

**Developing Probability Distributions for Model Variables**

Due to inherent natural variability, the exposure model variables are defined in terms of Probability Distribution Functions (PDFs) estimated from a limited set of observations. The PDFs are either derived from data presented in previous studies (EPA, 1997; Mayer et al., 1998; EPA, 2000) or taken directly from the literature (EPA, 1995). Separate PDFs for natural variability ( $PDF_v$ ) and knowledge uncertainty ( $PDF_u$ ) are used in a nested Monte Carlo (EPA, 1995) simulation. Most model variables conform to a lognormal distribution. When normality of the variable can be assumed, the true mean can be estimated from the student's-t distribution, and the true standard deviation from the chi-squared distribution (Kottegoda and Rosso, 1997). Thus, when the data follow a lognormal distribution, the geometric mean is obtained from the inverse student's-t distribution, and geometric standard deviation from the inverse chi-squared distribution. During the evaluation of the student's-t and chi-squared distributions, a quality factor (qf) is employed (EPA, 1995) instead of the sample size of the observed data. Values of 100, 25, or 10 are assigned depending on the sample size and the quality of fit to the distribution. The frequency of use of shower, toilet and washing machine conform to the gamma distribution. When the gamma distribution is used, no  $PDF_u$  is implemented. This implies that the distribution has no associated knowledge uncertainty and that the values of the parameters are constant. The parameters for the distributions are estimated using the least-squares method. To test the appropriateness of the derived distributions, goodness-of-fit tests were conducted. Due to the data richness (for example, more than 48,000 data points for the shower event), the goodness-of-fit test fails even when the fit is excellent (Figure S1). It has been suggested that goodness-of-fit tests should not be the

primary method for determining adequacy of fit for large data sets, because the method becomes extremely sensitive when the sample size is large (EPA, 1999). In these cases, visual inspection was used instead.

When none of the conventional PDFs were appropriate, an Empirical Distribution Function (EDF) comprised of the actual sample data was used (see Figure S2). The number of residents in the house ( $p_{num}$ ) and the frequency of use of the bath ( $f_{bath}$ ) and dishwasher ( $f_{DW}$ ) were represented as EDFs (Tables S1 and S2). Based on over 6000 observations (EPA, 1995), the probability that a household has 1 to 6 occupants is computed (Table S1). For the frequency of operation of bath and dishwasher, the measurements by Mayer et al. (1998) are adopted as EDFs. A random number between 0 and 1 is compared with the cumulative frequency to assign an appropriate value for the variable with EDFs. For example, if the random number is less than 0.192, one person is assumed to live in the house ( $p_{num} = 1$ ), and if it is between 0.192 and 0.520 ( $0.192 + 0.328$ ), two people reside in the house. Uncertainty for these EDFs is not considered in this study.

Although the volume of bath water does not fit a generic distribution, an EDF with the observed data also does not appear to represent the true values for the population. Therefore, a lognormal distribution with a very low  $qf$  of 10 was assumed (Figure S3). A complete description of the distributions for the input variables is presented elsewhere (Kim et al., 2002). Tables S1 and S2 summarize the PDFs used for all the model input variables.

### **Predicting Mass Transfer Coefficients as a Function of Operating Conditions**

Mass transfer coefficients may vary substantially depending on the characteristics of a water device and different operating conditions (temperature, volume, water flow rate, and air flow rate). Several studies have experimentally characterized the major factors influencing the mass transfer coefficient of the shower (Andelman et al., 1986; Giardino et al., 1992; Little, 1992; Keating et al., 1997; Corsi and Howard, 1998). A more complete listing of the wide range of operating conditions used in experiments involving showers is provided in Table 3. Despite the available data, a systematic approach to include the effects of operating conditions on the shower mass transfer coefficient has not been established. In this study, correlation equations are developed for as many of the water-using devices as possible. Variables representing the measured conditions such as water flow rate ( $Q_L$ ) in  $L \text{ min}^{-1}$ , air flow rate ( $Q_G$ ) in  $L \text{ min}^{-1}$ , temperature ( $T$ ) in  $^{\circ}\text{C}$ , and volume ( $V$ ) in  $L$ , are correlated with the liquid-phase mass transfer coefficient ( $K_L A$ ) in  $L \text{ min}^{-1}$  and gas-phase mass transfer coefficient ( $K_G A$ ) in  $L \text{ min}^{-1}$  or in some cases with the overall mass-transfer coefficient ( $K_{OL} A$ ) in  $L \text{ min}^{-1}$ .

According to the two-resistance theory, the overall resistance is the sum of two resistances in series (Little, 1992), or

$$\frac{1}{K_{OL} A} = \frac{1}{K_L A} + \frac{1}{H \cdot K_G A} \quad (\text{S1})$$

where  $H$  is the Henry's law constant. To use Equation S1,  $K_L A$  and  $K_G A$  need to be evaluated separately. Thus, nonlinear regression analysis was conducted to determine the effects of operating conditions on both  $K_L A$  and  $K_G A$ . In some instances,  $K_L A$  and  $K_G A$

could not be separately determined due to insufficient experimental measurements (bath) or due to a rapid approach to equilibrium (dishwasher). In these cases, the effect of operating conditions on the overall mass transfer coefficient ( $K_{OLA}$ ) was determined instead. When no experimentally measured mass transfer coefficient was available (toilet),  $K_{LA}$  and  $K_{GA}$  were computed from the transfer efficiency (TE) for radon and an estimate of the ratio of the gas phase to liquid phase mass transfer coefficient ( $K_G/K_L$ ) (Little and Chiu, 1998).

### Shower

The shower contributes significantly to inhalation exposure in residential environments. Although many studies measured mass transfer coefficients for the shower (Table S3), only two papers (Tancrede et al., 1992; Corsi and Howard, 1998) published sufficient data to separately evaluate  $K_{LA}$  and  $K_{GA}$ . For this analysis, TCE is selected as the reference chemical. Because Corsi and Howard (1998) did not measure mass transfer coefficients for TCE, their measured values for toluene are corrected to TCE according to the suggested method.

$K_{LA}$  is expected to increase with water flow rate ( $Q_L$ ) and water temperature (T).

The following relationship was therefore assumed:

$$K_{LA} \propto Q_L^{\beta_1} \cdot \beta_2^{(T-20)} \quad (S2)$$

By taking the logarithm of both sides, the non-linear regression equation is transformed to a multiple linear regression equation, or

$$\log K_L A = \beta_0 + \beta_1 \cdot \log Q_L + \beta_2 \cdot (T - 20) + \epsilon \quad (\text{S3})$$

where  $\beta_0$ ,  $\beta_1$ ,  $\beta_2$  are fitting parameters, and  $\epsilon$  is a random error that is assumed to be normally distributed with mean 0 and variance  $\sigma_\epsilon^2$  (Kottegoda and Rosso, 1997). The regression analysis gives

$$\log K_L A = 0.84 \cdot \log Q_L + 0.0057 \cdot (T - 20) + 0.20 + \epsilon \quad (R^2 = 0.9) \quad (\text{S4})$$

with the normal distribution for  $\epsilon$  of

$$\epsilon \sim N(0, 0.0018)$$

Introducing the normally distributed uncertainty allows natural variability of the mass transfer coefficient to be incorporated in the exposure model.

While  $K_G$  is mainly dependent on the air flow rate ( $Q_G$ ), the interfacial surface area ( $A$ ) depends on the water flow rate ( $Q_L$ ). Therefore, shower flow rate ( $Q_L$ ) and air flow rate between shower stall and bathroom ( $Q_G$ ) were chosen to represent  $K_G A$ . The regression analysis yielded

$$\log K_G A = 0.52 \cdot \log Q_L + 0.74 \cdot \log Q_G + 0.83 + \epsilon \quad (R^2 = 0.9) \quad (\text{S5})$$

$$\epsilon \sim N(0, 0.016)$$

The mass transfer coefficients for the shower can be estimated from either liquid-phase concentration of the chemical entering and leaving the shower compartment or the changing gas-phase concentration in the shower stall (Little, 1992). The mass transfer coefficients inferred from gas-phase concentrations are up to six times higher than those obtained from aqueous-phase concentrations (Table S4). Reasons that may explain this difference include possible gas-phase sinks inside the shower stall, incomplete mixing of the shower air, or inaccuracies in experimental measurements. However, as shown in Table S4, when mass recovery (mass recovered in the air/mass lost from water) approaches 100%, the mass transfer coefficients obtained from water and air data become similar. To test the overall approach  $K_{OLA}$  values predicted using Equations S1 and S5 (based only on the data of Tancrede et al. (1992) and Corsi and Howard (1998)) are compared in Figure S4 to the measured values obtained from the entire range of experimental studies shown in Table S3 (based on aqueous-phase concentrations). Despite the substantial variation in operating conditions shown in Table S3, many of the measured values are predicted to within 20%.

Unlike the shower, only one study (Corsi and Howard, 1998) measured mass transfer coefficients for bath, faucet, washing machine, and dishwasher, and reported the corresponding operating conditions. These values were used in the nonlinear regression approach described above. In these cases, toluene is selected as the reference chemical. When no clear relationship between the mass transfer coefficient and the operating conditions can be inferred, a simple uniform distribution is assumed.

Bath

The same value of the overall mass transfer coefficient for the bath was obtained in three separate experiments. Thus, a constant overall mass transfer coefficient is assumed regardless of the operating conditions

$$K_{OL}A = 1.2 \quad (S6)$$

The published range for the ratio between gas- and liquid-phase mass transfer coefficient is implemented as a uniform distribution, or

$$K_G/K_L \sim U(54, 78) \quad (S7)$$

Faucet

The flow rate of the faucet is the only varying operating condition (Corsi and Howard, 1998), so a correlation between the water flow rate and liquid-phase mass transfer coefficient is assumed:

$$\log K_L A = 0.90 \cdot \log Q_L - 0.56 + \epsilon \quad (R^2 = 0.4) \quad (S8)$$

$$\epsilon \sim N(0, 0.016)$$

No meaningful relationship could be deduced between flow rate of the faucet and gas-phase mass transfer coefficient, so a uniform distribution is used:

$$K_{GA} \sim U(21, 108) \quad (S9)$$

### Dishwasher

$K_{LA}$  and  $K_{GA}$  could not be determined separately for the dishwasher (Corsi and Howard, 1998) because the high temperature and significant agitation led to rapid establishment of equilibrium inside the dishwasher even for chemicals of low volatility. Thus only the impact of temperature is included in the regression analysis

$$\log K_{OL}A = 0.0027 \cdot (T - 20) + 1.4 + \epsilon \quad (R^2 = 0.3) \quad (S10)$$

$$\epsilon \sim N(0, 0.0012)$$

The ratio between gas- and liquid-phase mass transfer coefficient of 160 suggested by Corsi and Howard (1998) is used

$$K_G/K_L = 160 \quad (S11)$$

### Washing Machine – Fill Cycle

$K_{LA}$  and  $K_{GA}$  are evaluated for three separate cycles (fill, wash, and rinse) of the washing machine using the approach taken for the shower data

$$\log K_{LA} = 0.0077 \cdot (T - 20) + 0.69 + \epsilon \quad (R^2 = 0.3) \quad (S12)$$

$$\epsilon \sim N(0, 0.025)$$

$$\log K_G A = 0.97 \cdot \log Q_G - 1.8 \cdot \log Q_L + 1.8 + \epsilon \quad (R^2 = 0.8) \quad (S13)$$

$$\epsilon \sim N(0, 0.023)$$

Washing Machine – Wash Cycle

$$\log K_L A = 0.20 \cdot (T - 20) - 9.8 \cdot \log Q_G + 17 + \epsilon \quad (R^2 = 0.7) \quad (S14)$$

$$\epsilon \sim N(0, 0.19)$$

$$\log K_G A = 0.056 \cdot (T - 20) - 2.6 \cdot \log Q_G + 5.3 + \epsilon \quad (R^2 = 0.4) \quad (S15)$$

$$\epsilon \sim N(0, 0.056)$$

Washing Machine – Rinse Cycle

$$\log K_L A = -2.5 \cdot \log V + 5.7 + \epsilon \quad (R^2 = 0.3) \quad (S16)$$

$$\epsilon \sim N(0, 0.32)$$

$$\log K_G A = -1.5 \cdot \log V + 4.1 + \epsilon \quad (R^2 = 0.4) \quad (S17)$$

$$\epsilon \sim N(0, 0.090)$$

where V is the volume of water used for the rinse cycle.

Toilet

No experimental data to determine mass transfer coefficients for the toilet are available. In the absence of experimental data, the transfer efficiency for radon (Little et al., 1998) together with an estimate of  $K_G/K_L$  is used to calculate the  $K_G A$  and  $K_L A$  values for the toilet:

$$K_{OL} A_{Rn,toilet} = \frac{V_{L,toilet}}{\text{day}} \cdot \frac{TE_{Rn}}{1 - TE_{Rn}} \quad (S18)$$

$$K_L A_{Rn} = K_{OL} A_{Rn} \frac{\left( \left( \frac{K_G}{K_L} \right)_{Rn} \cdot H_{Rn} + 1 \right)}{\left( \left( \frac{K_G}{K_L} \right)_{Rn} \cdot H_{Rn} \right)} \quad (S19)$$

$$K_G A_{Rn} = K_L A_{Rn} \cdot \left( \frac{K_G}{K_L} \right)_{Rn} \quad (S20)$$

where  $K_{OL} A_{Rn}$  is the overall mass transfer coefficient for radon,  $V_{L,toilet}$  is the volume of water used in toilet flushing per day,  $TE_{Rn}$  is the transfer efficiency for radon,  $K_L A_{Rn}$  is the liquid phase mass transfer coefficient for radon,  $K_G/K_L$  is the ratio of the gas phase to liquid phase mass transfer coefficient, and  $H_{Rn}$  is the Henry's law constant for radon.

Mass transfer coefficients for a reference chemical, in combination with Henry's law constant, diffusion coefficients, and water temperature, are employed to determine the overall mass transfer coefficient of the contaminant of interest in the given device. Theoretical and experimental studies indicate that turbulent mass-transfer coefficients

generally depend on the diffusion coefficient raised to some power, the magnitude of which lies between 1/2 and 2/3 (Little, 1992). If no better information is available, the following relationships are sometimes assumed (Corsi and Howard, 1998)

$$\frac{K_L A_i}{K_L A_{Ref}} \propto \left( \frac{D_{L,i}}{D_{L,Ref}} \right)^{\frac{2}{3}} \quad (S21)$$

$$\frac{K_G A_i}{K_G A_{Ref}} \propto \left( \frac{D_{G,i}}{D_{G,Ref}} \right)^{\frac{2}{3}} \quad (S22)$$

where  $D_L$  is liquid phase diffusion coefficient,  $D_G$  is gas phase diffusion coefficient, and  $i$  and  $ref$  stand for the chemical of interest and a reference chemical respectively. For temperatures other than 20°C, the Henry's law constant,  $H$ , is adjusted (Staudinger and Roberts, 2001) according to

$$\log H = A - \frac{B}{T} \quad (S23)$$

where  $H$  has units of gas-phase concentration/aqueous-phase concentration,  $T$  is in K and  $A$  (-) and  $B$  (K) are fitting parameters.

### **Water Use and Activity Patterns**

Water use and activity patterns for the people residing in the house are needed to compute the gas-phase concentrations in each compartment and subsequently to estimate absorbed dose of the chemicals. With water consumption data measured for 1800 households in the United States and Canada (Mayer et al., 1998), typical daily water use patterns for each device are identified (Figure S5). Two water use peaks occurred (one in the morning and the other in the evening), with slightly lower water use during the day. Most people took a shower in the morning between 6:00 a.m. and 9:00 a.m., or in the evening between 6:00 p.m. and 11:00 p.m. Thus, consistent with the activity pattern adopted by EPA (1995), it is assumed that the first person starts to shower at 7:00 a.m., the second shower starts after the first person leaves the bathroom, and so on. When a person takes showers more than once per day, they are assumed to take the first shower in the morning and the others in the evening. The time spent in the bathroom after each shower is assumed to be the same for all the occupants with the value between 1 and 30 minutes ( $t_b$  in Table S1). The other devices are assumed to be operated at any time within the allowed time periods. The toilet is flushed between 5:00 a.m. and 12:00 a.m. The faucet can be used between 6:00 a.m. and 12:00 a.m. The washing machine and dishwasher can be operated between 9:00 a.m. and 10:00 p.m., and between 6:00 p.m. and 11:00 p.m., respectively. A person may take a bath between 7:00 p.m. and 11:00 p.m. Because this was not explicitly evaluated by Mayer et al. (1998), the use of a bathroom faucet is not considered in this study. The frequency ( $f$ ) of each water device operation is implemented as a distribution using the data from Mayer et al. (1998) (Table S2). The total amount of water consumed in a household is determined by the number of

residents, the frequency of operation of each device, flow rate of shower and faucet, and the volume of water used for each operation of dishwasher, washing machine, and toilet. The gas-phase concentration profile in each of the three compartments is coupled with the human activity pattern to calculate individual exposure via inhalation. An Occupancy Factor (OF), the fraction of time a person spends at home, is applied to quantify the time a person spends in the home (EPA, 1995).

### **Validation of the Model**

For the validation of the model, the indoor air concentrations of chemicals reported in previous studies are compared to the results of the model simulations. EPA's Total Exposure Assessment Methodology (TEAM) studies measured the concentrations of volatile organic chemicals in residential drinking water and in personal, indoor, outdoor and exhaled air for about 800 participants in eight cities (Wallace, 1997). Chloroform is chosen for the comparison, because it is one of the most commonly measured compounds. To measure exposure concentration, participants in the TEAM studies wore personal air quality monitors throughout the day except when they took a shower or bath. Personal air concentration for chloroform ranged from  $6.1 \times 10^{-7}$  to  $1.0 \times 10^{-3}$   $\text{mg L}^{-1}_{\text{air}}$  per  $\text{mg L}^{-1}_{\text{water}}$  with a sample size weighted mean and median of  $7.7 \times 10^{-5}$  and  $4.3 \times 10^{-5}$   $\text{mg L}^{-1}_{\text{air}}$  per  $\text{mg L}^{-1}_{\text{water}}$ , respectively (Table S5). The average chloroform exposure concentration from the Monte Carlo simulation ( $4.6 \times 10^{-5}$   $\text{mg L}^{-1}_{\text{air}}$  per  $\text{mg L}^{-1}_{\text{water}}$ ) is comparable to the measured personal air concentrations obtained in the TEAM studies. While indoor chloroform concentration in Canada varied by an order of magnitude ranging from  $3 \times 10^{-6}$  to  $3 \times 10^{-5}$   $\text{mg L}^{-1}$ , indoor concentrations normalized to the water

concentration ranged from  $2.5 \times 10^{-5} \text{ mg L}^{-1}_{\text{air}}$  per  $\text{mg L}^{-1}_{\text{water}}$  (Weisel et al., 1999) to  $4.5 \times 10^{-4} \text{ mg L}^{-1}_{\text{air}}$  per  $\text{mg L}^{-1}_{\text{water}}$  (Lévesque and Ayotte, 2002) (Table S6). The indoor chloroform concentration normalized to water concentration in Quebec City (Lévesque and Ayotte, 2002) is higher than the values in the TEAM studies by an order of magnitude. The discrepancy may arise due to different housing characteristics including ventilation and size. Lévesque and Ayotte (2002) also presented chloroform concentrations during and after a 15 minute shower (Table S6). Although the predicted concentration in the main house before the shower is lower than the measured chloroform concentration by an order of magnitude, the predicted shower and bathroom air concentrations during and after the shower are similar to the measured values.

### Nomenclature

$A_{\text{body}}$	surface area of body ( $\text{L}^2$ )
$A_{\text{hand}}$	surface area of hands ( $\text{L}^2$ )
BR	breathing rate ( $\text{L}^3\text{t}^{-1}$ )
$D_{\text{L}}$	liquid-phase diffusion coefficient ( $\text{L}^2\text{t}^{-1}$ )
$D_{\text{G}}$	gas-phase diffusion coefficient ( $\text{L}^2\text{t}^{-1}$ )
$f_{\text{sh}}$	frequency of showering ( $\text{cap}^{-1}\text{t}^{-1}$ )
$f_{\text{bath}}$	frequency of bath ( $\text{cap}^{-1}\text{t}^{-1}$ )
$f_{\text{toilet}}$	frequency of toilet flush ( $\text{cap}^{-1}\text{t}^{-1}$ )
$f_{\text{faucet}}$	frequency of faucet use ( $\text{cap}^{-1}\text{t}^{-1}$ )
$f_{\text{DW}}$	frequency of dishwasher operation ( $\text{cap}^{-1}\text{t}^{-1}$ )
$f_{\text{WM}}$	frequency of washing machine operation ( $\text{cap}^{-1}\text{t}^{-1}$ )
H	dimensionless Henry's law constant ( $\text{ML}^{-3}_{\text{air}}$ per $\text{ML}^{-3}_{\text{water}}$ )
$K_{\text{GA}}$	gas-phase mass transfer coefficient ( $\text{L}^3\text{t}^{-1}$ )
$K_{\text{LA}}$	liquid-phase mass transfer coefficient ( $\text{L}^3\text{t}^{-1}$ )
$K_{\text{OLA}}$	overall mass transfer coefficient ( $\text{L}^3\text{t}^{-1}$ )
$K_{\text{G}}/K_{\text{L}}$	ratio of gas phase to liquid phase mass transfer coefficient (1)
OF	occupancy factor (1)
pnum	number of people in the house (1)
$Q_{\text{G},\text{f}}$	ventilation rate of the fan in the bathroom ( $\text{L}^3\text{t}^{-1}$ )
$Q_{\text{L},\text{sh}}$	shower flow rate ( $\text{L}^3\text{t}^{-1}$ )
$Q_{\text{G},\text{sh}}$	ventilation rate in the shower stall ( $\text{L}^3\text{t}^{-1}$ )
$Q_{\text{L},\text{faucet}}$	faucet flow rate ( $\text{L}^3\text{t}^{-1}$ )
$R_{\text{s}}$	residence time of air in the shower stall (t)

$R_{b1}$	residence time of air in the bathroom when the bathroom door is closed (t)
$R_{b2}$	residence time of air in the bathroom when the bathroom door is open (t)
$R_a$	residence time of air in the main house (t)
$t_b$	time spent in the bathroom after showering (t)
$t_{sh}$	time in shower (t)
$t_{bath}$	time in bath (t)
$t_{faucet}$	time of faucet use (t)
$t_{DW}$	time of dishwasher operation (t)
$t_{WM}$	time of washing machine operation (t)
$T_{sh}$	temperature of shower water (T)
$T_{bath}$	temperature of bath water (T)
$T_{faucet}$	temperature of faucet water (T)
$T_{toilet}$	temperature of toilet water (T)
$T_{DW}$	temperature of water used for dishwasher (T)
$T_{WM}$	temperature of water used for washing machine (T)
$TE_{Rn}$	transfer efficiency for radon (1)
$V_s$	volume of shower stall ( $L^3$ )
$V_b$	volume of bathroom ( $L^3$ )
$V_a$	volume of main house ( $L^3$ )
$V_{t1}$	total volume of the house when 1 person is living in the house ( $L^3$ )
$V_{t2}$	total volume of the house when 2 people are living in the house ( $L^3$ )
$V_{t3}$	total volume of the house when 3 people are living in the house ( $L^3$ )
$V_{t4}$	total volume of the house when 4 people are living in the house ( $L^3$ )
$V_{t5}$	total volume of the house when 5 people are living in the house ( $L^3$ )
$V_{t6}$	total volume of the house when 6 people are living in the house ( $L^3$ )
$V_{bath}$	volume of bath water ( $L^3$ )
$V_{toilet}$	volume of toilet water ( $L^3$ )
$V_{DW}$	volume of water used for dishwasher ( $L^3$ )
$V_{WM}$	volume of water used for washing machine ( $L^3$ )
$V_{I,dir}$	volume of water consumed directly ( $L^3$ )
$V_{I,tot}$	total tap water consumed both directly and indirectly ( $L^3$ )
$VR_a$	air exchange rate in the main house ( $t^{-1}$ )

Footnote: M is mass, L is length, t is time, T is temperature, cap is capita and 1 represents a dimensionless quantity.

**References**

- (S1) Andelman, J.B. *Environmental Health Perspectives* **1985**, 62, 313-318.
- (S2) Andelman, J.B. *The Science of the Total Environment* **1985**, 47, 443-460.
- (S3) Andelman, J.B.; Couch, A.; Thurston, W.W. In *Environmental Epidemiology*; Kopfler, F.; Craun, G.F. Eds; 1986; pp 201-213.
- (S4) Chan, C. C.; Vainer, L.; Martin, J. W.; Williams, D. T. *J. Air Waste Manage. Assoc.* **1990**, 40, 62-67.
- (S5) Corsi, R.; Howard, C. *Volatilization Rates from Water to Indoor Air, Phase II*; U.S. Environmental Protection Agency, Office of Research and Development; Washington, DC, 1998.
- (S6) *Uncertainty Analysis of Risks Associated with Exposure to Radon in Drinking Water*; U.S. Environmental Protection Agency; Washington, DC, 1995; EPA-822-R-96-005.
- (S7) *Exposure Factors Handbook*; U.S. Environmental Protection Agency, Office of Research and Development National Center for Environmental Development; Washington, DC, 1997; EPA-600-P-95-002Fa.
- (S8) *Report of the Workshop on Selecting Input Distributions for Probabilistic Assessments*; U.S. Environmental Protection Agency, Risk Assessment Forum; Washington, DC, 1999; EPA-630-R-98-004.
- (S9) Estimated Per Capita Water Ingestion in the United States: Based on Data Collected by the United States Department of Agriculture's 1994-96 Continuing Survey of Food Intakes by Individuals; U.S. Environmental Protection Agency, Office of Water; Washington, DC, 2000; EPA-822-00-008.
- (S10) Giardino, N.J.; Andelman, J.B. Poster paper presented at the annual conference of the American Water Works Association, Philadelphia, PA, June 1991.
- (S11) Giardino, N.J.; Gumerman, E.; Esmen, N.A.; Andelman, J.B. *Journal of Exposure Analysis and Environmental Epidemiology* **1992**, Supplement 1, 147-158.
- (S12) Giardino, N.J.; Esmen, N.A.; Andelman, J.B. *Environ. Sci. Technol.* **1992**, 26, 1602-1606.
- (S13) Giardino, N.J.; Andelman, J.B. *Journal of Exposure Analysis and Environmental Epidemiology* **1996**, 6, 413-423.
- (S14) Giardino, N.J.; Hageman, J.P. *Environ. Sci. Technol.* **1996**, 30, 1242-1244.
- (S15) Guy, R. H.; Maibach, H. I. In *Percutaneous Absorption*; Bronaugh, R. L.; Maibach, H. I., Eds.; Marcel Dekker Inc.: New York, 1989; p 301.
- (S16) Hodgson, A.T.; Garbesi, K.; Sextro, R.G.; Daisey, J.M. Lawrence Berkeley Laboratory Report No. LBL-25465; Berkeley, CA, 1988.
- (S17) Jo, W.K.; Weisel, C.P.; Liroy, P.J. *Risk Analysis* **1990**, 10, 575-580.
- (S18) Jo, W.K.; Weisel, C.P.; Liroy, P.J. *Risk Analysis* **1990**, 10, 581-585.
- (S19) Keating, G.A.; McKone, T.E.; Gillett, J.W. *Atmospheric Environment* **1997**, 31(2): 123-130.
- (S20) Kerger, B.D.; Schmidt, C.E.; Paustenbach, D.J. *Risk Analysis* **2000**, 20(5): 637-651.
- (S21) Kim, E.; Little, J.C. *Exposure to Chemical Contaminants in Drinking Water*, Report to Environmental Protection Agency, Office of Water, Washington, DC, 2003.

- (S22) Kottegoda, N. T.; Rosso, R. *Statistics, Probability, and Reliability for Civil and Environmental Engineers*; McGraw Hill Companies Inc., 1997.
- (S23) Lévesque, B.; Ayotte, P. *J. Toxicol. Environ. Health Part A*, **2002**, *65*, 489-502.
- (S24) Little, J. C. *Environ. Sci. Technol.* **1992**, *26*, 1341-1349.
- (S25) Little, J. C.; Khanal, R.; Sankaran, K. *Uncertainty Analysis of Risk Associated with Exposure to Volatile Compounds in Drinking Water*; U.S. Environmental Protection Agency, Office of Water; Washington, DC, 1998.
- (S26) Little, J. C.; Chiu, N. In *Exposure to Contaminants in Drinking Water*; Olin, S. S. Ed.; ILSI Press: Washington, DC, 1998; pp 89-101.
- (S27) Mayer, P.W.; DeOreo, W.B.; Opitz, E.M.; Kiefer, J.C.; Davis, W.Y.; Dziegielewski, B. Nelson, J.O. *Residential end uses of water*; AWWA Research Foundation and American Water Works Association, 1998.
- (S28) McKone, T.E.; Knezovich, J.P. *J. Air Waste Manage. Assoc.* **1991**, *40*, 282-286.
- (S29) Otson, R.; Fellin, P.; Tran, Q. *Proceedings of Indoor Air* **1993**, *2*, 141-146.
- (S30) Staudinger, J.; Roberts, P. V. *Chemosphere* **2001**, *44*, 561-576.
- (S31) Tancrede, M.; Yanagisawa, Y.; Wilson, R. *Atmospheric Environment* **1992**, *26A*, 1103-1111.
- (S32) Wallace, L. A.; Pellizzari, E. D.; Hartwell, T. D.; Rosenzweig, M.; Erickson, M.; Sparacino, C.; Zelon, H. *Environ. Res.* **1984**, *35*, 293-319.
- (S33) Wallace, L. A. *Crit. Rev. Env. Sci. Technol.* **1997**, *27*, 113-194.
- (S34) Weisel, C. P.; Kim, H.; Haltmeier, P.; Klotz, J. B. *Environ. Health. Perspective* **1999**, *107*, 103-110.

Table S1. Distributions for model variables taken from EPA (1995)

Var	Units	Exp <sup>b</sup> Path	PDF <sub>v</sub> <sup>c</sup>	PDF <sub>u</sub> <sup>c</sup>	Values
pnum <sup>a</sup>		I	Empirical Distribution	NA <sup>d</sup>	1 = 0.192 2 = 0.328 3 = 0.183 4 = 0.164 5 = 0.083 6 = 0.049
V <sub>s</sub>	L	I	U(min, max)	min ~ U(a, b) max ~ U(c, d)	a = 1000 b = 1500 c = 2500 d = 3000
V <sub>b</sub>	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(14) s = ln(1.66) qf = 25 min = 4 max = 60
V <sub>t1</sub>	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(205,000) s = ln(1.78) qf = 100 min = 35,000 max = 1,100,000
V <sub>t2</sub>	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = t ln(144,000) s = ln(1.74) qf = 100 min = 30,000 max = 700,000
V <sub>t3</sub>	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(99,000) s = ln(1.68) qf = 100 min = 25,000 max = 450,000
V <sub>t4</sub>	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(89,000) s = ln(1.67) qf = 100 min = 20,000 max = 400,000
V <sub>t5</sub>	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(75,000) s = ln(1.70) qf = 100 min = 15,000 max = 350,000
V <sub>t6</sub>	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(54,000) s = ln(1.78) qf = 100 min = 10,000 max = 300,000
R <sub>s</sub>	min	I	U(min, max)	min ~ U(a, b) max ~ U(c, d)	a = 2 b = 3 c = 4 d = 6
R <sub>b1</sub>	min	I	U(min, max)	min ~ U(a, b) max ~ U(c, d)	a = 20 b = 30 c = 40 d = 50
R <sub>b2</sub>	min	I	U(min, max)	min ~ U(a, b) max ~ U(c, d)	a = 20 b = 30 c = 150 d = 250
Q <sub>G,f</sub>	L min <sup>-1</sup>	I	TRI(min, max, mode)	mode ~ U(a, b)	min = 1000 max = 5000 a = 2000 b = 2500
VR <sub>a</sub>	hr <sup>-1</sup>	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(0.68) s = ln(2.01) qf = 25 min = 0.1 max = 2
t <sub>b</sub>	min	I	U(min, max)	min ~ U(a, b) max ~ U(c, d)	a = 1 b = 10 c = 20 d = 30
BR	L min <sup>-1</sup>	I	TN(mean, std, min, max)	mean ~ t(m, s, qf) std ~ CHISQ(s, qf)	m = 9.1 s = 2.0 qf = 10 min = 2.6 max = 46.6
OF	-	I	B(mean, mode, min, max)	mean ~ U(a, b) mode ~ U(mean, max) or U(min, mean)	a = 0.65 b = 0.80 min = 0.33 max = 1.0

<sup>a</sup> Probability for the number of residents in a household. For example, the probability that a household has two occupants is 0.328.

<sup>b</sup> I: Inhalation exposure

<sup>c</sup> U: Uniform distribution, TLN: Truncated lognormal distribution, TN: Truncated normal distribution, TRI: Triangular distribution, B: Beta distribution, t: Student's-t distribution, CHISQ: Chi-squared distribution.

<sup>d</sup> NA: Not Applicable

Table S2. Distributions for model variables developed from data in other studies

Var	Units	Exp <sup>d</sup> Path	PDF <sub>v</sub> <sup>e</sup>	PDF <sub>u</sub> <sup>e</sup>	Values
$Q_{L,sh}^1$	L min <sup>-1</sup>	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(7.97) s = ln(1.44) qf = 100 min = 1.85 max = 24.6
$T_{sh}^2$	°C	I	U(min, max)	NA <sup>f</sup>	min = 19 max = 51
$t_{sh}^1$	min	I, D	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(7.13) s = ln(1.66) qf = 100 min = 1.66 max = 35.5
$f_{sh}^1$	cap <sup>-1</sup> day <sup>-1</sup>	I, D	G( $\alpha$ , $\beta$ )	NA	$\alpha = 3.40$ $\beta = 0.199$
$V_{bath}^1$	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(79.9) s = ln(1.60) qf = 10 min = 1.16 max = 305
$T_{bath}^a$	°C	I	U(min, max)	NA	min = 22 max = 46
$t_{bath}^3$	min	I, D	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(20.7) s = ln(1.72) qf = 10 min = 2 max = 60
$f_{bath}^1$	cap <sup>-1</sup> day <sup>-1</sup>	I, D	Empirical Distribution	NA	min = 0 max = 2.23 median = 0.019 stdev = 0.12
$V_{toilet}^1$	L cap <sup>-1</sup> day <sup>-1</sup>	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(62.8) s = ln(1.74) qf = 25 min = 9.14 max = 342
$T_{toilet}^a$	°C	I	constant	NA	20
$f_{toilet}^1$	cap <sup>-1</sup> day <sup>-1</sup>	I	G( $\alpha$ , $\beta$ )	NA	$\alpha = 2.43$ $\beta = 2.07$
$Q_{L,faucet}^1$	L min <sup>-1</sup>	I, O	TN(mean, std)	mean ~ t(m, s, qf) stdev ~ CHISQ(m, qf)	m = 4.37 s = 0.686 qf = 100 min = 1.85 max = 6.43
$T_{faucet}^4$	°C	I, O	constant	NA	23
$t_{faucet}^a$	min	I	constant	NA	1
$f_{faucet}^1$	cap <sup>-1</sup> day <sup>-1</sup>	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(8.18) s = ln(1.80) qf = 25 min = 0.61 max = 54.9
$V_{WM}^1$	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(150) gsd = ln(1.30) qf = 25 min = 40.9 max = 409
$T_{WM}^4$	°C	I	U(min, max)	NA	min = 18 max = 51
$t_{WM}^4$	min	I	constant	NA	32
$f_{WM}^1$	cap <sup>-1</sup> day <sup>-1</sup>	I	G( $\alpha$ , $\beta$ )	NA	$\alpha = 2.92$ $\beta = 0.125$
$V_{DW}^1$	L	I	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(35.7) gsd = ln(1.38) qf = 25 min = 5.26 max = 117
$T_{DW}^4$	°C	I	U(min, max)	NA	min = 38 max = 55
$t_{DW}^3$	min	I	constant	NA	42
$f_{DW}^1$	cap <sup>-1</sup> day <sup>-1</sup>	I	Empirical Distribution	NA	min = 0 max = 0.81 median = 0.08 stdev = 0.1
$V_{I,dir}^{5b}$	L day <sup>-1</sup>	O	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(0.351) s = ln(2.64) qf = 10 min = 0.01 max = 3.24
$V_{I,tot}^{5b}$	L day <sup>-1</sup>	O	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(0.702) s = ln(2.21) qf = 10 min = 0.01 max = 4.242
$A_{body}^{3c}$	cm <sup>2</sup>	D	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(18300) gsd = ln(1.11) qf = 25 min = 14500 max = 22800
$A_{hand}^3$	cm <sup>2</sup>	D	TLN(gm, gsd, min, max)	ln(gm) ~ t(m, s, qf) ln(gsd) ~ CHISQ(m, qf)	m = ln(910) gsd = ln(1.10) qf = 25 min = 730 max = 1170

Sources: <sup>1</sup> Mayer et al. (1998); <sup>2</sup> Giardino and Andelman (1996); <sup>3</sup> EPA (1997); <sup>4</sup> Corsi and Howard (1998);  
<sup>5</sup> EPA (2000)

<sup>a</sup> The value is assumed in this study. <sup>b</sup>  $V_{I,ind}$  is the difference between  $V_{I,tot}$  and  $V_{I,dir}$ . <sup>c</sup> Surface area exposed is 91% of total body surface area (Guy and Maibach, 1989). <sup>d</sup> I: Inhalation exposure, O: Ingestion exposure, D: Dermal sorption. <sup>e</sup> G: Gamma distribution. <sup>f</sup> NA: Not Applicable.

Table S3. Operating conditions of published shower experiments

Source	T (°C)	Q <sub>L</sub> (L min <sup>-1</sup> )	Q <sub>G</sub> (L min <sup>-1</sup> )	V (L)
Kerger et al. (2000)	31 – 41	11 – 19	NA <sup>a</sup>	2800 <sup>b</sup>
Corsi and Howard (1998)	21 – 36	6.1, 9.1	343 – 379	1745
Keating et al. (1997)	35 – 45	3.5	195	1530
Giardino and Andelman (1996)	19 – 51	5, 10	26 – 308	1500
Giardino and Hageman (1996)	22	2 – 4	32 – 39	1510
Bernhardt and Hess (1995)	NA <sup>a</sup>	2 – 6	0	NA <sup>a</sup>
Keating and McKone (1993)	45	2.8 – 6	65	1050
Tancrede et al. (1992)	25 – 46	9.7 – 14	34.8	1491
Giardino and Andelman (1991)	42 – 46	5	42 – 66	1200
McKone and Knezovich (1991)	22, 37	9.5	460	2300 <sup>b</sup>
Jo et al. (1990)	40	8.7	0	1666
Giardino et al. (1988)	21	6	0	1100
Hodgson et al. (1988)	40	13.7	1400	2800 <sup>b</sup>
Andelman (1985)	23, 43	0.28	5.4	100

<sup>a</sup> NA: Not Available

<sup>b</sup> shower stall is located inside the bathroom.

Table S4. Measured versus predicted mass transfer coefficients from gas-phase concentrations with fractional mass recovery in the air

Reference	Chemical	$K_{OLA_{meas}}$ ( $L\ min^{-1}$ )	$K_{OLA_{pred}}$ ( $L\ min^{-1}$ )	Mass Recovery
Keating et al. (1997)	Chloroform	5.3 – 6.9	5.4 – 6.2	0.7 – 1.1
Giardino and Andelman (1996)	TCE	17.9	8.3	1.2
	Chloroform	11	7.8	1.4
Tancrede et al. (1992)	CCl <sub>4</sub>	3.3 – 12	14 – 18	0.3 – 0.6
	PCE	5.5 – 7.1	13 – 17	0.5
	TCE	7.4 – 12	13 – 17	0.5 – 0.7
	Chloroform	7.4 – 9.9	12 – 17	0.5 – 0.6
	TCPA	1.0 – 3.0	3.0 – 4.1	0.1 – 0.3
Hodgson et al. (1988)	Freon-12	1.9 – 2.3	19	0.2 – 0.3
	Freon-11	3.3 – 4.4	17	0.2 – 0.4
	PCE	2.4 – 5.0	17	0.3 – 0.4
	TCA	7.5 – 15	18	0.4 – 0.6

Table S5. Concentration of chloroform in tap water and personal air measured in TEAM studies (Wallace et al., 1984; Wallace, 1997)

Location	Time	Tap water <sup>a</sup> ( $\mu\text{g L}^{-1}$ )	Sample <sup>e</sup> size	Personal air <sup>a</sup> ( $\mu\text{g L}^{-1}$ )	Sample <sup>g</sup> size
NJ	1980	128 (11-225)	9	0.0021 (0.00003-0.129)	9
NC	1980	120 (75-191)	3	0.0034 (0.00009-0.0176)	3
NJ	Sep.-Nov., 1981	67 <sup>b</sup> (170 <sup>c</sup> )	340	0.0032	344
NJ	Jul.-Aug., 1982	55 <sup>b</sup> (130 <sup>c</sup> )	156	0.00082 <sup>f</sup>	148
NJ	Feb., 1983	16 <sup>b</sup> (33 <sup>c</sup> )	49	0.0022	48
Greensboro, NC	May, 1982	44 <sup>b</sup> (91 <sup>c</sup> )	24	0.0017	24
Devil's Lake, ND	Oct., 1982	0.38 <sup>b</sup> (1.4 <sup>c, d</sup> )	24	0.00038	24
Los Angeles, CA	Feb., 1984	14	94	0.0013	112
Los Angeles, CA	May, 1984	33	86	0.00048	50
Antioch-Pittsburg, CA	Jun., 1984	49	94	0.00003	68
Los Angeles, CA	Feb., 1987	7.5	9	0.00097	45
Los Angeles, CA	Jun., 1987	9.6	7	0.00048 (0.0024 <sup>b</sup> , 0.076 <sup>c</sup> )	39
Baltimore, MD	Apr., 1987	24	10	0.0031	55

<sup>a</sup> Median values are presented as concentrations, noted otherwise.

<sup>b</sup> Mean concentration

<sup>c</sup> Maximum concentration

<sup>d</sup> It was measured in the rural area

<sup>e</sup> Sample size for tap water concentration.

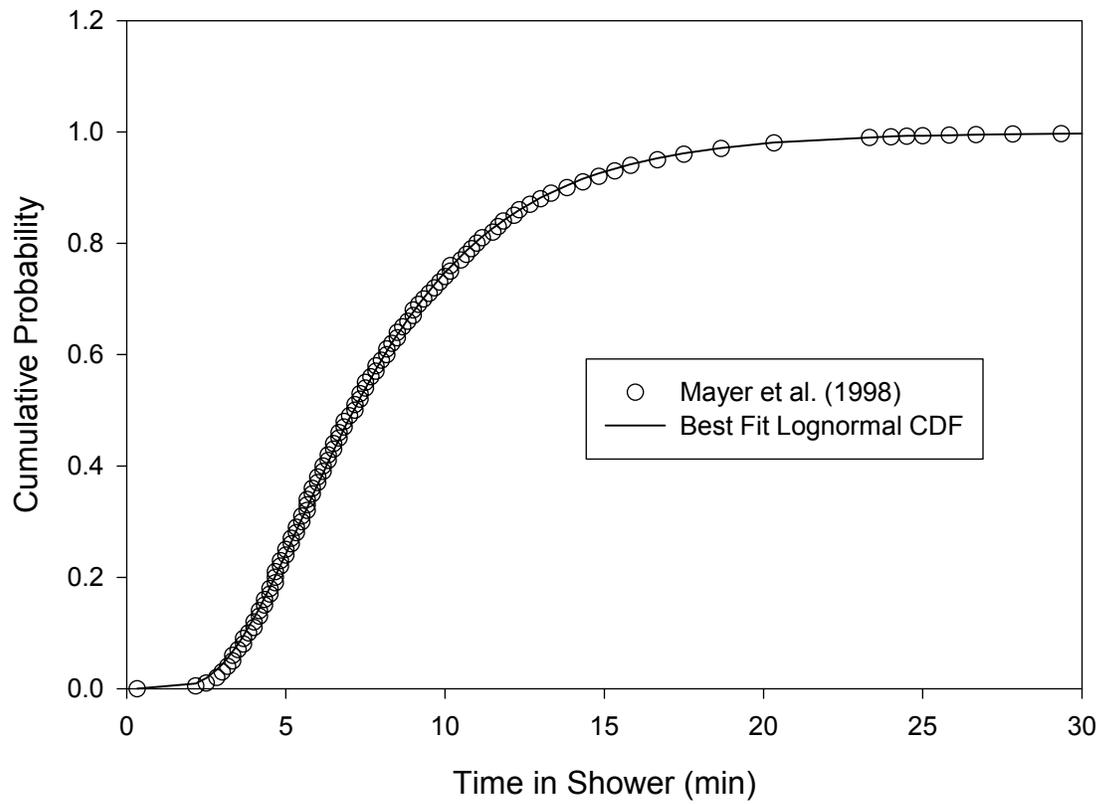
<sup>f</sup> There is a chance of contamination. The background level was  $5.5\mu\text{g m}^{-3}$ .

<sup>g</sup> Sample size for personal air concentration.

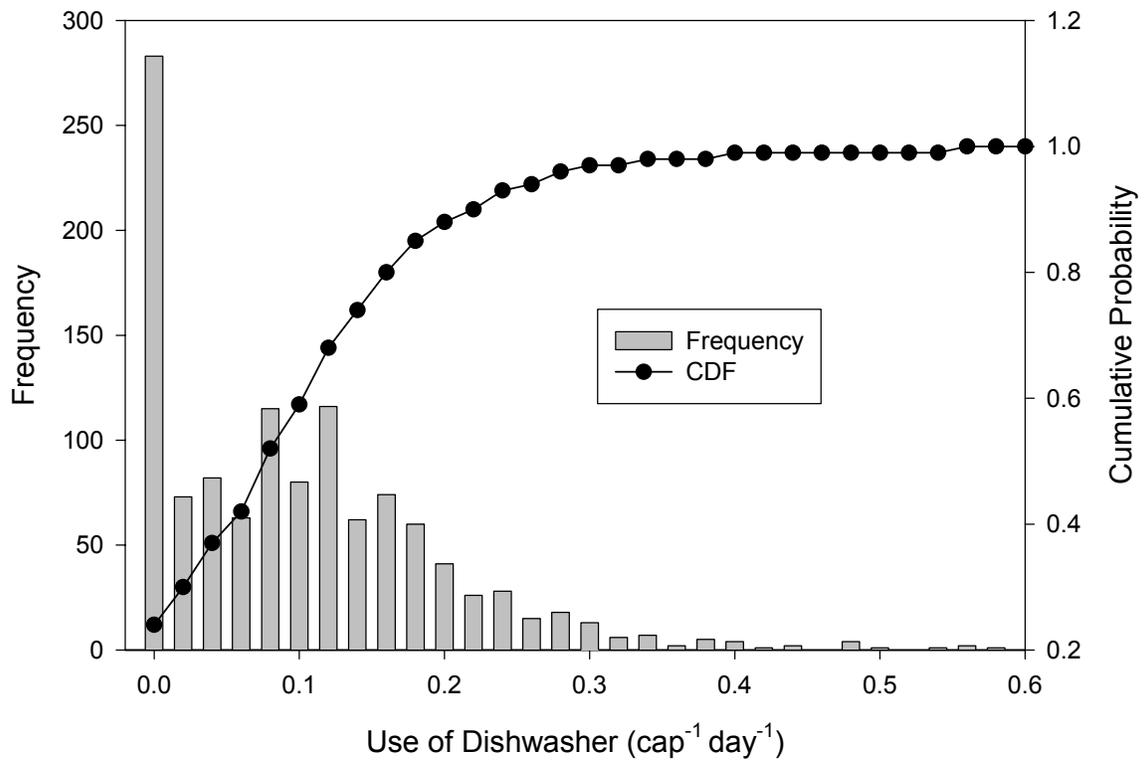
Table S6. Concentration of chloroform in tap water, indoor air, and breath air

Study	Medium	Conc. ( $\mu\text{g L}^{-1}$ )	Location	Sample Size	Comment <sup>b</sup>
Chan et al., 1990	indoor air	0.0251	Canada	12	mean (Nov/Dec, 1986) (Feb/Mar 1987)
		0.003		6	
Otson et al., 1993	indoor air	0.009	Canada	757	mean
Weisel et al., 1999	tap water	16	NJ	49	0.04 – 200 (mean: 31) < 0.0001 – 0.025 for $C_w < 10$ . for $C_w > 10$
	indoor air	0.0004		48	
		0.0002		25	
		0.00125		23	
Lévesque and Ayotte, 2002	shower water	20.1 <sup>a</sup>	Quebec City, Canada	18	14 – 53
	indoor air	0.0097		18	before shower
	shower air	0.147		18	during shower
	bathroom air	0.0358		18	first 15-min period after shower
		0.0204		18	second 15-min period after shower

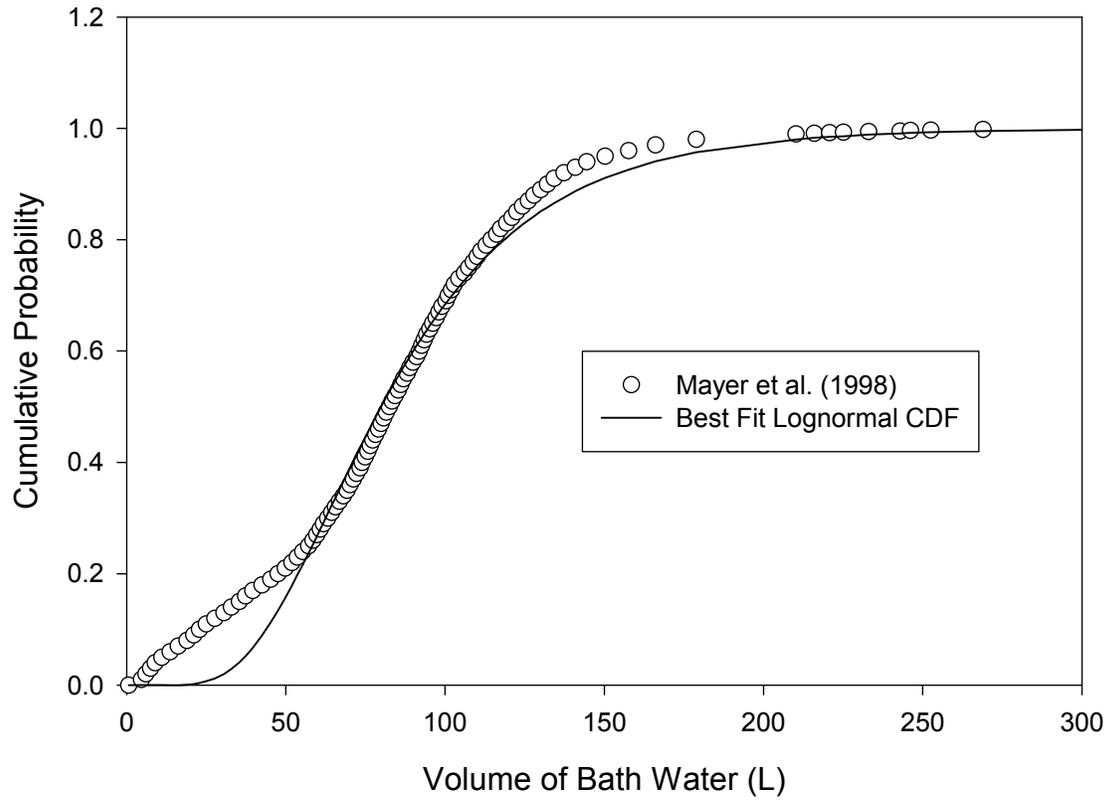
<sup>a</sup> average concentration<sup>b</sup>  $C_w$  is tap water concentration



**Figure S1. Fitting lognormal cumulative distribution function to time in shower**



**Figure S2. Histogram and cumulative distribution function (CDF) for dishwasher use**



**Figure S3. Fitting lognormal cumulative distribution function to the volume of bath water**

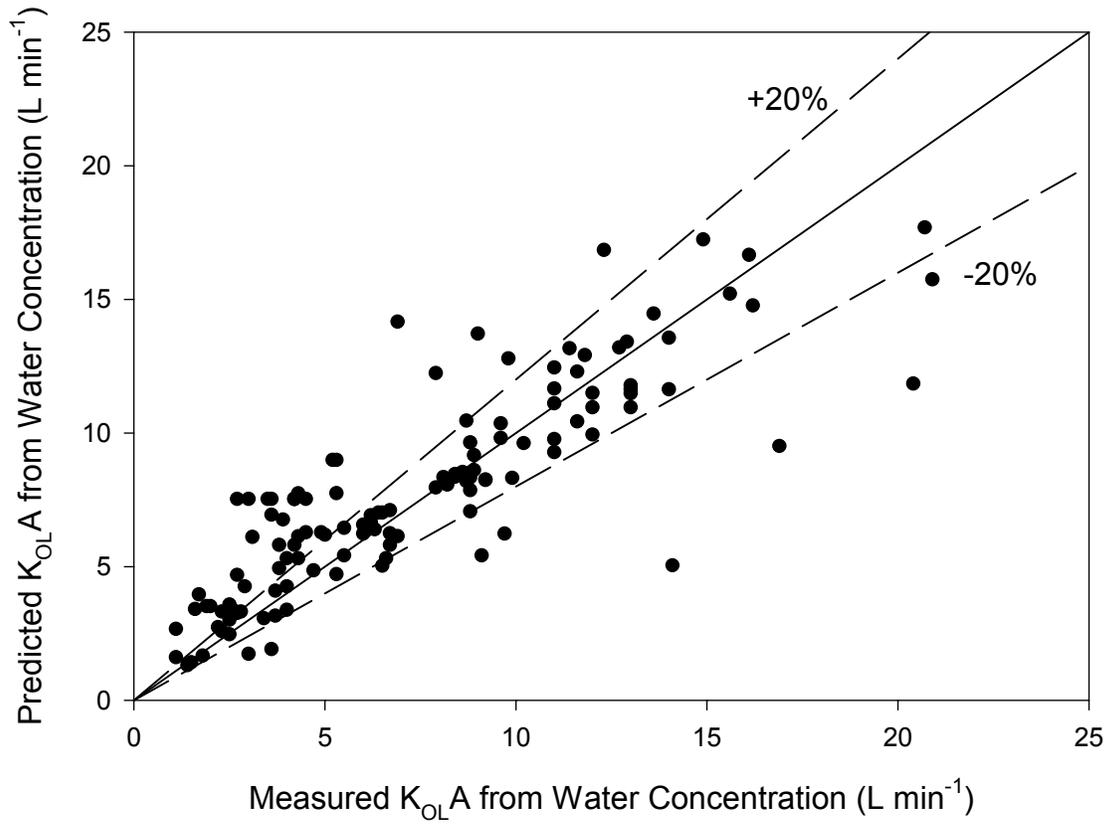


Figure S4. Predicted versus measured values of the overall mass transfer coefficient for shower

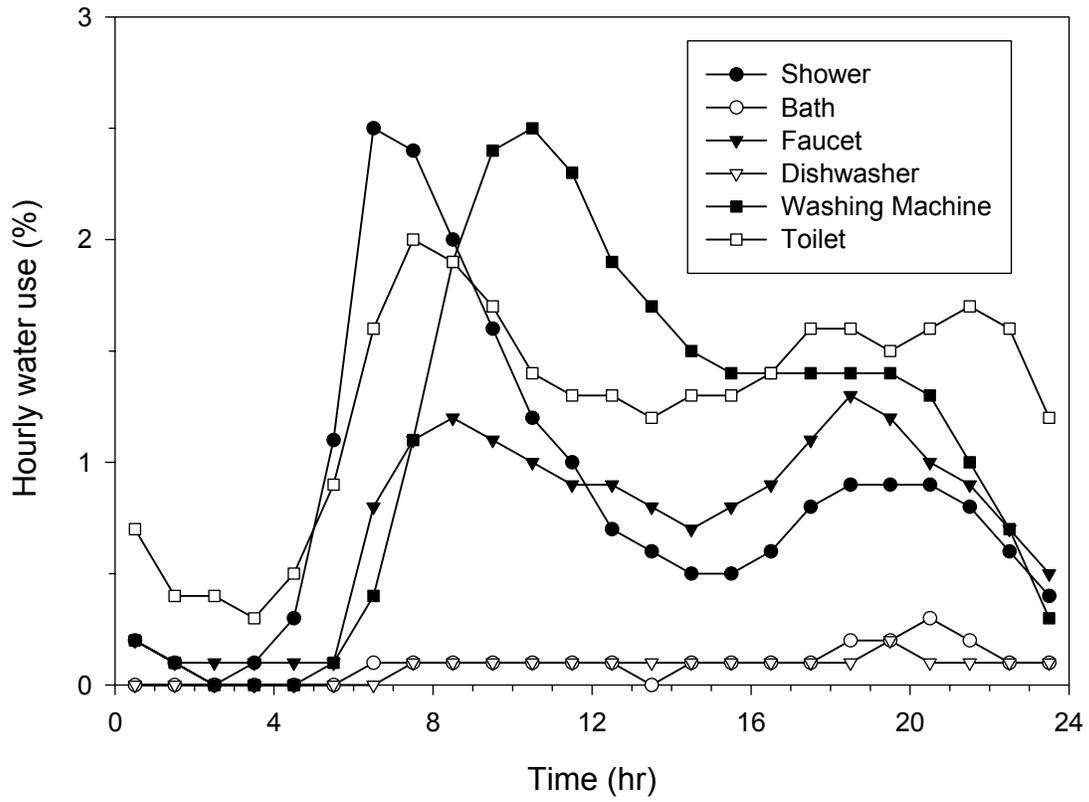


Figure S.5 Hourly water use for each device from the data of Mayer et al. (1998)