, benzene, toluene, ethylbenzene, and xylenes; C_s, concentration in soil; ELCR, excess lifetime cancer risk; HI, hazard index (sum of the HQs); HQ, hazard quotient; mg/kg, milligrams per kilogram.

[a] Assumes no analytical data specific to BTEX.

IV. **DISCUSSION**

Selection of surrogates to represent TPH released to the environment must consider a variety of issues. These include the composition of the TPH, often represented by the composition of the product that was released. This is an oversimplification, because weathering of TPH in the environment results in changes in TPH composition following release. Other issues include the toxicologic and chemical/physical properties of TPH components. Surrogates should represent the range of properties possessed by components present in the TPH; however, because information describing toxicity is available for only a small number of TPH components, it is difficult to ensure that surrogates represent the range of toxic effects, so conservative simplifications are often made. Most importantly, selection of surrogates must be site-specific.

As indicated in the previous section, different surrogates used in the same exposure and risk equations can result in very different conclusions. For in-

stance, predicted leachate concentrations of TPH ranged over two orders of magnitude, from 0.006 mg/l to 0.3 mg/l, depending on the surrogate used. Hazard indices also ranged over orders of magnitude, from 0.00004 to 0.01 for the excavation worker and 0.0002 to 0.07 for residents. Estimates of excess lifetime cancer risk ranged over several orders of magnitude, from 10^{-15} to 10^{-5} for excavation workers and from 10^{-14} to 10^{-4} for residents. Although not the intent of this article, these results demonstrate that the arbitrary soil criterion of 100 ppm TPH used in several states is excessively conservative, resulting in remediation of sites that do not present a significant risk.

Thus, it is clear that surrogate choice can significantly impact risk estimates. This is particularly important because these risk estimates are used to guide decisions about remediation activities that can range from thousands to millions of dollars, depending on the complexity of the site and the level of clean-up. Although conservative, simplified assumptions about surrogates for TPH can provide significant insight and contribute to screening of sites; more thoughtful, site-specific selection of surrogates can be a critical step in the remediation decision-making process.

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