# Multipathway risk assessment of trihalomethane exposure in drinking water of Lebanon

Lucy Semerjian and John Dennis

# ABSTRACT

The toxicological risks and lifetime cancer risks of trihalomethanes through oral ingestion, dermal absorption, and inhalation exposure from tap water in selected regions in Lebanon are estimated. Existing trihalomethane concentrations do not pose any non-carcinogenic and developmental risks in the exposed population via oral ingestion. Among the three pathways, residents have a higher risk of cancer through oral ingestion than through the other two pathways. The lifetime cancer risk through oral ingestion for dibromochloromethane makes the highest contribution to total risks, followed by bromodichloromethane, bromoform, and chloroform. The total multipathway cancer risk analysis suggests that no cancer risks exist during the summer and winter seasons; however, in the spring the total cancer risks exceeds the USEPA acceptable level of  $10^{-6}$  by a factor of 10.7.

Key words | drinking water, Lebanon, multipathway, risk assessment, trihalomethanes

Lucy Semerjian (corresponding author) Department of Civil and Environmental Engineering, American University of Beirut, Beirut, Lebanon Tel.: +961 1 34 79 52 Fax: +961 1 74 44 62 E-mail: Is07@aub.edu.lb

John Dennis

Department of Community Health Sciences, University of Calgary, Calgary, Alberta, Canada

# ABBREVIATIONS

AUB	American University of Beirut
BDCM	Bromodichloromethane
THM(s)	Trihalomethane(s)
ТСМ	Trichloromethane (chloroform)
DBCM	Dibromochloromethane
TBM	Tribromomethane (bromoform)
WHO	World Health Organization
USEPA	United States Environmental Protection Agency
EERC	Environmental Engineering Research Center
HQ	Hazard quotient
RfD	Reference Dose
LADD	Lifetime Average Daily Dose
CSF	Cancer Slope Factor
IRIS	Integrated Risk Information System
WTP	Water Treatment Plant

# **INTRODUCTION**

Since the beginning of the 20th century, chlorination has been a key treatment process for improving the doi: 10.2166/wh.2007.046

microbiological quality of drinking waters in urban distribution systems. However, the use of chlorine to disinfect drinking water leads to the formation of halogenated hydrocarbon by-products, which are potentially harmful to human health (Rook 1974; Bellar et al. 1974). Among such by-products, trihalomethanes (THM) (chloroform (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform (TBM)) have attracted extensive attention, as they have been found to exhibit potential carcinogenic activity in humans, namely bladder and rectum cancer (Morris et al. 1992), and adverse reproductive outcomes such as neural tube and central nervous system defects, low birth weight, stillbirth and miscarriage (Fawell 1999; WHO 2000; Nieuwenhuijsen et al. 2000; Hwang & Jaakkola 2003). THM are formed through the reaction of hypochlorous acid with natural organic matter in the presence or absence of bromide (Rook 1974; Bellar et al. 1974).

In Lebanon, the two most prevalent modes of water treatment are either direct disinfection at the source (boreholes and springs), or semi-conventional water treatment, namely screening, coagulation (mostly in winter), and filtration, followed by disinfection at centralized water treatment plants. Moreover, the most common, if not the sole, disinfection practice adopted by water authorities is chlorination using chlorine gas. No previous official investigations have been performed to assess and monitor the levels of THMs in public drinking waters of Lebanon although a large fraction of the population consumes chlorinated public drinking water, and thus may be prone to adverse health impacts due to chronic exposure to THM. Consumption of drinking water is not only for drinking purposes but also for cooking, showering, bathing, washing, laundering, cleaning, and so forth. Therefore, for many drinking water contaminants, there is the potential for exposure and uptake not only by ingestion but also through contact with the skin (dermal absorption) or by inhalation. Traditional risk assessments of water often consider only ingestion exposure to toxic chemicals, but scientists have proposed that inhalation and dermal absorption should be considered also in the risk assessment of drinking water (Weisel & Jo 1996; Weisel et al. 1999).

Therefore, the purpose of this study is to conduct a multipathway exposure assessment of selected public drinking waters of Lebanon based on the concentrations of THM within water distribution systems previously investigated by the author (Semerjian 2005; Semerjian *et al.* 2007).

# MATERIALS AND METHODS

#### Sample collection and study area

To investigate the occurrence of THM within water distribution systems, a seasonal sampling program was initiated during which a total of 196 samples were collected randomly from various water sources and water distribution systems throughout Lebanon. Three sampling points were selected for each sampling location to track the water from its source to the distribution network. However, THM levels recorded in the distribution systems only were included in the risk assessment since they represented the actual THM concentrations to which the assessed population was exposed. Therefore, duplicate samples were collected into pre-cleaned 60 ml amber screw cap glass vials fitted with

polytetrafluoroethylene (PTFE)-faced septa from each sampling point. Prior to sample collection, a homogeneous mixture of phosphate buffer and ammonium chloride was added to each sample vial to standardize the pH of all samples to 4.5-5.5, and to stop the action of residual chlorine thus preventing the formation of any additional THM compounds in chlorinated samples. Sample vials were filled to just overflowing without flushing out the buffer/dechlorinating agent mixture. Care was taken to ensure that no air bubbles passed through the sample as the bottle was filled, or were trapped in the sample when the bottle was sealed. All collected and preserved samples were stored at 4°C in a cooler and carried to the Environmental Engineering Research Center (EERC) at the American University of Beirut (AUB) for further analysis. Locations and geographical distributions of investigated water distribution systems are illustrated in Figure 1.

#### Analytical methods

Within specified allowable holding times, collected samples were processed and analyzed for trihalomethanes by liquidliquid extraction and gas chromatography with electron capture detector, using EPA Method 551.1 (USEPA 1995). This method is a reference method for chlorination by-products and is capable of speciating THM into individual compounds. A certified commercial trihalomethane calibration mix solution at a concentration of  $100 \,\mu$ g/ml of each THM species in methanol, obtained from Supelco (Bellefonte, PA, Cat. No. 4-7904), was used as the stock THM standard solution. Procedural calibration standards were prepared by adding appropriate volumes of the stock solution into 50-ml aliquots of buffered/dechlorinated reagent water. Decafluorobiphenyl (neat, Supelco 44-2538) was used as a certified commercial surrogate standard.

### **Risk assessment methodology**

Typical exposure routes to THMs in tap water are ingestion, inhalation, and dermal absorption. Traditionally, risk assessments for toxic chemical exposure from water often consider ingestion solely although showering has been shown to also increase the body burden of certain chemicals by inhalation and dermal absorption; thus these need to be



Figure 1 Geographical distribution of investigated water distribution networks.

considered in the analysis of total human exposure to volatile contaminants in tap water (Hsu *et al.* 2001). In general, a risk assessment process includes the following four components: data collection and evaluation, exposure assessment, toxicity assessment, and risk characterization. Results are then integrated and compared to estimates of intake with appropriate toxicological values to determine the likelihood of adverse effects in potentially exposed populations (Lee *et al.* 2004). In this study, two approved risk assessment models are adopted (1) the World Health Organization (WHO) index for additive toxicity, and (2) the USEPA-approved Risk Assistant model.

The WHO index for additive toxicity,  $I_{WHO}$ , for THMs is an overall guideline value to estimate the toxic (developmental and non-carcinogenic) risk associated with chlorinated drinking water. The  $I_{WHO}$  value should be  $\leq 1$  for compliance with WHO guidelines and is calculated as follows:

$$I_{WHO} = \frac{C_{TCM}}{GV_{TCM}} + \frac{C_{BDCM}}{GV_{BDCM}} + \frac{C_{DBCM}}{GV_{DBCM}} + \frac{C_{TBM}}{GV_{TBM}} \le 1$$

Where C is the surveyed concentration of each THM, and GV is the WHO guideline value. WHO GVs have been established separately at 200  $\mu$ g/L for TCM, 100  $\mu$ g/L for each of TBM and DBCM, and 60  $\mu$ g/L for BDCM (WHO 2004).

On the other hand, the USEPA Risk Assistant model is capable of estimating both toxic and carcinogenic risks.

Table 1 Exposure factors adopted in the risk assessment for the population of Lebanon

According to the USEPA model, toxicological risks, expressed as the hazard quotient (HQ), are calculated based on the comparison of actual exposure to a chemical to the reference dose (RfD) of that substance as follows:

 $HQ = [total amount ingested/body weight \times exposure time$ 

#### × reference dose]

Reference doses are extrapolated from toxicological studies of exposure which demonstrate a critical effect, are expressed in units of mg/kg/day, and are available in the Integrated Risk Information System (IRIS) database maintained by the USEPA (USEPA 2006). The total amount of chemical ingested depends on several typical or population-specific exposure factors such as the chemical concentration in local waters, water consumption rate, exposure frequency, and exposure duration. Body weight and exposure time estimates are also needed to calculate HQ. Using the USEPA approach, HQs in the Lebanese population were estimated based on the exposure factors summarized in Table 1.

In addition to toxic risks, carcinogenic risks of exposure to surveyed THM levels were calculated using the USEPA methodology. Carcinogenic compounds differ from toxic compounds in that there is no lower limit for the existence of risk. Thus, carcinogen risk assessment models are generally based on the premise that risk is proportional to

Factor	Estimate/Remarks						
Chemical concentration	• Actual recorded network THM concentrations, mg/L (Semerjian 2005; Semerjian <i>et al.</i> 2007)						
Exposure rate (ER)	• Based on a typical water consumption rate of 2 litres/day						
	• Note that in the HQ estimations, ingestion was the only exposure route considered						
Exposure frequency (EF)	• Equivalent to events per year, i.e. 365 days per year for water consumption						
Exposure duration (ED)	• Equivalent to life expectancy of 71.2 years. Data obtained from US Census Bureau, 2000. (http://www.census.gov)						
Average exposure time (AT)	- Based on life expectancy. Expressed in days; calculated as 71.2 years $\times$ 365 days/year = 25,988 days						
Body weight (BW)	• A typical adult weight of 70 kg is considered						

total lifetime dose, and the exposure metric used for carcinogenic risk assessment is the Lifetime Average Daily Dose (LADD). The LADD is typically used in conjunction with the Cancer Slope Factor (CSF) to calculate individual excess cancer risk. It is an estimate of the daily intake of a carcinogenic agent throughout the entire life of an individual. The CSF is the gradient of the line of the dose-response curve derived from laboratory toxicological studies, and values for each substance are available in the USEPA IRIS databases (USEPA 2006). For THM species, the USEPA range of concern is for an increased carcinogenic risk of  $10^{-6}$  i.e.1:1,000,000 (USEPA 2003).

In this study, multi-pathway (oral, dermal, and inhalation) exposures were considered in the cancer risk assessment of THM in the Lebanese population. Dermal exposure may occur via skin contact with THM contaminated water mainly during showering, bathing, and swimming; other activities such as washing and handling wet clothing also can be contamination contributors but to a lesser extent. According to USEPA, available skin-surface areas for chemical absorption vary between males and females; therefore, risk assessment for THM via dermal exposure was conducted separately for males and females (Miles et al. 2002; Lee et al. 2004). Inhalation exposure occurs when the air breathed contains compounds volatilized during water usage, such as bathing, showering, washing, and cooking. Showering has been identified as the activity contributing the greatest amount to inhalation exposure to volatile compounds (Lee et al. 2004). As a result, the calculation of cancer risks of THM through inhalation is only carried out for chloroform. In inhalation risk calculations,

the daily dose was calculated by assuming  $20 \text{ m}^3$  aspirated air per day (Lee *et al.* 2004). The chloroform concentration in air used for the estimation of risk through inhalation was calculated using a volatilization factor of 0.5 as suggested by USEPA (1991).

The following relationships were used to calculate the cancer risks for THMs through ingestion, dermal absorption, and inhalation. Exposure factors previously included in the calculations of non-carcinogenic risks were the same estimates as summarized in Table 1. Additional exposures factors related to dermal and inhalation THM exposure are discussed in Table 2.

THM carcinogenic risk of oral route =  $LADD_{oral} \times CSF_{oral}$ 

THM carcinogenic risk of dermal absorption

$$=$$
 LADD <sub>dermal</sub>  $\times$  CSF<sub>oral</sub>

THM carcinogenic risk of inhalation

= LADD<sub>inhalation</sub>  $\times$  CSF<sub>inhalation</sub>

Where LADD<sub>oral</sub> = [total amount ingested/body weight

× life time]

= (Conc. THM in water  $\times$  IR  $\times$  EF  $\times$  ED)/(BW  $\times$  AT)

 $LADD_{dermal} = (Conc. THM in water \times SA \times PC \times ET \times EF$ 

 $\times$  ED)/(BW  $\times$  AT)

 $LADD_{inhalation} = (Conc. TCM in water \times AA \times VF \times ET)$ 

 $\times\, \mathrm{EF} \times \mathrm{ED})/(\mathrm{BW} \times \mathrm{AT})$ 

Table 2 | Exposure factors adopted in the THM risk assessment via dermal absorption and inhalation exposure

Factor	Estimate/Remarks
Surface area (SA)	• Skin-surface available for contact in cm <sup>2</sup> . According to USEPA, male SA = 19,400 cm <sup>2</sup> , and female SA = 16,900 cm <sup>2</sup> (Lee <i>et al.</i> 2004)
Permeability constant (PC)	• Chemical-specific dermal permeability constant = $0.0020 \text{ cm/h}$ (Lee <i>et al.</i> 2004)
Exposure time (ET)	• Equivalent to exposure time per day or per event, estimated as $0.2 \text{ h/day}$ (Lee <i>et al.</i> 2004)
Aspirated air (AA)	• $20 \mathrm{m^3} \mathrm{per} \mathrm{day} = 0.83333 \mathrm{m^3/hr}$
Volatilization factor for chloroform (VF)	• $0.5 L/m^3$

# **RESULTS AND DISCUSSION**

# Non-carcinogenic risks for THM

#### The WHO index for additive toxicity approach

Applying this approach to network THM levels (Semerjian 2005) in Lebanon resulted in  $I_{WHO}$  values of less than 1 for all samples collected from the various distribution networks, and for the three sampling seasons. Computed  $I_{WHO}$ values ranged between 0.024-0.276, 0.020-0.200, and 0.020-0.110 for samples collected during spring, summer, and winter, respectively. Therefore, it can be concluded that the additive toxicity of recorded THM levels in the distribution networks of investigated sources is compliant with the WHO guideline value, and consequently such concentrations do not pose any adverse toxic health impacts. Calculated I<sub>WHO</sub> values for network THM levels recorded for individual locations are summarized in Table 3. The highest  $I_{WHO}$  values were observed for the distribution network of Hazmieh-Brazilia water treatment plant (WTP) during spring, the distribution network of Fanar well during summer, and the distribution network of Zheyma well during winter. It is noteworthy to mention that the water source for both Hazmieh-Brazilia WTP and Zheyma well is the Dayshounieh spring.

#### The USEPA risk assistant model approach

As previously mentioned, according to this USEPA model, toxicological risks are estimated and expressed as the (HQ). Hazard quotients in the Lebanese population were estimated based on the exposure factors summarized in Table 1. All investigated sampling locations, during the three seasons, exhibited acceptable HQ values ranging between 0-0.0814, 0-0.0114, 0-0.0117, and 0-0.0202, for TCM, BDCM, DBCM, and TBM (Table 4), respectively for a water consumption rate of 2 litres/day. Considering a consumption rate of 3 litres/day during the hot season, computed HQ values were also within acceptable norms, namely ranging between 0-0.1221, 0-0.0171, 0-0.0176, and 0-0.0303, for TCM, BDCM, DBCM, and TBM, respectively. In conclusion, THM concentrations found in local networks do not pose adverse developmental and non-carcinogenic risks in the Lebanese population.

	I <sub>WHO</sub>		
Sampling location	Spring	Summer	Winter
Dbayye WTP	0.058	0.051	0.075
Naameh chlorination tank	0.182	0.052	ND
Jamhour station	0.139	0.023	0.078
Hazmieh-Brazilia WTP	0.276	0.142	ND
	0.172		
Zheyma, Dayshounieh	0.227	0.159	0.110
Al Rayyes, Shoueifat	ND	0.042	ND
Al-Rishani, Shoueifat	0.082	0.048	0.083
Kanaan, Kfarshima	0.056	0.026	0.040
		0.022	0.035
Assaily, Kfarshima	ND	ND	0.033
Dekwaneh	0.113	0.045	0.065
Al-Zeghzeghy, Al-Sabtiyeh	0.120	0.111	ND
Jisr El-Basha	0.082	ND	0.046
Bonjus, Fanar	0.070	0.200	0.069
Mar Mtanios, Nahr El-Mot	0.068	0.075	0.078
Der Tamish	0.188	0.058	ND
Ayntoura	0.038	ND	ND
Jeita	0.055	ND	ND
Chananiir	0.061	ND	ND
Jbeil WTP	0.185	0.121	ND
Kfarhelda WTP	0.095	0.032	0.020
	0.115	0.037	0.064
	0.153	0.038	0.082
Kousba WTP	0.093	0.051	0.074
Chekka	0.047	0.032	ND
Anfeh	0.053	0.033	0.024

Table 3 Computed WHO additive toxicity values for network THM concentrations

#### Table 3 (continued)

	I <sub>WHO</sub>		
Sampling location	Spring	Summer	Winter
Fih	0.062	0.023	0.097
Tripoli WTP	0.056	0.036	ND
Abu Samra, Tripoli	0.027	0.020	ND
Al-Qebbeh, Tripoli	0.038	0.095	ND
	0.024		
	0.108		
Al-Jisr, Tripoli	0.067	ND	ND
Min	0.024	0.020	0.020
Max	0.276	0.200	0.110
Mean	0.100	0.063	0.063

Multi-pathway evaluations of lifetime cancer risks for THM

#### **Ingestion route**

Considering a water ingestion of 2 litres/day, computed cancer risks via oral exposure revealed that 90.3% (N = 28) of investigated networks during spring exceeded the set USEPA range of concern for an increased carcinogenic risk of 10<sup>-6</sup> for at least one THM species. Increased oral cancer risks for the spring season ranged between 1.19-1.39 folds, 1.08-14.14 folds, 1.54-19.70 folds, and 1.05-3.19 folds for TCM, BDCM, DBCM, and TBM, respectively. The highest cancer risk increases were recorded for DBCM followed by BDCM, TBM, and TCM. Regarding networks investigated during summer, 100% (N = 25) of investigated networks exceeded the set USEPA range of concern for at least one THM species. Computed increases in oral cancer risks for a consumption rate of 2 litres/day was 4.96 folds, and ranged between 1.22-8.19 folds, 1.26-16.12 folds, and 1.07-2.36 folds for TCM, BDCM, DBCM, and TBM, respectively. Finally, regarding networks investigated during winter, 94.1% (N = 16) of investigated networks exceeded the set USEPA range of concern for at least one THM species at a water consumption rate of 2 litres/day. Computed increases in oral cancer risks ranged between 1.67–5.45 folds, 1.14–8.26 folds, and 1.32– 1.82 folds for BDCM, DBCM, and TBM, respectively. It is noted that highest cancer risk increases were recorded for DBCM followed by BDCM, and TBM. No cancer risk increases were recorded for TCM during winter. Computed oral cancer risks of sampled networks exhibiting high THM carcinogenic risks for any of THM species are depicted in Figures 2, 3, and 4 for the spring, summer, and winter sampling seasons, respectively.

#### **Dermal absorption**

Risk assessment by dermal absorption was conducted separately for males and females because of gender-specific exposure factors. Computations revealed that dermal carcinogenic risks from exposure to all surveyed THM levels were below the lower end of the range of acceptable risk by the USEPA for both males (TCM: 0-1.93E-08; BDCM: 0- 5.48E-08; DBCM: 0-7.65E-08; TBM: 0-1.24E-08) and females (TCM: 0-1.68E-08; BDCM: 0- 4.78E-08; DBCM: 0-6.66E-08; TBM: 0-1.08E-08) although computed risks were higher for males possibly due to the higher skin-surface area.

#### Inhalation exposure

Estimation of THM cancer risks through inhalation (during bathing) was conducted only for TCM. Computations revealed that dermal carcinogenic risks from exposure to all surveyed TCM levels were below the lower end of the range of acceptable risk by the USEPA during spring (0-7.69E-07), summer (0-3.51E-07) and winter (0-3.24E-07).

# Total cancer risk of THM from multi-pathway evaluations

The seasonal average cancer risks for THM in investigated distribution networks through oral ingestion, genderspecific dermal absorption, and inhalation are illustrated in Figure 5. The graphs indicate that the exposed population has a higher risk of cancer through oral ingestion, and that DBCM is the highest contributor, followed by BDCM, TBM, and TCM during all seasons. To understand the total risk of

Table 4	Estimated non-carcinogenic risks of surveyed network THM levels in the Lebanese population for a consumption rate of 2 litres/day

	HQ <sub>TCM</sub>			HQ <sub>BDCM</sub>			HQ <sub>DBCM</sub>			HQ <sub>твм</sub>		
Sampling location	Spring	Summer	Winter	Spring	Summer	Winter	Spring	Summer	Winter	Spring	Summer	Winter
Dbayye WTP	0.0025	0.0006	0.0019	0.0016	0.0006	0.0017	0.0028	0.0016	0.0043	0.0022	0.0045	0.0031
Naameh chlorination tank	0.0053	0.0000	ND	0.0043	0.0004	ND	0.0092	0.0000	ND	0.0084	0.0068	ND
Jamhour station	0.0091	0.0009	0.0031	0.0053	0.0006	0.0034	0.0064	0.0015	0.0037	0.0024	0.0006	0.0010
Hazmieh-Brazilia WTP	0.0228	0.0814	ND	0.0114	0.0000	ND	0.0117	0.0000	ND	0.0030	0.0000	ND
	0.0100			0.0065			0.0086			0.0027		
Zheyma, Dayshounieh	0.0139	0.0104	0.0096	0.0076	0.0066	0.0044	0.0102	0.0068	0.0049	0.0060	0.0023	0.0011
Al Rayyes, Shoueifat	ND	0.0014	ND	ND	0.0004	ND	ND	0.0011	ND	ND	0.0040	ND
Al-Rishani, Shoueifat	0.0026	0.0006	0.0000	0.0010	0.0002	0.0000	0.0021	0.0010	0.0003	0.0073	0.0055	0.0115
Kanaan, Kfarshima	0.0031	0.0007	0.0000	0.0011	0.0003	0.0005	0.0020	0.0016	0.0014	0.0034	0.0014	0.0035
		0.0006	0.0000		0.0002	0.0006		0.0013	0.0010		0.0015	0.0031
Assaily, Kfarshima	ND	ND	0.0000	ND	ND	0.0007	ND	ND	0.0003	ND	ND	0.0033
Dekwaneh	0.0035	0.0016	0.0007	0.0015	0.0003	0.0000	0.0035	0.0017	0.0008	0.0093	0.0038	0.0084
Al-Zeghzeghy, Al-Sabtiyeh	0.0031	0.0008	ND	0.0018	0.0013	ND	0.0016	0.0049	ND	0.0118	0.0086	ND
Jisr El-Basha	0.0032	ND	0.0008	0.0012	ND	0.0000	0.0028	ND	0.0012	0.0062	ND	0.0052
Bonjus, Fanar	0.0030	0.0034	0.0009	0.0000	0.0036	0.0014	0.0017	0.0096	0.0039	0.0076	0.0122	0.0036
Mar Mtanios, Nahr El-Mot	0.0026	0.0009	0.0000	0.0009	0.0003	0.0000	0.0009	0.0008	0.0002	0.0066	0.0091	0.0109
Der Tamish	0.0041	0.0027	ND	0.0013	0.0000	ND	0.0035	0.0014	ND	0.0202	0.0062	ND
Ayntoura	0.0027	ND	ND	0.0000	ND	ND	0.0000	ND	ND	0.0047	ND	ND
Jeita	0.0028	ND	ND	0.0011	ND	ND	0.0020	ND	ND	0.0034	ND	ND
Chananiir	0.0000	ND	ND	0.0014	ND	ND	0.0021	ND	ND	0.0043	ND	ND
Jbeil WTP	0.0195	0.0006	ND	0.0082	0.0003	ND	0.0068	0.0016	ND	0.0011	0.0149	ND
Kfarhelda WTP	0.0092	0.0008	0.0001	0.0045	0.0006	0.0004	0.0034	0.0016	0.0007	0.0005	0.0018	0.0014
	0.0116	0.0013	0.0016	0.0056	0.0010	0.0019	0.0039	0.0022	0.0034	0.0004	0.0011	0.0021
	0.0065	0.0012	0.0028	0.0048	0.0010	0.0023	0.0068	0.0023	0.0047	0.0056	0.0012	0.0024
Kousba WTP	0.0000	0.0042	0.0076	0.0055	0.0023	0.0039	0.0042	0.0018	0.0022	0.0000	0.0005	0.0000
Chekka	0.0029	0.0000	ND	0.0010	0.0003	ND	0.0018	0.0014	ND	0.0024	0.0026	ND
Anfeh	0.0000	0.0006	0.0000	0.0016	0.0002	0.0005	0.0024	0.0011	0.0016	0.0024	0.0031	0.0010

 Table 4
 (continued)

	HQ <sub>TCM</sub>			HQ <sub>BDCM</sub>			HQ <sub>DBCM</sub>			НQ <sub>твм</sub>		
Sampling location	Spring	Summer	Winter	Spring	Summer	Winter	Spring	Summer	Winter	Spring	Summer	Winter
Fih	0.0025	0.0006	0.0033	0.0021	0.0002	0.0031	0.0033	0.0010	0.0049	0.0015	0.0018	0.0029
Tripoli WTP	0.0000	0.0006	ND	0.0015	0.0005	ND	0.0026	0.0018	ND	0.0030	0.0025	ND
Abu Samra, Tripoli	0.0000	0.0006	ND	0.0000	0.0002	ND	0.0016	0.0008	ND	0.0023	0.0017	ND
Al-Qebbeh, Tripoli	0.0000	0.0006	ND	0.0000	0.0000	ND	0.0000	0.0084	ND	0.0054	0.0050	ND
	0.0000			0.0000			0.0000			0.0034		
	0.0056			0.0000			0.0056			0.0085		
Al-Jisr, Tripoli	0.0000	0.0000	ND	0.0000	ND	ND	0.0000	ND	ND	0.0095	ND	ND



Figure 2 | Cancer risks of THM through oral route in tap waters in Lebanon during spring.



Figure 3 | Cancer risks of THM through oral route in tap waters in Lebanon during summer.



Figure 4 | Cancer risks of THM through oral route in tap waters in Lebanon during winter.



Figure 5 | Multi-pathway average cancer risks for THM in investigated networks in Lebanon.

 Table 5
 Total cancer risks from multipathways for exposed population in Lebanon

Parameter	Total cancer risk
Multipathway cancer risk for males (spring)	1.07E-05
Multipathway cancer risk for females (spring)	1.07E-05
Multipathway cancer risk for males (summer)	6.01E-06
Multipathway cancer risk for females (summer)	6.01E-06
Multipathway cancer risk for males (winter)	6.55E-06
Multipathway cancer risk for females (winter)	6.55E-06

THM on human health through the different pathways, the total cancer risks from all three pathways and during the various seasons are summarized in Table 5. Total multipathway cancer risks suggest that no cancer risks exist during the summer and winter seasons; however, in the spring the total cancer risk exceeds the USEPA acceptable level of  $10^{-6}$  by a factor of 10.7.

# CONCLUSIONS

The study evaluated the association between THM exposure through three different pathways and lifetime cancer risks in selected regions in Lebanon. Results indicate that a higher risk of cancer may exist through oral ingestion. In fact, 90.3-100% of investigated networks, depending on the season, exceed the set USEPA range of concern for an increased carcinogenic risk of 10<sup>-6</sup> for at least one THM species. In Lebanon, DBCM and BDCM posed a higher cancer risk in the exposed population than did TBM, and TCM. Dermal and inhalation carcinogenic risks from exposure to all surveyed THM levels were below the lower end of the range of acceptable risk by the USEPA for both males and females. A total multipathway cancer risk analysis suggested that no cancer risks exist during the summer and winter seasons, however, in the spring the total cancer risk exceeds the USEPA acceptable level of  $10^{-6}$  by a factor of 10.7.

Non-carcinogenic risk assessment for THM was also conducted using the WHO index for additive toxicity approach as well as the USEPA Risk Assistant model approach. Both approaches indicated that network THM concentrations do not pose adverse developmental and non-carcinogenic risks in the Lebanese population.

# REFERENCES

- Bellar, T. A., Bienlien, J. J. & Kroner, R. C. 1974 The occurrence of organohalides in chlorinated drinking water. J Am. Wat. Works Assoc. 66(12), 703–706.
- Fawell, J. 1999 The risks of DBPs in perspectives. In Disinfection By-Products in Drinking Water: Current Issues (ed. M. Fielding & M. Farrimond), Royal Society of Chemistry, Cambridge, UK. pp. 157–164.
- Hsu, C. H., Jeng, W. L., Chang, R. M., Chien, L. C. & Han, B. C. 2001 Estimation of potential lifetime cancer risks for trihalomethanes from consuming chlorinated drinking water in Japan. *Environ. Res. Sec. A* 85, 77–82.
- Hwang, B. F. & Jaakkola, J. K. 2003 Water chlorination and birth defects: a systematic review and meta-analysis. Arch Environ Health 58(2), 83–89.
- Lee, S. C., Guo, H., Lam, S. M. J. & Lau, S. L. A. 2004 Multipathway risk assessment on disinfection by-products of drinking water in Hong Kong. *Environ Res.* 94, 47–56.
- Miles, A. M., Singer, P. C., Ashley, D. L., Lynberg, M., Mendola, P., Langlois, P. & Nuckols, J. R. 2002 Comparison of trihalomethanes in tap water and blood. *Environ. Sci. Technol.* 36(8), 1692–1698.
- Morris, R. D., Audet, A. M., Angelillo, I. F., Chalmers, T. C. & Mosteller, F. 1992 Chlorination, chlorination by-products, and cancer: a meta-analysis. *Am. J. Public Health* 82(7), 955–963.
- Nieuwenhuijsen, M. J., Toledano, M. B., Eaton, N. E., Fawell, J. & Elliott, P. 2000 Chlorination disinfection by-products in water and their association with adverse reproductive outcomes: a review. Occup. Environ. Med. 57, 73–85.
- Rook, J. J. 1974 Formation of haloforms during the chlorination of natural water. *Water Treatment Exam.* 23(2), 234–243.
- Semerjian, L. 2005 Trihalomethanes in Drinking Waters of Lebanon. PhD thesis. Department of Geographical and Environmental Sciences, University of Bradford, UK.
- Semerjian, L., Dennis, J. & Ayoub, G. 2007 Spatial and seasonal evolution of trihalomethanes in water distribution systems in Lebanon. J. Wat. Supp. Res. Tech. - AQUA 56(4).
- USEPA (United States Environmental Protection Agency) 1995 Determination of Chlorination DBPs, Chlorinated Solvents, and Halogenated Pesticides/Herbicides in Drinking Water by Liquid-liquid Extraction and Gas Chromatography with Electron Capture Detector. Method 551.1. Office of Research and Development, National Exposure Research Laboratory, Cincinnati, Ohio.
- USEPA (United States Environmental Protection Agency) 2003 Draft Final Guidelines for Carcinogen Risk Assessment, Risk Assessment Forum, EPA/630/P-03/001A, NCEA-F-0644A, Washington, DC (www.epa.gov/ncea/raf/cancer2003.htm).

- USEPA (United States Environmental Protection Agency) 2006 Integrated Risk Information System (Electronic database). Office of Research and Development, National Center for Environmental Assessment, Washington, DC, (http://www.epa.gov/iris).
- USEPA 1991 *Risk Assessment Guidance for Superfund–Vol. I, Part B.* US Environmental Protection Agency, Washington, DC, EPA/540/R-92/003.
- Weisel, C. P. & Jo, W. K. 1996 Ingestion and dermal exposure to chloroform and trichloroethene from tap water. *Environ. Health Persp.* 104, 48–51.
- Weisel, C. P., Kim, H., Haltmeier, P. & Klotz, J. B. 1999 Exposure estimates to disinfection by-products of chlorinated drinking water. *Environ. Health Persp.* **107**, 103–110.
- WHO (World Health Organization) 2000 *Disinfectants and Disinfectant By-Products*. Environmental Health Criteria 216, Geneva, Switzerland.
- WHO (World Health Organization) 2004 Draft Third Edition of the WHO Guidelines for Drinking-Water Quality. WHO, Geneva, Switzerland, http://www.who.int/water\_sanitation\_health/ dwq/guidelines3rd/en/.

Available online May 2007