APPENDIX C

INHALATION EXPOSURE TO TAP WATER THROUGH SHOWERING LITERATURE AND MODEL REVIEW

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In response to public comments, the following articles containing shower inhalation exposure models have been evaluated for possible inclusion of these pathways in the groundwater risk analysis for the petroleum waste listing:

- Andelman, Julian B. 1990. Total Exposure to volatile organic compounds in potable water, Chapter 20 in *Significance and Treatment of Volatile Organic Compounds in Water Supplies*,. N.M. Ram, R.F. Christman, and K.P. Cantor (eds.). Lewis Publishers Chelsea MI. pp. 485-504.
- McKone, Thomas E. 1987. Human exposure to volatile organic compounds in household tap water: The Indoor Inhalation Pathway. *Environmental Science and Technology* 21:1194-1201.
- McKone, Thomas E., and J.P. Knezovich. 1991. The transfer of trichloroethylene (TCE) from a shower to indoor air: Experimental measurements and their implications. *Journal of Air and Waste Management Association.* 41(3):282-286.
- Little, John C. 1992a. Applying the two resistance theory to contaminant volatilization in showers. *Environmental Science and Technology* 26:1341-1349.
- Little, John C. 1992b. Correspondence. Comment on "Human exposure to volatile organic compounds in household tap water: the indoor inhalation pathway." *Environmental Science and Technology* 26(4):836-837.
- McKone, Thomas E., and K.T. Bogen, 1992. Uncertainties in health-risk assessment: an integrated case study based on tetrachloroethylene in California groundwater. *Regulatory Toxicology and Pharmacology* 15:86-103.

Andelman (1990) is the simplest model; it assumes that all volatile constituents (i.e., constituents with a Henry's law constant of 2E-6 atm-m^3/mol or greater) are equally volatilized and that below a threshold Henry's law constant of 2E-6 atm-m^3/mol no volatilization occurs. In the case of very volatile compounds, this approach may be adequate, but it will tend to over-estimate exposure if semivolatile constituents are included in risk assessment. McKone and Bogen (1992) offered a simple screening model to estimate exposure in the shower based on compound-specific data, Henry's law constant, and air and water diffusivities, but it does not consider the time-dependent variations of the exposure concentrations (uses "steady-state" concentration analysis). Both Little (1992a) and McKone (1987) consider the variation in shower concentration with time based on both the volumes of the different model compartments (house, bathroom, shower) and the air exchange rates between the compartments.

The simple exposure models presented by Andelman (1990) and McKone and Bogen (1992) can be used to calculate the shower air exposure concentrations directly. The more detailed shower models presented in Little (1992a) and McKone (1987) require more complicated solution algorithms to calculate the contaminant concentration over time. These concentrations are then averaged over the exposure duration to calculate the average exposure concentrations to be used in the risk analysis. The shower models of Little (1992a) and McKone (1987) use basically the same model construct, except that Little (1992b) suggests a slight modification to the equation proposed by McKone (1987) to estimate the overall mass transfer coefficient. Due to the nature of the variations in the models evaluated, the model evaluation is divided into three parts. First we evaluate and compare the predicted overall mass transfer coefficients by McKone (1987) and Little (1992b). Second we compare the impact of the predicted overall mass transfer coefficient on the estimated emissions when applied with a detailed shower model. Third we compare the predicted exposure concentrations using Andelman (1990), McKone and Bogen (1992), and the detailed shower model based on Little (1992a) and McKone (1987). These comparisons provide support for the selection of a model to be used in the risk analysis.

C.1.1 Estimation and Comparison of Overall Mass Transfer Coefficients

The basic equation presented in McKone (1987) for estimating the overall mass transfer coefficient from tap water to air is

$$K_{ol} = \beta \, [2.5/(D_{ab,l}^{2/3}) + 1/(D_{ab,g}^{2/3} \, H')]^{-1} \qquad (6\text{-}1)$$

where,

K_{ol} = overall mass transfer coefficient (cm/s)
β = proportionality constant $(\text{cm-s})^{-1/3}$.
$D_{ab,l}$ = diffusivity of constituent in liquid phase (cm^2/s)
$D_{ab,g}$ = diffusivity of constituent in gas phase (cm^2/s)
H' = $H/RT = 41\,H$ = dimensionless Henry's law constant
H = Henry's law constant (atm-m^3/mol)
R = Ideal gas law constant = 8.205E-5 atm-m^3/mol-K
T = temperature (K).

In a correspondence to the editor, Little (1992b) suggested that another proportionality constant should be included in the equation presented in McKone (1987). The suggested change in the mass transfer equation is as follows:

$$K_{ol} = \beta \, [2.5/(D_{ab,l}^{2/3}) + \alpha/(D_{ab,g}^{2/3} \, H')]^{-1} \qquad (6\text{-}2)$$

where

α = proportionality constant (dimensionless).

Based on his compilation and analysis of experimental shower emission data, in which he developed a means to estimate the gas and liquid phase mass transfer coefficients from experimentally observed overall mass transfer coefficients for compounds with differing Henry's law constants, Little (1992a) suggests $\alpha \approx 70$ in Equation 6-2 (Little, 1992b). Consequently, the main difference in Equation 6-1 (McKone, 1987) and Equation 6-2 (Little, 1992b) is the suggested value of α. McKone (1987) uses $\alpha = 1$; Little (1992b) suggests $\alpha \approx 70$.

The change in the shower water concentration as the water falls, which equals the contaminant emission rate, is calculated using the overall mass transfer coefficient as follows:

$$\delta c / \delta t = - K_{ol} \, (A/V)(c - y_s/H') \qquad (6\text{-}3)$$

where

c = liquid phase (droplet) constituent concentration ($\mu\text{g/cm}^3$ or mg/L)
t = time (s)

A = total surface area for mass transfer (cm^2)
V = total volume of water within the shower compartment (cm^3)
y_s = gas phase constituent concentration in the shower ($\mu g/cm^3$ or mg/L)
H' = dimensionless Henry's law constant.

Consequently, in addition to the overall mass transfer coefficient, the emission rate of a contaminant within the shower is dependent on the surface-area-to-volume ratio of the shower water (within the shower) and the concentration driving force between the water and the shower air.

Instead of estimating the surface area for mass transfer, Little (1992a) lumps the overall mass transfer coefficient and the total surface area for mass transfer (i.e., $K_{ol}A$). McKone (1987) defined transfer efficiencies and used radon emission data to calibrate the model. This treatment eliminates the need to calculate the total surface area for mass transfer, but the approach ideally includes a calibration run for a similar shower nozzle type and flow rate. In effect, the transfer efficiency approach lumps the total surface area into the proportionality constant of Equation 6-1 as follows:

$$K_{ol}A= \beta' \; [2.5/(D_{ab,l}^{2/3}) + 1/(D_{ab,g}^{2/3} \, H')]^{-1} \qquad \text{(Eqn. 6-6)}$$

where

β' = lumped proportionality constant $(cm^5/s)^{1/3}$.

The shower emissions can be modeled based on falling droplets as a means for estimating the surface-area-to-volume ratio for mass transfer and the residence time of the water in the shower compartment. Equation 6-3 can then be integrated assuming the compound concentration in the gas phase is constant over the time frame of the droplet fall (Little, 1992a, similarly assumes that the gas phase is constant over the residence time of the water in the shower). The time required for a droplet to fall equals the nozzle height divided by the water droplet velocity. The ratio of the surface-area-to-volume for the droplet is calculated as $6/d_p$ (i.e., by assuming a spherical shape). By assuming the drops fall at terminal velocity, the surface-area-to-volume ratio and the residence time can be determined based solely on droplet size. A droplet size of approximately 1 mm (0.1 cm) was selected by calibrating the model for benzene emissions. The terminal velocity for the selected droplet size is approximately 400 cm/s.

The fraction of constituent emitted from a water droplet at any given time can then be calculated by integrating Equation 6-3 and rearranging as follows:

$$f_{em} = 1 - c_{out}/c_{in} = (1 - f_{sat})(1 - e^{-N}) \qquad (6\text{-}4)$$

where

f_{em} = fraction of constituent emitted from the droplet (dimensionless)
c_{out} = droplet constituent concentration at shower floor/drain (mg/L)
c_{in} = droplet constituent concentration entering the shower (mg/L)
f_{sat} = $y_s/(H' c_{in})$ = fraction of gas phase saturation (dimensionless)
N = dimensionless overall mass transfer coefficient = $K_{ol}\,(6/d_p)\,(h/v_t)$
d_p = droplet diameter = 0.1 (cm)
v_t = terminal velocity of droplet = 400 (cm/sec)
h = nozzle height (cm).

Equation 6-4 is equivalent to the equation presented in Little (1992a, Equation 5) for liquid phase concentration change, except that Little uses the total surface area for mass transfer (lumped with K_{ol}) and the shower flow rate to dimensionalize the overall mass transfer. Equating the two approaches, the total surface area for mass transfer can be as follows:

$$A = Q_l\,(6/d_p)\,(h/v_t) \qquad (6\text{-}5)$$

where

A = total surface area for mass transfer (cm^2)
Q_l = volumetric shower water flow rate(cm^3/s).
d_p = droplet diameter = 0.1 (cm)
v_t = terminal velocity of droplet = 400 (cm/s)
h = nozzle height (cm)

The experimentally observed overall mass transfer coefficients (i.e., $K_{ol}A$), as compiled by Little (1992a), are summarized in Table C.1. Most of these experimental overall mass transfer coefficients are based on measurements of the constituent loss from the water, although some are based on measured air concentration data. Overall mass transfer coefficients were also calculated using Equation 6-6 ("McKone (1987) model"), Equation 6-1 combined with Equation 6-5 ("modified McKone, 1987 model"), and Equation 6-2 combined with Equation 6-5 ("Little model"). The value of the proportionality constants was estimated by calibrating the models to predict identical overall mass transfer coefficients for benzene for the model shower (see Section C.3). For the McKone model (1987), $\beta' = 5.2E+5\ (cm^5/s)^{1/3}$ was calculated; for the modified McKone model (1987): $\beta = 216\ (cm\text{-}sec)^{-1/3}$; and for the Little model: $\beta = 276\ (cm\text{-}sec)^{-1/3}$ and $\alpha = 70$. Table C.1 also compares the calculated overall mass transfer coefficients with the experimental overall mass transfer coefficients as compiled by Little (1992a). Note that most of the contaminants for which shower data are available are highly volatile. For the two compounds with the lowest volatility (1,2,3-trichloropropane and 1,2-dibromo-3-chloropropane), the Little model shows a sharp contrast to the modified McKone (1987) model, but it significantly underestimates the overall mass transfer coefficient for 1,2-dibromo-3-chloropropane. For the most part, the modified McKone (1987) model provides better estimates of the overall mass transfer coefficients for the different shower studies (i.e., shower conditions) than the McKone (1987) model.

Table C.1. Comparison of Overall Mass Transfer Coefficients

Referenced Study (Q_l; h)[a] Compound	Reported $K_{ol}A$[a] (L/min)	Calc'd $K_{ol}A$ McKone[b] (L/min)	Calc'd $K_{ol}A$ modified McKone[c] (L/min)	Calc'd $K_{ol}A$ Little[d] (L/min)
Study T (13.5 L/min; 1.7 m)[a]				
carbon tetrachloride	11 - 21	5.3	13	16
tetrachloroethene	7 - 16	5.0	12	14
1,1,1-trichloroethane	9 - 15	5.3	13	15
chloroform	8 - 15	5.7	14	13
1,2,3-trichloropropane	2 - 6	4.6	11	3
Study G (5 L/min; 1.8 m)[a]				
trichloroethene	9	5.4	5.2	5.7
chloroform	4 - 7	5.7	5.5	5.1
1,2-dibromo-3-chloropropane	2	3.4	3.3	0.24
Study H (13.7 L/min; 1.6 m)[a]				
trichlorofluoromethane	25 -34	5.6	13	17
1,1,1-trichloroethane	21 - 29	5.3	12	14
tetrachloroethene	26	5.0	12	14
trichloroethene	30	5.4	13	14
Study M (9.5 L/min; 1.6 m)[a]				
trichloroethene	8½ - 9.7	5.4	8.7	9.7
Study J (8.7 L/min; 1.7 m)[a]				
chloroform	2.5	5.7	9.5	8.8

[a] Data and referenced study abbreviations as reported in Little (1992a).

[b] Calculated assuming $\beta' = 5.2E+5\ (cm^5/sec)^{1/3}$ using the Equation 6-6 (McKone, 1987).

[c] Calculated assuming $\beta = 216$ using the Equation 6-1 and Equation 6-5 (modified McKone, 1987).

[d] Calculated assuming $\beta = 276$ and $\alpha = 70$ using the Equation 6-2 and Equation 6-5 (Little, 1992b).

C.1.2 Comparison of Predicted Shower Emissions for Detailed Shower Model

The question arises: How sensitive are the model estimates to the estimated overall mass transfer coefficient? An evaluation of Equation 6-4 reveals that, if the dimensionless overall mass transfer coefficient is large ($N>3$), the fraction emitted during the droplet fall becomes independent of the overall mass transfer coefficient because the term $(1 - e^{-N})$ approaches 1. That is, the McKone and Little models may predict different overall mass transfer coefficients, but both models may predict the same amount of contaminant being emitted from the water droplet because the term $(1 - e^{-N})$ approaches 1 for both models. Therefore, to assess the actual variation of the predicted emissions for the different shower models, we evaluated the total mass of contaminant emitted during a shower event. Although Andelman (1990) is referenced in Risk*Assistant as applying a set fraction emitted from the shower, Andelman (1990) suggests that the set emission fraction (or transfer efficiency) will become limited as the shower air concentration builds up. Therefore, Equation 6-4 is applied with $N=1$ (transfer efficiency of 63.2 percent) for Henry's law constants greater than or equal to 2.4E-6 atm-m^3/mol ($H' \geq 10^{-4}$) and $N=0$ for Henry's law constants greater than 2.4E-6 atm-m^3/mol. The evaluation in this section offers a comparison of a simple model for estimating the overall mass transfer coefficient (Andelman, 1990, as just described) with the constituent-specific overall mass transfer equations presented in Section C.1.1 using the same detailed shower model algorithm. The detailed shower model estimates the change in the shower air concentration based on the mass of constituent lost by the water (fraction emitted or emission rate) and the air exchange rate between the various model compartments (shower, the rest of the bathroom, and the rest of the house) following the same basic model construct described by Little (1992a). The resulting differential equations were solved using finite difference numerical integration. The pertinent equations are summarized in Appendix A.

The McKone, Little, and Andelman models were compared for constituents exhibiting a wide range of Henry's law constants. Ten compounds were selected for the analysis, representing the 5th, 15th, 25th,..., and 95th percentile of Henry's law constants for the compounds in the exposure model database. The total mass of compounds emitted over a standard shower event, as described in Section C.2 (Table C.5), is presented in Table C.2. [Note: Because the McKone and modified McKone models were calibrated using the same model shower parameters as this simulation, both the McKone and modified McKone models provide identical emission predictions for this model simulation.]

In comparing the model predictions presented in Table C.2, the simple Andelman (1990) model, as constrained by the saturation level in the shower using the detailed shower model, provided emission results very similar to the McKone (1987) model. Apparently, both the Andelman and McKone models become "saturation rate" limited at approximately the same Henry's law value for this model simulation. The Little model predicts much lower emissions for compounds in the semivolatile range than the other models, because the Little model predicts that the emissions are mass-transfer-rate-limited for these compounds rather than saturation-rate-limited for this model simulation. It is important to note that the comparison of model emission predictions provided in Table C.2 is somewhat dependent on the shower model parameters selected for the simulation. If shower water parameters that increase the time required to saturate the shower were used (i.e., decreased shower water use rates, increased shower volumes, and increased shower air exchange rates), larger differences would be expected in the emissions predicted by the Andelman and McKone models. If shower water parameters that would decrease the time required to saturate the shower were used (i.e., increased shower water use rates, decreased shower volumes, and decreased shower air exchange rates), the emissions predicted by the Little model would more closely compare to the emissions predicted by the Andelman and McKone models.

Unfortunately, emission measurement data on which to base a model recommendation are very limited for compounds in the semivolatile range. The experimental overall mass transfer rate coefficient data presented in Table C.1 does not provide conclusive evidence as to which model best simulates semivolatile compounds (i.e., the Little model provided excellent prediction of K_{ol} for 1,2,3-trichloropropane, but it significantly underestimated the K_{ol} for 1,2- dibromo-3-chloropropane). From the comparison provided in Table C.2, it appears that a very simple mass transfer rate model of Andelman (1990) provides the highest emission estimates; the McKone (1987) model provides similarly conservative emission estimates for the shower model parameters used in this simulation; and the Little model predicts lower shower emissions (or exposure levels) for semivolatile compounds for the model shower parameters used in this simulation.

Table C.2. Model Predictions for Total Mass Emitted During Model Shower[a]

Compound	Total Mass Emitted During Model Shower (mg)		
	Andelman[b]	McKone[c]	Little[d]
bisphenol A	0	0.00	0.00
phthalic anhydride	0	0.00	0.00
2,4-dinitrophenol	0	0.06	0.01
quinoline	0.71	0.35	0.08
heptachlor epoxide	1.3	1.1	0.11
diallate	6.0	5.8	1.7
1,2-dibromo-3-chloropropane	15	13	2.3
1,2-dichloroethane	31	31	21
1,1-dichloroethane	36	37	36
ethyl ether	36	36	40

[a] All calculations performed using the complete shower model presented in Appendix A and using model shower parameters provided in Table 6.5 , but with non-shower water use rates set to zero and C_{water} = 1 mg/L.
[b] Calculated assuming N = 1 for $H' \geq 10^{-4}$ and N = 1 for $H' < 10^{-4}$ (Andelman, 1990).
[c] Calculated assuming $\beta = 216$ using Equation 6-1 and Equation 6-5 (modified McKone, 1987).
[d] Calculated assuming $\beta = 276$ and $\alpha = 70$ using Equation 6-2 and Equation 6-5 (Little, 1992b).

C.1.3 Comparison of Predicted Average Exposure Concentrations

The detailed shower model equations provided in Appendix A were compared to less complicated models that directly estimate the average air exposure concentrations. Again the model comparison was conducted using the 10 compounds selected to represent the range of Henry's law constants for the compounds in the exposure model database. The exposure concentrations predicted by the equations presented in Andelman (1990) and in McKone and Bogen (1992) were compared with the detailed shower model using the McKone (1987) mass transfer correlation as applied using the equations in Appendix A. Table C.3 compares the results of the model predictions of the average shower air exposure concentrations.

During the evaluation, it was evident that the simple models - Andelman (1990) and McKone and Bogen (1992) - predicted shower air exposure concentrations greater than saturation (i.e., greater than the equilibrium air concentration based on Henry's law partitioning) for compounds of low volatility. The equilibrium air concentrations based on Henry's law partitioning are also provided in Table C.3 for comparison with the predicted exposure concentrations. The results presented in Table C.3 illustrate the inadequacy of the Andelman (1990) exposure concentration equations for anything but highly volatile compounds. The McKone and Bogen (1992) exposure model provides constituent-dependent exposure variations, but does not account for shower volume (additional dilution air) or the time it takes to reach the maximum concentration. As discussed in Section C.1.2, the detailed shower model using McKone (1987) mass transfer dependency predicts near-saturation exposure concentrations for the mid to low volatility compounds. Thus, it appears that the McKone (1987) model provides a conservative estimate of the shower exposure.

The full shower model described in Appendix A also provides direct estimates of the bathroom and whole house exposure. McKone and Bogen (1992) also provide simple equations to estimate bathroom exposure based on shower emissions and whole house exposure based on other water use rates. Table C.4 compares the McKone and Bogen (1992) bathroom air exposure concentration estimates to the bathroom air exposure concentrations estimated by the detailed shower model described in Appendix A for 45 minutes immediately following the shower event. Again, the McKone and Bogen (1992) model provides very high exposure estimates because of its lack of dependence on the bathroom volume (additional dilution air) or the time required to build up the concentration.

The risk from inhalation exposures in the remainder of the house was several orders of magnitude less than the risk from inhalation exposures in the bathroom and during showering (Coburn, 1996). Thus, it appears that the whole house compartment of the analysis may be disregarded without significant impacts on the inhalation exposure risk analysis.

Table C.3. Model Predictions For Average Shower Air Exposure Concentration[a]

Compound	Average Shower Air Concentration (mg/L)			
	Andelman[b]	McKone and Bogen[c]	McKone[d]	Saturated Air[e]
Bisphenol A	0	6.0E-8	3.7E-9	4.1E-9
Phthalic anhydride	0	1.9E-5	6.3E-7	6.7E-7
2,4-Dinitrophenol	0	2.7E-4	1.6E-5	1.8E-5
Quinoline	7.9E-3	2.4E-4	1.0E-4	1.1E-4
Heptachlor epoxide	7.9E-3	2.9E-3	3.1E-4	3.9E-4
Diallate	7.9E-3	1.8E-2	1.6E-3	2.0E-3
1,2-Dibromo-3-chloropropane	7.9E-3	1.8E-2	3.0E-3	6.0E-3
1,2-Dichloroethane	7.9E-3	3.0E-2	6.1E-3	4.0E-2
1,1-Dichloroethane	7.9E-3	3.1E-2	7.0E-3	2.3E-1
Ethyl ether	7.9E-3	2.9E-2	6.8E-3	1.4

[a] Used model shower parameters provided in Table C.5 , but with non-shower water use rates set to zero.

[b] Calculated assuming transfer efficiency, $t_{eff} = 0.63$ for $H' \geq 10^{-4}$ and $t_{eff} = 0$ for $H' < 10^{-4}$ (Andelman, 1990).

[c] Calculated using Equation 11 from McKone and Bogen (1992).

[d] Calculated using Appendix A shower model with McKone (1987) mass transfer dependency.

[e] Calculated assuming Henry's law equilibrium partitioning based on C_{water} = 1 mg/L.

Table C.4. Model Predictions For Average Bathroom Air Exposure Concentration[a]

Compound	Average Bathroom Air Concentration (mg/L)		
	McKone and Bogen[b]	McKone[c]	Saturated Air[d]
Bisphenol A	2.0E-8	4.0E-10	4.1E-9
Phthalic anhydride	6.3E-6	6.8E-8	6.7E-7
2,4-Dinitrophenol	9.0E-5	1.8E-6	1.8E-5
Quinoline	7.9E-4	1.1E-5	1.1E-4
Heptachlor epoxide	9.6E-4	3.6E-5	3.9E-4
Diallate	1.1E-3	1.8E-4	2.0E-3
1,2-Dibromo-3-chloropropane	1.1E-3	4.1E-4	6.0E-3
1,2-Dichloroethane	9.9E-3	9.5E-4	4.0E-2
1,1-Dichloroethane	1.0E-2	1.1E-3	2.3E-1
Ethyl ether	9.7E-3	1.1E-3	1.4

[a] Used model shower parameters provided in Table C.5 , but with non-shower water use rates set to zero.

[b] Calculated using Equation 11 from McKone and Bogen (1992).

[c] Calculated using Appendix A shower model with McKone (1987) mass transfer dependency.

[d] Calculated assuming Henry's law equilibrium partitioning based on C_{water} = 1 mg/L.

C.2 References

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