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Trihalomethane exposures in indoor swimming pools: A level III fugacity model

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ABSTRACT

The potential for generation of disinfection byproducts (DBPs) in swimming pools is high due to the concentrations of chlorine required to maintain adequate disinfection, and the presence of organics introduced by the swimmers. Health Canada set guidelines for trihalomethanes (THMs) in drinking water; however, no such guideline exists for swimming pool waters. Exposure occurs through ingestion, inhalation and dermal contact in swimming pools. In this research, a multimedia model is developed to evaluate exposure concentrations of THMs in the air and water of an indoor swimming pool. THM water concentration data were obtained from 15 indoor swimming pool facilities in Quebec (Canada). A level III fugacity model is used to estimate inhalation, dermal contact and ingestion exposure doses. The results of the proposed model will be useful to perform a human health risk assessment and develop risk management strategies including developing health-based guidelines for disinfection practices and the design of ventilation system for indoor swimming pools.

1. Introduction

In Canada, swimming is a popular activity for leisure and exercise and is ranked fourth among leisure activities, after walking, gardening and home exercise. Many of the swimmers using indoor public pools are children, pregnant women and seniors who may be at greater risk for health effects from chemical exposures in swimming pool water; therefore, it is important to quantify the associated exposure and risk.

The objective of this paper is to develop an integrated model to evaluate exposure concentrations of trihalomethanes (THMs) in the air and water of an indoor swimming pool facility (natatorium). The fugacity approach is used to develop a multimedia environmental exposure model to assess the inhalation and dermal contact exposures, and minor ingestion exposure. The results of the proposed model will be useful to perform a human health risk assessment and develop risk management strategies including the development of health-based guidelines for disinfection practices and the design of ventilation system for swimming pools.

1.1. Disinfection byproducts (DBPs)

Chlorine has been used to disinfect drinking water for over a century and has helped eliminate most waterborne diseases in developed countries, such as typhoid, and cholera. Approximately 90% of the water supply systems in Canada use chlorine for disinfection purposes (Health Canada, 2009). During the disinfection process, reactions between natural organic matter in the source water and chlorine added to the

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water generate disinfection byproducts (DBPs), such as THMs and haloacetic acids (HAAs). Since the discovery of DBPs in 1974 (Rook, 1974), many studies have been done on their formation, prevalence and associated risks. Of the more than 600 DBPs identified to date, very few have been the subject of exposure and toxicological studies (Richardson et al., 2007). The DBPs that are currently most studied are THMs and HAAs in part due to the availability of exposure and toxicology studies, and in part due to the fact that THMs are present in the highest concentrations, followed by HAAs (Who, 2000). THMs consist of four distinct but related compounds: chloroform ($CHCl_3$), bromodichloromethane ($CHCl_2Br$), dibromochloromethane ($CHClBr_2$), and bromoform ($CHBr_3$). For the purposes of testing and regulation, the concentrations of these four compounds are often added to form a parameter commonly referred to as total trihalomethanes (TTHMs).

The presence of DBPs in water has been linked to an increased risk of bladder cancer (Villanueva et al., 2007), reproductive effects (Nieuwenhuijsen et al., 2000), and immediate respiratory effects such as asthma (Levesque et al., 2006; Goodman and Hays, 2008; Jacobs et al., 2007; Thickett et al., 2002). Health Canada (2008) has set guidelines for some groups of DBPs in drinking water: namely, TTHMs (0.10 mg/L), HAAs (0.08 mg/L), bromate (0.01 mg/L), bromodichloromethane (0.016 mg/L) and chlorite (1 mg/L), however, no such guideline is in use for swimming pool waters.

1.2. DBPs formation in swimming pools

Disinfection in swimming pools is important to reduce the risk of exposure to pathogens present in the water that originate either from the source water or from the swimmers in the water. Viruses, bacteria, parasites and fungus present in the water can cause health effects and even death (WHO, 2006).

Chlorine is the most common disinfectant used for indoor pools (WHO, 2006). Chlorine is added to the pool waters as chlorine gas, calcium/sodium hypochlorite, or through electrolytic generation of sodium hypochlorite. Regardless of the method of application, once the chlorine is in the water it forms hypochlorous acid which dissociates into hydrogen atoms and hypochlorite ions. The "free chlorine" residual is the sum of the concentrations of hypochlorous acid, (HOCl), hypochlorite ion (OCl^-), and aqueous chlorine ($Cl_{2(aq)}$) (Weaver et al., 2009). The residual chlorine is measured frequently to ensure adequate disinfection power is retained in the pool throughout operation. Regulations in Quebec require free chorine in swimming pools to be between 0.8 and 2.0 mg/L (Ministère du Développement Durable, de l'Environnement et des Parcs (MDDEP), 2006). In British Columbia (BC), the required amount of free chlorine varies with pH: 0.5 mg/L for pH of 7.4–7.8, and 1.0 mg/L for pH of greater than 7.8 (BC, 2010). Other parameters that are measured and regulated in swimming pools include fecal coliforms, *Escherichia coli, Pseudomonas aeruginosa, Staphylococcus aureus*, alkalinity, hardness, pH and turbidity.

The formation of other DBPs is dependent on the presence of precursors which are usually naturally occurring organic material. A recent investigation in swimming pools in Spain identified more than 100 different DBPs in swimming pools, including brominated DBPs which are generally more mutagenic and carcinogenic than chlorinated DBPs (Richardson et al., 2007). The potential for DBP formation in swimming pools is high due to the concentrations of chlorine required for disinfection and the presence of organic matter from swimmers in the form of hair products, lotion, mucus, skin excretions and urine (Kim et al., 2002).

1.3. Exposure to DBPs

In a swimming pool, although children may ingest a small amount of pool water, exposure to DBPs is mainly through inhalation and dermal contact (Whitaker et al., 2003). The amount and impact of the exposure may be influenced by many factors including: the type and dose of disinfectant, the resultant concentration of DBPs in the water, the number of swimmers, temperature of water and air (Chu and Nieuwenhuijsen, 2002), ventilation rates in the building, duration of swimming, and the turbulence in the water generated by moving-water features such as fountains, water slides, wave pools, and hot tub jets (Hery et al., 1995). For chlorination THMs, inhalation may constitute a significant impact due to the volatility of the compounds. In this study, exposures to THMs through dermal contact, ingestion and inhalation are modeled using concentrations of THMs detected in water samples collected from indoor pools in Quebec.

2. Data collection

The concentration data used to model environmental exposures were obtained from a study by Simard (2008). Pools were selected in various areas of Quebec City served by several different potable water distribution systems. The 15 indoor pools chosen were those most attended in each area with similar disinfection practices (chlorination). Indoor pools are considered due to the comparative ease of modeling the air exchange in the environment to account for volatilization of DBPs and also due to the presumed increased risk from the enclosed indoor pool environment. Samples were collected from each pool once per month over a year (Jan. 2007–Feb. 2008).

Water samples were stored in accordance with the protocol suggested by MDDEP (2009). Analysis for chloroform, BDCM, DBCM and bromoform was performed by gas chromatograph with a mass spectrophotometry column in compliance with US EPA methods 524.2 and 552.2 (US EPA, 1995a,b). The risk analysis software @Risk (Palisade, 2010) and the Excel add-in EasyFitXL Version 5.3 (MathWave Technologies, 2010) were used to fit probability density functions to the results of the analysis of the water samples. Lognormal distribution was found to be the best fitted distribution in all cases. A summary of the results of the laboratory analyses is presented in Table 1 along with the means and standard deviations of the lognormal distributions. These distributions were used as input into the Monte Carlo simulations in the model to represent the concentrations in the water in the pools in Quebec as well as the variability in that data.

3. Fugacity model

The concept of fugacity is used for describing the conditions of equilibrium among multimedia environments. Fugacity is

Table 1 – Results of laboratory analyses.

Analyte	Detection limit μg/L	Range μg/L	# of samples (out of 176) below detection limit	Mean (μ)	Standard deviation (σ)
Chloroform	0.3	12.93–215	0	55.2	31.6
BDCM	0.4	0–23.94	11	1.23	2.55
DBCM	0.4	0–27.13	142	0.26	1.94
Bromoform	0.5	0–19.23	173	0.26	1.41
Temperature	–	26.5–31.1	NA	28.2	0.88

NA – not applicable.

defined as *the escaping tendency of a chemical to leave one medium in preference for another*. At the low concentrations expected for environmental sampling, there is a linear relationship between concentration, fugacity (f) and fugacity capacity (Z):

$$C = fZ \quad (1)$$

The mass balance equations used in fugacity modeling can include terms that correspond to chemical and biochemical reactions, inter-media transport, diffusion between media, and advection in or out of an "evaluative environment". The use of fugacity makes it possible to consider complex inter-relationships between environmental media such as air, water, and soil, as well as sediment underlying water, suspended solids within the water, and aerosols or particulates within the air. Biota in the form of fish, plants or humans can also be included as media (Mackay, 2001).

Fugacity offers advantages over the use of dual-phase partition coefficients when there are several media and many processes occurring. Traditional partition coefficients can only describe equilibrium conditions between two media at a time, while fugacity models can generate equations to consider all the media at the same time. Four levels of fugacity models have been proposed (Mackay, 2001). Level III deals with steady state, but includes flow and non-equilibrium conditions.

3.1. *Evaluative environment*

The first step in fugacity modeling is the definition of the evaluative environment. In the case of an indoor swimming pool, the environment consists of the water in the pool, the air above the pool within the natatorium, and the people in the pool (biota).

During the regular operation of most community swimming pools, water is filtered and chlorinated during recirculation of the water from the pool (BC, 2010). Some water is lost during back-washing of the filters and by evaporation, therefore fresh water is added at a rate of approximately 1% of the total pool volume per day or 30 L per swimmer (MDDEP, 2006). Some DBPs may be adsorbed to suspended solids in the pool and removed with the filtration; however, for this study that process is neglected. The concentration of DBPs is assumed to remain constant and the processes of DBP formation due to swimmers and chlorination, as well as losses to filtration are assumed to be represented by that constant concentration of DBPs.

The other component of the environment, the air, is also re-circulated and filtered. Many ventilation systems in natatoriums are operated with the goal of maintaining constant and comfortable humidity. The American Society of Heating, Refrigerating and Air-Conditioning Engineers (ASHRAE, 2007) recommends maintaining the humidity between 40 and 60% and the air temperature 2–4 °C higher than the temperature of the water. For energy efficiency and protection of structural elements, the humidity is removed from the air and a large portion of the air is returned to the natatorium. It is unclear what effect dehumidifying the air has on the concentration of DBPs in the air.

In Quebec, the recommended rate of ventilation using outside air is 9 $m^3/h\ m^2$, based on the area of the pool surface and surrounding deck. The total ventilation including re-circulated and outside air should equal 4–6 air changes per hour (ACH). For the "typical" pool facility considered here, 10–15% of the air entering the natatorium is outside air and the rest is re-circulated. For the purposes of modeling, the outside air is assumed to have negligible concentrations of DBPs, which would be the case, provided the exhaust and air intake have sufficient separation between them.

A previous study has shown that inhalation of DBPs in aerosols contributes significantly to the average daily dose; however, that study focussed on HAAs which are less volatile than THMs (Xu and Weisel, 2003). In the present study we assumed that THMs are sufficiently volatile to not exist in any appreciable concentrations within aerosols of respirable size (<10 μm). Another assumption made in this study is that the air in the natatorium is completely and instantaneously mixed. A degree of variation of concentration and temperature with height in natatoriums has been shown (Hsu et al., 2009); however, considering such variation increases the complexity of the model. A schematic of the evaluative environment is presented in Fig. 1. The compartments are

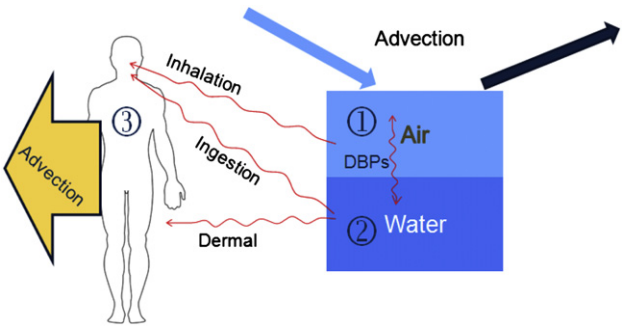

Fig. 1 – System schematic of with flows and exposure routes. Evaluative environment used in the fugacity model showing model compartments, human exposure routes, inter-media transport and advective transport processes.

numbered as used in the model. The inter-media transport, human exposure routes, and advection processes are also highlighted. Depending on the amount absorbed, inhaled and ingested by the humans, the removal of contaminants when the people leave the pool can also be considered an advection process.

For the typical pool in Quebec considered in this study, the size of the pool and natatorium is assumed from the dimensions of the pools that were sampled (where they were known) and other similar pools. The size and recirculation parameters of the typical pool considered in the model are presented in Table 2.

3.2. Physico-chemical properties

Once the evaluative environment has been described, the chemicals of concern must be characterized in order to model their behavior in the environment. Chemical properties of THMs were determined from chemical handbooks and literature (Mackay, 2006). Where several values were presented for a chemical property, the multiple data points were fitted to a probability density function using the decision support software @Risk. The inclusion of multiple data points for these parameters reflects a degree of uncertainty in some of the chemical properties that were used as input into the model. As such, it was desirable to propagate the uncertainty in each of these parameters through the model using Monte Carlo simulations so that the final model results reflected the uncertainties in all of the input parameters. Some of the chemical properties were the same in all sources, and in that case deterministic (or "crisp") values were used as model input. The chemical properties are presented in Table 3, with the properties for which distributions were used shown in shading.

The parameter Z, the fugacity capacity constant, has units mol/m^3 Pa. Each compartment of the natatorium environment has a fugacity capacity that is determined by the chemical properties of the contaminant. The equations for Z are shown in Table 3.

In their paper on unified dermal uptake model, McKone and Howd (1992) present a unitless partition coefficient between water and skin, K_M. The values of K_M are related to the octanol–water partition coefficient, K_{OW}, by the empirically-derived equation in the notes below Table 3. The calculated fugacity capacities and parameters are also presented in Table 3.

3.3. Removal from the environment

In a typical environmental fugacity model, removal of contaminants from the environment can occur in various ways including reactions, diffusion, and advection. In the case of a swimming pool, it is assumed that the predominant reaction occurring is the formation of the DBPs, which is already incorporated in the constant water concentration. The advective processes include the flow of the re-circulating air and water, and the movement of people in and out of the pool. The maximum allowable capacity calculated for our assumed pool size is 417 bathers (MDDEP, 2006); however, the maximum number of swimmers observed in any of the pools during the sampling was 100. The number of swims per month and minute per month spent swimming vary by age. Data from the US EPA Exposure Factors Handbook (US EPA, 2009) was fitted to distributions for calculating the average minutes per swim in order to vary the duration of exposure by age.

For simplicity in the fugacity model, 1 h duration was used as an average time of swimming as recommended by US EPA (2009). This is done so that the flow of people in and out of the pool is simplified for all age groups. Therefore we can calculate that every hour 100 people leave the pool with THMs absorbed into their skin. The ages of the 100 people were distributed according to their minutes swimming per month. The volume of the people in the pool was calculated using age specific exposure factors (US EPA, 2009) and a relationship between height, surface area and weight (Sendrov and Collison, 1966). The volume and surface area were adjusted for the assumption that people do not have their head submerged (Xu and Weisel, 2005).

Table 2 – Evaluative environment parameters for Quebec pools and swimming pool facility characteristics in Modena, Italy (Fantuzzi et al., 2001).

Swimming pool	Pool surface area (m^2)	Pool volume (m^3)	Natatorium area (m^2)	Air volume of natatorium (m^3)	Ventilation rate (ACH)	Maximum # swimmers during sampling
Quebec typical pool[a]	500[b]	1200[c]	770	9390[d]	4–6[e]	417[f]
Modena 1	312	420	700	3500	6	16
Modena 2	312 + 42[g]	500 + 30[g]	600	4200	5	27
Modena 3	300	420	450	3375	5	3
Modena 4	312	670	440	3000	6	20
Modena 5	250	565	525	3150	5	13

a "half olympic sized pool" assumed to represent Quebec pools.
b 25 m × 20 m.
c based on 2.4 m depth.
d based in 12.2 m (40 ft) ceiling.
e uniform distribution used.
f maximum bather load based on 1.2 m^2 per bather (Ministère du Développement Durable, de l'Environnement et des Parcs, 2006).
g additional dimensions given for wading pool in the same natatorium.

Table 3 – Estimates for fugacity capacities (after Mackay et al., 1985).

Medium (compartment number)	Air (1)	Water (2)		Skin (3)	
	$Z_1 = 1/RT$ (mol/Pa m^3)	H (Pa m^3/mol)	$Z_2 = 1/H$ or C^S/P^S (mol/Pa m^3)	K_M (unitless)	$Z_3 = K_M/H$ (mol/Pa m^3)
Chloroform	4.04E-04	390.87	2.56E-03	10.053	0.026
BDCM	4.04E-04	242.8	4.12E-03	12.606	0.052
DBCM	4.04E-04	119.60	8.36E-03	16.126	0.135
Bromoform	4.04E-04	59.56	1.68E-02	20.682	0.347

Notes: R = 8.314 Pa m^3/mol K; T = absolute temperature (298 K); H = Henry's Law Constant (Pa m^3/mol); C^S = aqueous solubility (mol/m^3); P^S = vapor pressure (Pa); K_M = skin water partition coefficient = $0.64 + 0.25(K_{OW})^{0.8}$.

Based on the recommendations for operation of pool facilities made by ASHRAE (2007) the air in the natatorium is re-circulated at a rate of 56,340 m^3/h. The percentage of re-circulated air that is replaced with fresh air was calculated using 9 m^3 of fresh air per m^2 of surface area of the room. The flow of re-circulated water is disregarded because the concentration of THMs in the water is held constant. The movement of the people in and out of the pool is assumed to be 4.13 m^3/h. These unconventional units are useful for calculating inter-media and advective flows in the model.

3.4. *Inter-media transport*

Movement of THMs between media is governed by diffusive and mass transport processes. The net transfer rate is described by the following equation:

$$N = D_{ij} \times f_i - D_{ji} \times f_j \left(\frac{\text{mol}}{\text{h}}\right) \quad (2)$$

where D_{ij} is the transfer from medium i to medium j and D_{ji} is the transfer from j to i. The transport coefficient is generally described by:

$$D = k \times A \times Z \left(\frac{\text{mol}}{\text{Pa}} \cdot \text{h}\right). \quad (3)$$

where k is the mass transfer coefficient and A is the inter-phase area.

3.4.1. *Air and water*

The mass transport between the air and the water was calculated using following relationship:

$$D_{12} = \frac{1}{\frac{1}{k_G A_{12} Z_1} + \frac{1}{k_L A_{12} Z_2}} \quad (4)$$

where k_G is the air side mass transfer coefficient, k_L is the water side mass transfer coefficient, and A_{12} is the area over which the air and water are in contact. This relationship is based on the two-resistance theory presented by Mackay and Yeun (1983).

The mass transfer coefficients, k, are physico-chemical properties that were generated based on formulas provided by Guo and Roache (2003). The air side mass transfer coefficient k_G was calculated using seven formulas from the following sources:

1. Bennett and Myers (1982), Sparks et al. (1996),
2. Higbie (1935), Reinke and Brosseau (1997),
3. Mackay and Matsugu (1973), Reinke and Brosseau (1997),
4. Geankoplis (1993), Reinke and Brosseau (1997),
5. Jayjock (1994), van Veen et al. (1999),
6. Sparks et al. (1996) and
7. Reinke and Brosseau (1997), Geankoplis (1993).

Using the distribution fitting function of the decision support software @Risk, the results were fitted to a lognormal distribution to be later used in performing Monte Carlo simulations. The water side mass transfer coefficient, k_L, was determined using an equation from Southworth (1979) provided by Guo (2002) and Guo and Roache (2003). The transfer coefficients k_G, k_L, and k_p are presented in Table 4. The probability distributions and their characteristic parameters are also provided where applicable.

3.4.2. *Water and skin*

The transport coefficient for water and skin, D_{23}, is described by:

$$D_{23} = K_p \times A_{23} \times Z_3 \quad (5)$$

where K_p is the diffusion coefficient for skin and A_{23} is the surface area of the skin in contact with the water (McKone and Howd, 1992). The diffusion of the chemical into skin is also a two-resistance model, with the permeability coefficient, K_p.

Table 4 – Mass transfer coefficients.

Transfer coefficient (m/h)	Distribution parameters	Chloroform	BDCM	DBCM	Bromoform
k_G	Lognormal mean (std dev)	46.25 (20.70)	44.87 (20.71)	43.93 (20.78)	43.21 (20.98)
k_L	Deterministic	0.399	0.341	0.302	0.274
k_p	Deterministic	0.0296	0.0311	0.0349	0.0402

3.5. Mass balance

Mass balance equations were generated for each compartment. The equations were of the general form:

$$E_i + G_i C_{Bi} = G_i C_i + \sum D_{ij} f_i - \sum D_{ji} f_j \quad (6)$$

where

E_i = direct emission to the compartment,
$G_i C_{Bi}$ = advective flow into the compartment, in this case the air re-circulated back into the natatorium with some of it removed and replaced with clean air,
$G_i C_i$ = advective flow out of the compartment,
$\Sigma D_{ij} f_i$ and $\Sigma D_{ji} f_j$ = inter-media transport from compartment i to compartment j, and from compartment j to compartment i, respectively.

The concentration of chemicals in the water, C_2, is known and the concentrations C_1 in the air, and C_3 in the skin can be expressed as $C_i = Z_i \cdot f_i$. The remaining mass balance equations are:

For air:

$$f_1 = \frac{\frac{D_{12} \times C_2}{Z_2}}{0.12 \times G_1 \times Z_1 + D_{12}} \quad (7)$$

For Water:

$$E_2 = D_{23} \times \left(\frac{C_2}{Z_2}\right) - D_{23} \times f_3 + D_{21} \times \left(\frac{C_2}{Z_2}\right) - D_{21} \times f_1 \quad (8)$$

For Skin:

$$f_3 = \frac{\frac{D_{32} \times C_2}{Z_2}}{G_3 \times Z_3 + D_{32}} \quad (9)$$

3.6. Model validation

The proposed model was validated using a data set provided by Fantuzzi et al. (2001). In that study, water and air concentrations of the four THMs were measured for five swimming pools located in Modena, Italy. The air and water concentrations of THMs are presented in Table 5 and the pool facility characteristics are presented in Table 2. These concentrations and pool characteristics were used as input into the fugacity model. The equations given in the previous section were used to determine the concentrations in air and skin.

The modeled air concentrations were compared to the air concentrations measured by Fantuzzi et al. (2001) as shown in Fig. 2. The normalized mean bias (NMB) and mean fractional bias (MFB), shown in Table 5, were calculated for each THM using the following equations:

$$\mathrm{NMB}(\%) = \frac{\sum_{i=1}^{N}\left(Y_{\mathrm{predicted}} - Y_{\mathrm{measured}}\right)}{\sum_{i=1}^{N} Y_{\mathrm{measured}}} \times 100 \quad (10)$$

$$\mathrm{MFB}(\%) = \frac{1}{N}\sum_{i=1}^{N} \frac{2 \times \left(Y_{\mathrm{predicted}} - Y_{\mathrm{measured}}\right)}{\left(Y_{\mathrm{predicted}} + Y_{\mathrm{measured}}\right)} \quad (11)$$

The modeled concentrations were closest to the measured concentrations for chloroform and total trihalomethanes with NMB and MFB from 1.7% to 17%, followed by BDCM which were 28% and 43%, respectively. Bromoform was only detected in one swimming pool in the air. The poorest fit was for the DBCM with NMB of 74% and MFB of 109%.

The air recirculation characteristics used in the model for the Italian pools are presented in Table 2. The Italian guidelines for swimming pool facilities (Conferenza Permanente per i Rapporti tra lo Stato, 2003; Fantuzzi, 2011 personal communication) suggest air recirculation which results in air

Table 5 – THM concentrations in pools in Modena, Italy (Fantuzzi et al., 2001) and comparison of measured and modeled air concentrations for model validation.

			Modena 1	Modena 2	Modena 3	Modena 4	Modena 5	NMB	MFB
Chloroform	Water (μg/L)		26	47	18.7	68.4	6.1		
	Air (μg/m^3)	Measured	58.6	42.00	67.7	43	19		
		Modeled	87.67 (37.81)	40.15 (15.83)	50.78 (18.11)	73.25 (29.07)	16.86 (6.33)	16.68	9.36
BDCM	Water (μg/L)		5.3	4.3	4.5	2	5		
	Air (μg/m^3)	Measured	13	4.80	14.7	2.9	8		
		Modeled	11.17 (4.79)	2.29 (0.88)	7.62 (2.62)	1.35 (0.53)	8.67 (3.19)	−28.34	−42.86
DBCM	Water (μg/L)		0.6	1.3	2	0.4	1.3		
	Air (μg/m^3)	Measured	3.5	1.20	4.3	0.3	6		
		Modeled	0.64 (0.3)	0.35 (0.14)	1.71 (0.63)	0.14 (0.06)	1.14 (0.46)	−73.99	−108.58
Bromoform	Water (μg/L)		0.1	0.1	0.3	-nd	1.3		
	Air (μg/m^3)	Measured	nd	nd	nd	nd	0.8		
		Modeled	nd	nd	nd	nd	0.58 (0.25)		
TTHM	Water (μg/L)		32	52.7	25.5	70.8	17.8		
	Air (μg/m^3)	Measured	75.1	48.00	86.7	46.2	33		
		Modeled	99.54 (38.15)	42.80 (15.86)	60.24 (18.32)	74.74 (29.07)	27.25 (7.08)	5.39	1.73

Values in parentheses are standard deviations.
nd – not detected.
NMB – normalized mean bias.
MFB – mean fractional bias.

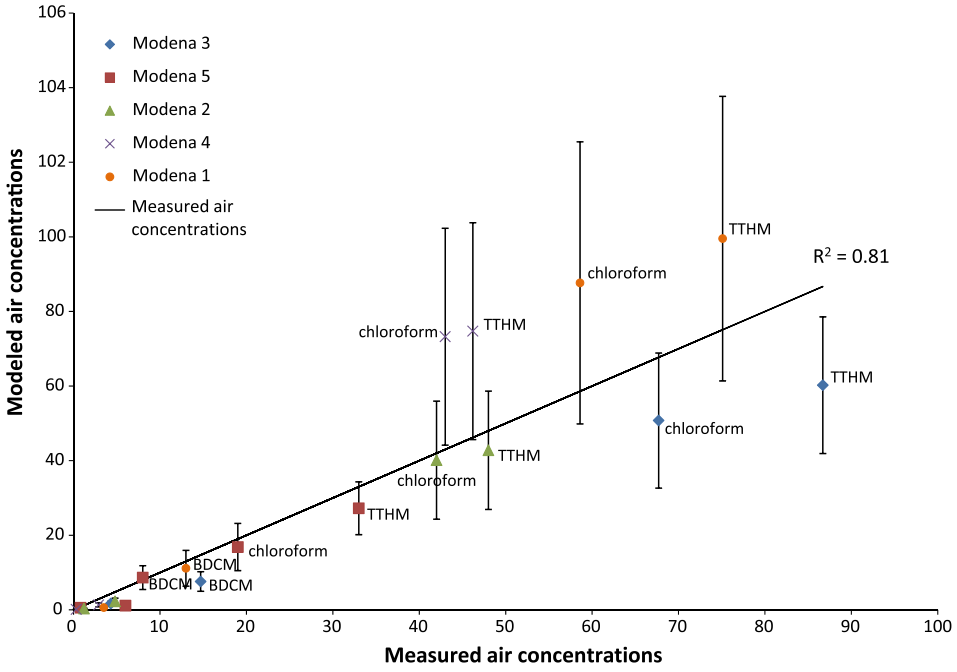

Fig. 2 – Comparison of measured air concentration to modeled air concentration for model validation.

velocities of no more than 0.10 m/s with an outdoor air component of at least 20 m^3/h for each m^2 of swimming pool surface area. The number of swimmers considered in the model for each facility were the maximum number of bathers reported by Fantuzzi (personal communication, 2011) during the sampling event. For pools 2 and 4, the model fit was poor when using the default assumed values for air recirculation. The model was re-run using 100% outside air for these pools, resulting in a better fit. In general, the fit for these two swimming pools was not as favorable. In the case of pool 2, there were two swimming pool basins in the natatorium, which would make it more difficult for the model to predict the air concentrations. The poor fit for pool 4 could not be completely explained; therefore, further analysis would be required to investigate how to best fit the model to various swimming pool and natatorium configurations. The ratio of air concentrations to water concentrations for these pools was lower in those two pools than the other three by a factor of 2–5, suggesting that the conditions specific to those two pools make the air concentrations much lower than the other three pools.

3.7. *Results and discussion on fugacity modeling for Quebec pools*

To evaluate the exposure doses for the swimming pools in Quebec, Equations (6) through (9) were used to calculate the fugacities and to estimate the concentrations, amount, and % in each compartment, presented in Table 6. Because the concentration in the water was known, there was an unknown term E_2, which represented the formation of the THMs. This number should be equal to the total amount of THMs leaving with the people, and vented from the exhaust fans.

The fugacities, concentrations, mass, and advective and interphase transports for each compound are presented in Fig. 3.

The partitioning of the THMs between the water and the air is consistent with other studies in which concentrations of chloroform in water and air were measured. In Table 7, the results of the model are compared to empirical data from the literature. Chloroform is presented because it is the most commonly measured THM in the literature. In Fig. 4, a plot of measured chloroform concentrations in water and air are

Table 6 – Fugacities, concentrations and emissions.

Compound	Fugacity in Pa			C_1 µg/m³	C_2 µg/L	C_3 µg/m³	E_2 g/h
	f_1	f_2	f_3				
Chloroform	4.83E-03	1.81E-01	1.64E-03	232.6	55.2	5266	1.64
BDCM	4.98E-05	1.82E-03	1.75E-05	3.29	1.23	148.0	2.32E-02
DBCM	4.12E-06	1.46E-04	1.56E-06	0.35	0.26	44.40	2.62E-03
Bromoform	1.78E-06	6.05E-05	7.45E-07	0.18	0.26	66.17	1.54E-03

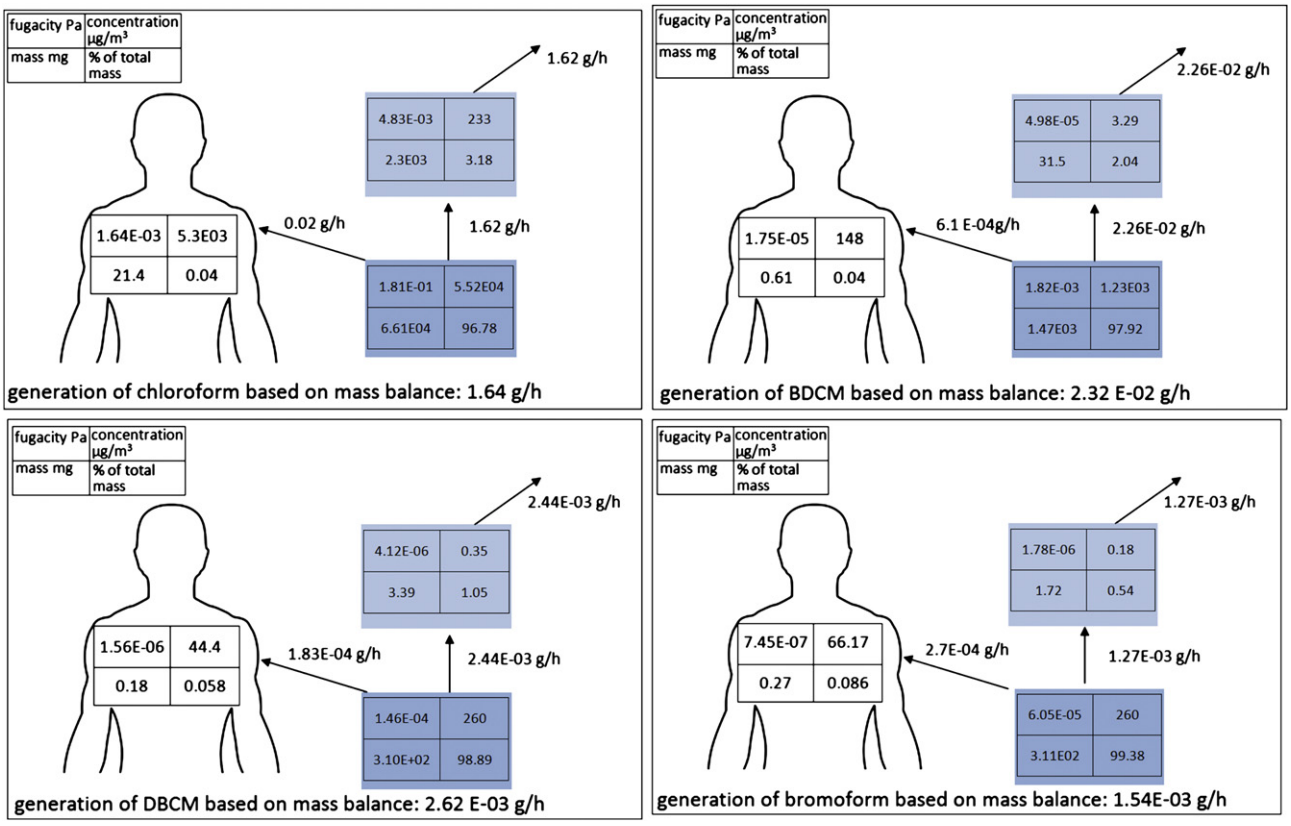

Fig. 3 – Fugacity model results for the four THM species. Visual representation of the model results showing concentration, fugacity, amount (in grams) present and percent of total amount for each compartment of the model, and for each of the four THMs considered.

Table 7 – Comparison of literature with model results (concentrations in water and air).

Data source	Pool water µg/L	Pool air µg/m^3	Data source	Pool water µg/L	Pool air µg/m^3
Aggazzotti et al. (1990)	32.75 (10.76)	209.52 (129.6)	Erdinger et al. (2004)	17.5 (3.81)	195 (25.1)
Aggazzotti et al. (1993)	36.59 (18.24)	138.68 (78.45)	Fantuzzi et al. (2001)	39.8 (21.7)	58 (22.1)
Aggazzotti et al. (1995)	89.69 (31.24)	261.74 (110.8)		33.2 (24.6)	46.1 (18.6)
	44.59 (18.03)	310.33 (134.0)		4.2 (1.3)	8.7 (5.1)
	57.44 (11.03)	120.52 (38.03)		1.9 (2)	3.1 (2.3)
	99.79 (31.85)	376.54 (107.19)	Jo (1994)	23.9 (6.6)	50.9 (2.2)
	36.5 (5.10)	96 (17.6)		19.5 (7.5)	33.6 (12.8)
	78.44 (6.95)	253.78 (56.65)	Levesque et al. (1994)	158.6 (7.5)	507 (45.8)
	19.5 (2.65)	48 (1.63)		200 (10.7)	1490 (438.7)
	114.5 (12.45)	459.5 (160.6)		307.1 (16)	1120 (157.1)
	47.5 (1.02)	302.5 (88.8)		567.5 (5.0)	1296 (36.5)
	98.78 (5.92)	387.83 (124.7)		538.3 (21.4)	1630 (196.4)
Aggazzotti et al. (1998)	33.7 (9.6)	91.7 (15.4)	Levesque et al. (2000)	37.79 (21.02)	208.59 (91.12)
	2.3 (0.60)	10.5 (3.1)	Model results using Simard (2008) data	**55.20**	**233**
	0.8 (0.2)	5.2 (1.5)	Model results using simulated	65	275
Cammann and Hubner (1995)	11.38 (1.17)	12.285 (2.61)	water concentrations	80	338
	10.12 (4.09)	42.8 (11.95)		100	423
	23.6 (2.42)	93.85 (8.75)		150	634

Values given are the mean, values in parentheses are standard deviation, model results in bold.

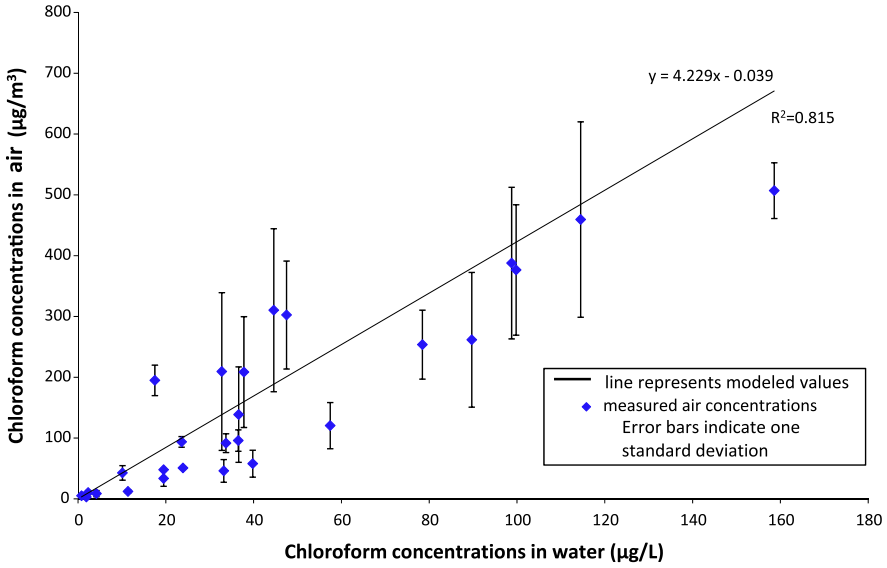

Fig. 4 – Chloroform air and water concentrations from model and previous studies. Chart showing the relationship between measured concentrations of chloroform in air and water in previous studies, and the model results of air concentrations for given water concentrations.

presented with a line representing the modeledair concentrations.

The known water concentrations of THMs and modelled air concentrations can now be used in exposure assessment to determine the exposure doses for swimmers through dermal contact, inhalation and ingestion.

4. Exposure assessment

In a swimming pool, the exposure routes for THMs are dermal contact, inhalation and ingestion. The exposures vary for different age groups based on average time swimming, body surface area, inhalation rate and rate of inadvertent ingestion of the pool water. The exposure factors used below are listed in Table 8.

In the equations for each exposure route below, some common terms are:

EF = exposure frequency (min/month) × 12 months/year
ED = exposure duration (years) – to calculate chronic daily intake (CDI)
BW = body weight (kg)
AT = averaging time (yr) – for chronic only (70 yrs)

4.1. *Inhalation*

The dose of inhaled THMs was calculated using the following equation:

$$\text{Dose}\left(\frac{\mu g}{kg \cdot d}\right) = \frac{C_a \times IR_A \times EF \times ED}{BW \times AT \times 365\left(\frac{\text{days}}{\text{year}}\right)} \quad (12)$$

where

Table 8 – Exposure factors (US EPA, 2009).

Exposure factor		Age				
		1–4	5–11	12–17	18–64	>65
Body weights (kg)	Mean	14.53	32.03	56.56	80	74.49
	Standard deviation	2.41	11.2	16.58	20.15	16.16
Time spent swimming (min/month)	Mean	81.53	68.76	87.41	51.59	53.67
	Standard deviation	133.1	54.59	83.4	47.54	59.45
Respiration rates (m^3/minute)	Mean	2.11E-02	2.13E-02	2.54E-02	2.72E-02	2.56E-02
	Standard deviation	4.55E-03	4.82E-03	6.46E-03	7.25E-03	5.00E-03
Surface area of whole body (m^2)	Mean	0.63	1.09	1.59	1.95	1.88
	Standard deviation	0.07	0.22	0.27	0.28	0.24
Surface area of head (m^2)	Mean	0.085	0.109	0.111	0.116	0.115
	Standard deviation	0.0017	0.0042	0.0039	0.0035	0.0031
Surface area of body, no head (m^2)	Mean	0.545	0.98	1.475	1.836	1.768
	Standard deviation	0.069	0.21	0.27	0.28	0.24
Water ingestion (mg/L)		49	49	49	21	21

C_a = concentration of contaminant in air (μg/m^3)
IR_A = receptor air intake rate (m^3/min)

Lognormal distributions of the respiration rates, body weight, and minutes per month spent swimming for bathers of different ages were determined from the US EPA Exposure Factors Handbook (US EPA, 2009).

4.2. Dermal contact

The dose due to dermal contact with THMs in water was calculated using the fugacity based model. The concentrations of THMs in the skin (biota compartment) in the model was multiplied by age specific volume per swimmer:

$$\text{Dose}\left(\frac{\mu g}{kg\cdot d}\right) = \frac{C_B \times V \times EF \times ED}{BW \times AT \times 365\left(\frac{\text{days}}{\text{year}}\right)} \times CF \quad (13)$$

where

C_B = concentration of contaminant in swimmers skin (μg/L)
V = body volume of the swimmer (m^3)
CF = conversion factor $\frac{10^3 L}{m^3} \times \frac{1\ h}{60\ min} = 16.7$

The surface areas and body volume for each age group were obtained from the EFH.

4.3. Ingestion

The dose of inadvertent ingestion of pool water was calculated using the equation:

$$\text{Dose}\left(\frac{\mu g}{kg\cdot d}\right) = \frac{C_W \times IR \times EF \times ED}{BW \times AT \times 365\left(\frac{\text{days}}{\text{year}}\right)} \times CF \quad (14)$$

where

C_W = concentration of contaminant in water (μg/L)
IR = ingestion rate (ml/h)
CF = conversion factor $\frac{1\ L}{10^2\ ml} \times \frac{1\ h}{60\ min} = 1.67 \times 10^{-5}$

Ingestion of pool water is a larger issue for children; however, the EFH (Dufour et al., 2006) provides ingestion rates for ages under sixteen and over 18. They were allocated to the ages in this study using one value for those under 17, and one value for those older than 18.

4.4. Results and discussion of exposure assessment

The exposure dose for each age group, each THM and each exposure route were determined using Monte Carlo simulations using the software @Risk (Palisade, 2010). The results presented in Table 9 are the daily dose in μg/kg day. The results are also illustrated in Fig. 5.

The chronic daily intake (CDI) was calculated by weighting the age appropriate dose by the number of years out of the lifetime (70 years) in the age group.

$$\text{CDI} = \left(\text{dose}_{1-4} \times \frac{4}{70}\right) + \left(\text{dose}_{5-11} \times \frac{7}{70}\right) + \left(\text{dose}_{12-17} \times \frac{6}{70}\right) + \left(\text{dose}_{18-64} \times \frac{47}{70}\right) + \left(\text{dose}_{>65} \times \frac{5}{10}\right) \quad (15)$$

The calculated CDIs for each THM are:

- chloroform 3.19E-01 μg/kg day
- BDCM 5.92E-03 μg/kg day
- DBCM 9.65E-04 μg/kg day
- bromoform 7.32E-04 μg/kg day

For the exposure doses presented in Table 9 and Fig. 5, the ingestion exposure was predominantly related to children.

Table 9 – Doses of THMs by age and exposure route (μg/kg day).

THMs	Exposure route	(1–4)		(5–11)		(12–17)		(18–64)		>65	
		μ	σ	μ	σ	μ	σ	μ	σ	μ	σ
Chloroform	Inhalation	9.62E-01	2.09E+00	4.33E-01	5.52E-01	3.23E-01	4.84E-01	1.43E-01	2.16E-01	1.47E-01	2.04E-01
	Dermal contact	2.11E-01	4.86E-01	1.69E-01	2.34E-01	2.24E-01	3.49E-01	1.39E-01	2.11E-01	1.42E-01	2.00E-01
	Ingestion	8.74E-03	1.64E-02	3.91E-03	4.22E-03	2.45E-03	3.13E-03	4.35E-04	5.60E-04	4.78E-04	5.95E-04
	Total	1.18E+00	2.51E+00	5.76E-01	6.90E-01	4.52E-01	6.33E-01	2.08E-01	2.92E-01	2.16E-01	2.82E-01
BDCM	Inhalation	1.32E-02	4.97E-02	6.15E-03	2.26E-02	4.38E-03	1.19E-02	1.92E-03	5.84E-03	2.08E-03	5.86E-03
	Dermal contact	5.89E-03	1.99E-02	4.82E-03	1.30E-02	6.19E-03	1.63E-02	3.76E-03	9.66E-03	4.08E-03	1.16E-02
	Ingestion	1.91E-04	6.18E-04	8.82E-05	2.39E-04	5.32E-05	1.43E-04	9.33E-06	2.49E-05	1.07E-05	2.89E-05
	Total	1.92E-02	6.89E-02	1.22E-02	3.10E-02	7.92E-03	1.98E-02	3.95E-03	9.67E-03	3.81E-03	1.04E-02
DBCM	Inhalation	1.39E-03	1.46E-02	6.45E-04	5.68E-03	6.62E-04	2.17E-02	1.96E-04	1.31E-03	2.16E-04	1.41E-03
	Dermal contact	1.88E-03	2.46E-02	1.35E-03	7.33E-03	2.21E-03	4.00E-02	1.12E-03	6.29E-03	1.16E-03	6.39E-03
	Ingestion	4.28E-05	5.89E-04	1.79E-05	1.14E-04	1.40E-05	3.28E-04	1.88E-06	1.01E-05	2.21E-06	1.30E-05
	Total	3.31E-03	3.93E-02	1.86E-03	9.93E-03	2.07E-03	4.48E-02	5.49E-04	3.07E-03	6.08E-04	3.57E-03
Bromoform	Inhalation	6.81E-04	4.34E-03	3.13E-04	1.69E-03	2.18E-04	1.01E-03	1.07E-04	8.25E-04	1.16E-04	6.80E-04
	Dermal contact	2.51E-03	1.52E-02	2.03E-03	1.19E-02	2.67E-03	1.61E-02	1.74E-03	1.37E-02	1.86E-03	1.10E-02
	Ingestion	3.94E-05	2.50E-04	1.80E-05	1.08E-04	1.07E-05	5.89E-05	2.13E-06	2.21E-05	2.38E-06	1.47E-05
	Total	1.77E-03	1.07E-02	1.62E-03	6.57E-03	9.53E-04	5.16E-03	5.17E-04	4.72E-03	5.58E-04	3.33E-03

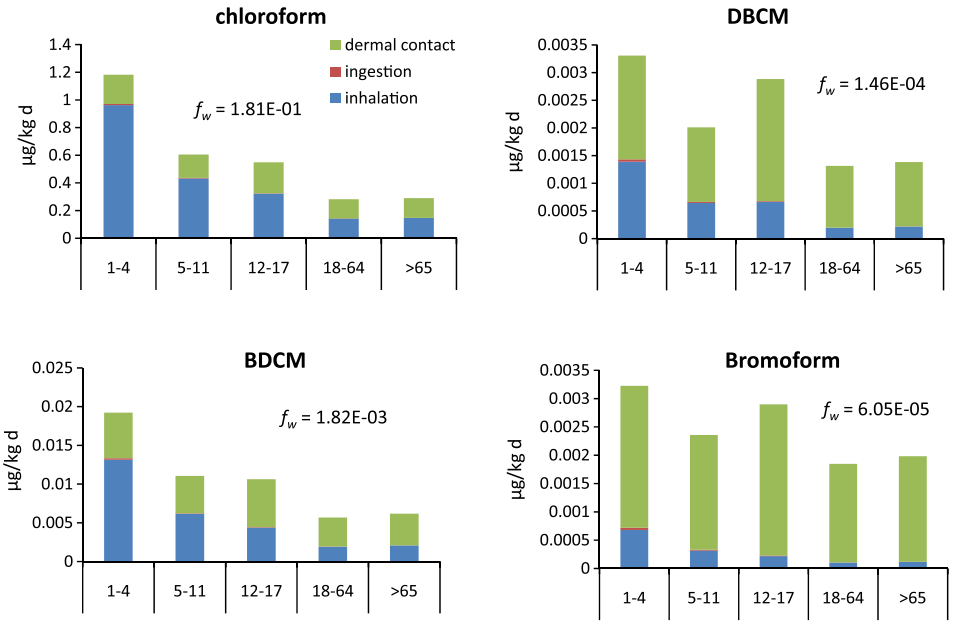

Fig. 5 – THM daily dose exposure by route and age. Graphical representation of exposure doses for each of the four THMs indicating total dose, dose for each exposure route, and exposures for each age group.

The contribution of inhalation, ingestion and dermal contact to the total dose changed for each compound as shown in Fig. 5. Note that the inhalation contribution decreases with decreasing water compartment fugacity (f_w).

The proportion of total dose attributable to each exposure route is inconsistent among previous studies. Lindstrom et al. (1997) estimated 80% of total exposure resulting from dermal contact. In studies using scuba tanks to eliminate inhalation exposure, Erdinger et al. (2004) and Levesque et al. (1994) estimated the contribution from dermal contact to be less than inhalation, 1/3 and 24% respectively. In studies on exposure during showering, Jo et al. (1990) and Weisel and Jo (1996) estimated that the exposure is roughly equal for dermal and inhalation routes. The results of this study indicate that the contribution from each exposure route changes dramatically for each age group and for the four THMs considered. Research on THMs is often generalized from studies considering chloroform only; therefore, the exposure to the other THMs is incompletely understood and requires further study.

Sensitivity analysis was conducted to find the most influential factors in the exposure doses for each age group for each exposure route, as shown in Table 10. THM concentration in water was the most important factor for all routes for all THMs except for chloroform, where minutes swimming were consistently more important than water concentration. The reason for this could be the much higher concentrations of chloroform present in water which would result in a smaller impact for variability.

4.5. *Comparison of models*

The United States Environmental Protection Agency (US EPA) Office of Pesticide Programs developed a screening level model, SWIMODEL, to assess swimmers' exposures to chemicals by inhalation, ingestion, and dermal contact routes, as well as buccal/sublingual, nasal/orbital, and aural routes (US EPA, 2003). The model was based on the worst case exposure of swimmers to trihalomethanes (Beech, 1980). The user is recommended to input the air concentration of the chemical of concern, however, the model can also calculate the air concentration from water concentration and chemical properties using either Henry's Law or Raoult's Law.

The model allows selection of swimmer age, gender and activity level (competitive or non-competitive swimming). The default exposure pathways considered by the model are inhalation, ingestion, and dermal contact with optional consideration of sublingual/buccal, nasal/orbital and aural, or any combination of these exposure routes.

Table 10 – Most sensitive factors for each exposure route.

Ingestion	Inhalation	Dermal contact
Water concentration	Water concentration	Water concentration
Minutes swimming	Minutes swimming	Minutes swimming
Body weight	k_G – inter-media transport coefficient	K_{OW} – octanol-water partition coefficient (chloroform only)
	Inhalation rate	Body surface area
	Body weight	Thickness of fully hydrated skin
	Henry's law constant (for chloroform and bromoform)	Body weight
	Air Changes per Hour (ACH)	Volume, surface area and weight of other age groups
		Height

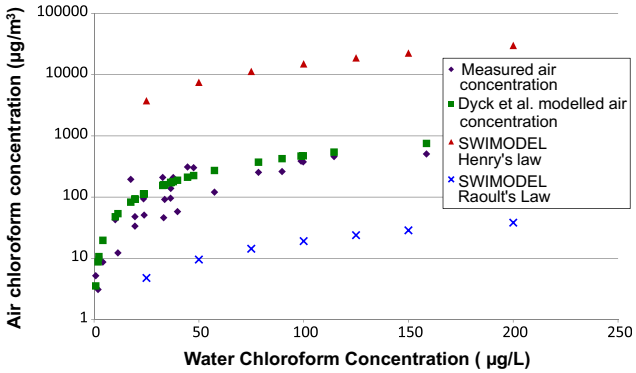

Fig. 6 – Comparison of models based on measured chloroform air concentrations.

The results of this study were compared to the results using SWIMODEL to illustrate the comparative fit to empirical data for each model. Using a range of concentrations of chloroform in water from empirical data shown in Fig. 3, SWIMODEL was used to calculate concentrations of chloroform in air. Fig. 6 shows the empirical data and modeled concentrations from this study; along with air concentrations determined using the two methods in SWIMODEL, Henry's Law and Raoult's Law. The air concentrations generated using the fugacity model appear to fit the empirical data better than the SWIMODEL results for either Henry's Law or Raoult's law. Also, the results from the two methods used by SWIMODEL differed by several orders of magnitude.

Exposure doses were calculated using SWIMODEL with default values for a male, adult non-competitive swimmer. Fig. 7 shows the percentage of the total exposure to all four THMs that is attributable to each exposure route. This figure also provides a comparison between the results of the fugacity model and SWIMODEL. One major difference between the models is the use of additional exposure routes in SWIMODEL. Obviously, the distribution between exposure routes will be different when using additional routes, however, it appears that for some compounds the alternative routes (sublingual/buccal, nasal/orbital and aural) have a far greater impact on exposure than the more commonly considered routes of

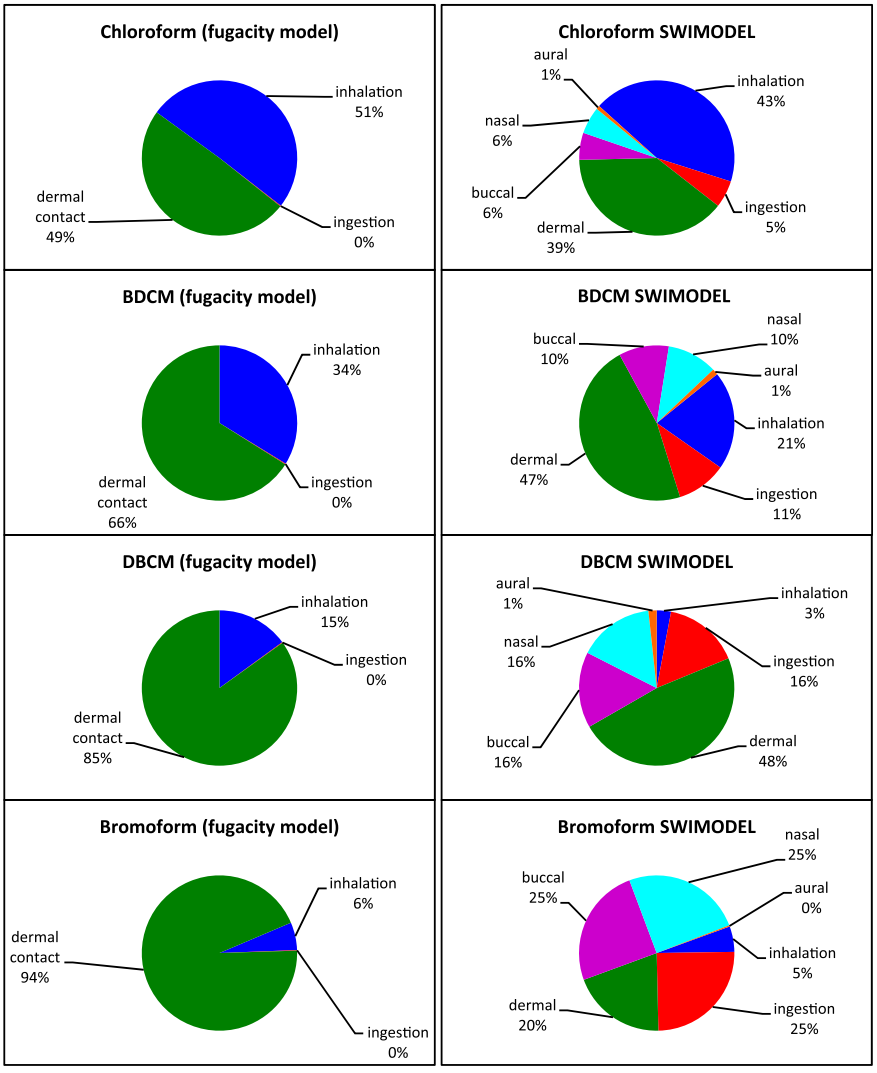

Fig. 7 – Comparison of proposed fugacity model with SWIMODEL.

ingestion, inhalation and dermal contact. Also for all THMs SWIMODEL predicts a much larger contribution to total exposure from ingestion than the fugacity model.

The influence of these alternative exposure routes may be exaggerated in the SWIMODEL approach. Little literature was found to support the use of these exposure routes (Beech, 1980) and they are not among the exposures commonly recommended by the US EPA (1992) or Health Canada (2010).

5. Discussion

A number of assumptions were made in this model that may affect the outcome. For the dermal contact exposures, the thickness of hydrated skin was used in the model as a probabilistic parameter. According to McKone and Howd (1992), the absorption of organic compounds into the skin is influenced by the skin hydration rate, with the absorption rate changing over time and reaching a steady state condition for absorption. In this study, the absorption rate was assumed to be constant. It was also assumed that there is no threshold concentration under which there would be no absorption, and also that there is no maximum dose absorbed after which the body is saturated and no more can be absorbed. There is also a potential that a swimmer's percentage of body fat could affect THM absorption and distribution in the body.

In this study, the inhalation dose was calculated using the air concentration of THMs and the breathing rate for each age group. The breathing rates used in the model were for moderate activity. Intake of THMs can be measured using breath samples (Jo et al., 1990; Lindstrom et al., 1997; Aggazzotti et al., 1998) and the concentration of THM in the breath is correlated to the dose within the body. The inhalation pathway was not considered a mass transfer process for the fugacity model due to the difficulty in calculating the amount of THM exhaled, assuming that it would change with the dose that had been taken up by the body. In that case, the exhaled amount with change over the time that the swimmer is in the pool area and it would no longer be possible to maintain a steady state condition in the model.

The swimming pool environment was assumed to be instantaneously and completely mixed both in the water and in the air. In the large volume of air in the natatorium, there is the potential for air currents caused by temperature gradients from the water surface to the ceiling, as well as near features such as hot tubs and saunas. Depending on the placement of air ventilation outlets and inlets, short circuits can develop in the air flow so that the mixing is not complete within the natatorium causing localized concentration of THMs close to the water surface. The inclusion of spatial variability would add to the complexity of the model with uncertain benefit. The air circulation system in a natatorium is typically designed to remove some of the moisture in the air. It is unknown what effect de-humidification has on the concentrations of THMs present in the air.

In a further study, the exposure doses generated in this study with the use of a fugacity model can be used to determine cancer and non-cancer health risks, including respiratory and reproductive effects. Risk management recommendations will be made based on sensitivity analysis of the model. Factors that should be explored include the impact on the results of the following parameters:

- pool size, temperature and humidity,
- number of bathers and activities they are involved in (high splashing, water slides etc.),
- pool water recirculation rates
- ventilation rates and water circulation rates, and
- disinfection agent chosen and the amount used.

An understanding of the partitioning and mass transport in the system of the pool environment and the risks generated could be used to calculate concentrations of chlorinated disinfection agents that can be safely added to the pool while balancing microbial risks with chemical risks.

6. Summary

Chlorination is important to preserve the health of swimmers in indoor pools. Microbial risks are effectively managed by the addition of chlorinated disinfectants to pool water. The generation of DBPs has been identified and characterized in many studies since their discovery. DBPs, specifically THMs, that are generated in the pool water are absorbed into the skin of swimmers, ingested by swimmers, and volatilized to the air where they can be inhaled by swimmers. The cancer and non-cancer risks associated with such exposure have been established by previous research.

A level III fugacity model has been developed to quantify the amount of THMs that are volatilized and absorbed by swimmers based on water concentration data collected from indoor pools in Quebec City. The modeled concentrations in air compared favorably with air concentrations measured in other studies. The resulting concentrations were used to determine exposure doses by dermal absorption, ingestion and inhalation exposure routes for 5 age groups: ages 1–4, ages 5–11, ages 12–17, ages 18–64 and ages >65. Future study is required to apply the doses to health risk formulas to determine the risks associated with swimming pool exposure to THMs. Risk management strategies should be developed that minimize THM exposure, without compromising disinfection efficiency.

Acknowledgments

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