Monitoring of chlorination disinfection by-products and their associated health risks in drinking water of Pakistan

Sidra Abbas, Imran Hashmi, Muhammad Saif Ur Rehman, Ishtiaq A. Qazi, Mohammad A. Awan and Habib Nasir

ABSTRACT

This study reports the baseline data of chlorination disinfection by-products such as trihalomethanes (THMs) and their associated health risks in the water distribution network of Islamabad and Rawalpindi, Pakistan. THM monitoring was carried out at 30 different sampling sites across the twin cities for 6 months. The average concentration of total trihalomethanes (TTHMs) and chloroform ranged between 575 and 595 μg/L which exceeded the permissible US (80 μg/L) and EU (100 μg/L) limits. Chloroform was one of the major contributors to the TTHMs concentration (>85%). The occurrence of THMs was found in the following order: chloroform, bromodichloromethane > dibromochloromethane > bromoform. Lifetime cancer risk assessment of THMs for both males and females was carried out using prediction models via different exposure routes (ingestion, inhalation, and dermal). Total lifetime cancer risk assessment for different exposure routes (ingestion, inhalation, and skin) was carried out. The highest cancer risk expected from THMs seems to be from the inhalation route followed by ingestion and dermal contacts. The average lifetime cancer risk for males and females was found to be 0.51×10^{-3} and 1.22×10^{-3}, respectively. The expected number of cancer risks per year could reach two to three cases for each city.

Key words | chlorination, disinfection by-products, distribution network, solid phase micro-extraction (SPME), trihalomethanes

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INTRODUCTION

The availability of clean and safe drinking water is one of the major concerns in developing countries. Intake of poor quality drinking water may lead to several health risks to the community, and eventually, it affects the magnitude of the healthcare budget. Thus, it is important to appropriately manage and treat drinking water for the benefit of society. Contaminated drinking water is a major carrier of disease causing organisms; these pathogenic organisms may pose a serious threat to human health (Ashbolt 2004; Duke *et al.* 2006).

Currently, almost one-half of the population of Pakistan has no or little access to potable drinking water (PCRWR 2006), whereas only one-quarter of the whole population has access to clean drinking water in Pakistan (Hashmi *et al.* 2009). Approximately 70% of Pakistan's surface and sub-surface water supply sources are not appropriate for drinking due to significant organic, inorganic, and biological contamination (PCRWR 2006; Baig *et al.* 2011). Therefore, disinfection plays a significant role in the supply of safe drinking water. The destruction of microbial pathogens is essential and very commonly involves the use of reactive chemical agents such as chlorine. Chlorination is one of the most common methods for water disinfection due to its effective oxidizing potential, cost–effectiveness, and simplicity of operation (Karim *et al.* 2011; Pardakhti *et al.* 2011; Chowdhury 2012a, 2012b). Chlorination inactivates pathogenic organisms responsible for waterborne diseases such as cholera, typhoid fever, and dysentery. Furthermore, residual chlorine restricts the microbial recontamination throughout the water distribution system

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(Hassan *et al.* 2010). However, chlorination may form chlorination disinfection by-products (CDBPs) due to the oxidation of organic matter, anthropogenic contaminants, and bromide/iodide present in most surface water resources (rivers, lakes, and many groundwaters). The residual chlorine reacts with natural organic matter (NOM). CDBPs normally include trihalomethanes (THMs), which are considered potential carcinogens (Bischel & von Gunten 2000a; Imo *et al.* 2007). Generally, disinfection of surface water produces more THMs as compared to groundwater due to the presence of high organic matter (Hassan *et al.* 2010).

More than 600 DBPs have been reported in drinking water after disinfection. There are two major types of chlorinated disinfection by-products, THMs and haloacetic acids (HAAs). According to Rook (1974), THMs have further been categorized into four main species: chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$; BDCM), dibromochloromethane ($CHBr_2Cl$; DBCM), and bromoform ($CHBr_3$). Bull *et al.* (2001) have reported that these CDBPs are not only mutagenic but also potential carcinogens. Many recent studies indicate a connection between cancer and exposure to THM-contaminated potable water (Goi *et al.* 2005). These CDBPs pose a threat to human health even at very low concentrations. The potential health effects include different kinds of cancers, reproductive disorders, miscarriages, and birth defects (Wright *et al.* 2004; Ristoiu *et al.* 2009). CDBPs formation depends on the amount and chemical composition of organic species present in water along with other factors such as temperature, pH, chlorine dose, and retention time. Humans are exposed to these THMs via oral, inhalation, and dermal routes. Thus, monitoring of THMs has become mandatory to ensure the public's health against their carcinogenic and non-carcinogenic risks, associated with public water supplies (Lee *et al.* 2004; Mallika *et al.* 2008; Chowdhury *et al.* 2011a, 2011b; Karim *et al.* 2011; Pardakhti *et al.* 2011).

This study aims at the monitoring of THMs in drinking water supplies in the twin cities of Rawalpindi and Islamabad, and to predict cancer and non-cancer risk associated with THMs via three different routes using different models.

MATERIALS AND METHODS

Sampling area

Rawalpindi and Islamabad are known as twin cities of Pakistan with more than two million inhabitants. These cities are supplied with drinking water from Rawal Lake, Simly Dam, and Khanpur Dam after being treated in their respective water treatment plants. Simly Dam is the largest water reservoir with a total depth of approximately 2,300 ft (701 m). Disinfection by chlorination is practiced at Simly, Khanpur, and Rawal Dam filtration plants as means of water disinfection, and treated water is supplied to the public via distribution networks. Surface water from these resources is filtered and then it is subjected to on-site chlorination (Hashmi *et al.* 2009). This chlorination is carried out by non-technical staff based on their experience rather than exact calculations. The chlorinated water is, thus, supplied to the twin cities. Thirty sites were selected for sampling across the twin cities (Figure 1). Three replicate samples were collected from each of the 30 locations. The samples were taken directly from consumers' taps after letting the water run for several minutes before collecting the water in pre-cleaned glass containers with sodium thiosulfate (10 mg for 10 mL of sample) preservative to eliminate any residual chlorine (APHA 2005). Samples were collected in 40 mL vials. Vials were completely filled with the samples leaving no headspace, and were stored below 4 °C in a dark room for further analysis (Norin & Renberg 1980).

All samples were analyzed within 14 days of collection for quality assurance. Much care is needed in sample collection. No air bubbles should pass through the sample as the bottle is filled, or be trapped in the sample when the bottle is sealed. Samples were collected from (1) underground storage tanks, (2) consumer taps, and (3) overhead reservoirs. All the samples were collected in triplicate. Measurement of pH, temperature, and residual chlorine were carried out in the field as mentioned in *Standard Methods for the Examination of Water and Wastewater* (APHA 2005). Other measurements were done in the laboratory.

Water samples were collected from the main water supply systems and treatment plants. The pretreated water samples were collected in 1 L sterilized glass bottles. The

Figure 1 | Map showing sampling sites of Rawalpindi and Islamabad (Courtesy Google Maps).

glass bottles were cleaned and air dried in a hot oven at 100 °C for 30 minutes prior to use. After drying, the glassware was sealed and stored in a clean environment to prevent any accumulation of dust or other contaminants. They were store inverted and capped with aluminum foil.

For THMs analysis, samples were collected in 40 mL clean glass vials. For head space solid phase micro-extraction (SPME) 25 mL of vial were filled with water and the remaining 15 mL was left for head space. For liquid SPME glass vials were fully filled with water.

Chemicals and standards

Chloroform ($CHCl_3$), bromodichloromethane ($CHBrCl_2$), dibromochloromethane ($CHBr_2Cl$), and bromoform ($CHBr_3$) were purchased from Dr Ehrenstrofer (Germany). Standard stock solution of individual THMs were prepared by mixing 10 μL of the standard analyte in 100 mL GC-grade methanol and stored in sealed glass bottles at 4 °C.

DBP analysis

THM concentration was measured with a gas chromatography/electron capture detector (GC/ECD) equipped with fused silica capillary column. Gas chromatographic analysis was performed using a Shimadzu 2010 series gas chromatograph coupled with ECD detector.

For sample extraction, the SPME technique using Supleco cat. No.57344-U manual solid-phase microextraction fiber assembly fitted with a 75 μm (Car PDMS) fiber was used. The column used was fused silica capillary with a length of 30 cm, inner diameter 0.53 mm, thickness 0.88 μm and filling material of 5% diphenyl, 95% dimethyl-polysiloxane.

Fiber was first conditioned at 280 °C for 1 h before use. All the samples were analyzed within 2 weeks of collection following USEPA method 551.1 and 552.2 (USEPA 1995a, 1995b). Extraction was performed with fiber immersed in the headspace for extraction for 10–15 min at 25 °C. Two microliters of the THMs extracts were analyzed. Procedural calibrations were developed using THMs (chloroform, bromodichloromethane, dibromochloromethane, bromoform) standards. Furthers details of the experimental procedures and calibrations can be found elsewhere (USEPA 1995a). More details of the GC-FED analysis can also be found in the literature (Ghaffar *et al.* 2012; Amjad *et al.* 2013).

Exposure assessment

Exposure assessments were conducted based on the measured concentration of THMs in drinking water. Carcinogenic risks of exposure to THMs levels were calculated on the basis of other recently reported studies (Basu *et al.* 2011; Pardakhti *et al.* 2011; Lee *et al.* 2013). Oral ingestion and dermal absorption were considered as the two important ways for exposure to THMs. The cancer risks through inhalation, ingestion, and dermal contact for each of the four THMs were calculated as the product of their chronic daily intake (CDI) and the potency or the slope factor (SF), that is, CDI × SF. The CDI of each THM species through oral ingestion and dermal absorption were estimated as follows. The assessment was performed on all three entry routes including oral ingestion, inhalation, and dermal absorptions using CDI estimation. Showering was considered as a major route for inhalation and dermal absorption (Wang et al. 2007; Pardakhti et al. 2011). CDI values for different routes were calculated using the following equations (Pardakhti et al. 2011; Lee et al. 2013):

$$\text{Oral ingestion (mg/kg-day)} = [\text{CW} \times \text{IR} \times \text{EF} \times \text{ED} \times \text{CF}]/(\text{BW} \times \text{AT}) \quad (1)$$

$$\text{Dermal absorption (mg/kg-day)} = [\text{DA} \times \text{SA} \times \text{F} \times \text{EF} \times \text{ED}]/(\text{BW} \times \text{AT}) \quad (2)$$

$$\text{DA (mg/m2.shower)} = 2\text{FA} \times \text{Pd} \times \text{CW}\sqrt{(6\tau \times t)/\Pi} \quad (2a)$$

$$\text{Inhalation absorption (mg/kg-day)} = [\text{C air} \times \text{R} \times \text{t} \times \text{F} \times \text{EF} \times \text{ED} \times \text{CF}]/(\text{BW} \times \text{AT}) \quad (3)$$

C air can be estimated as follows using the two-resistance theory (Little 1992):

$$\text{C air} = \text{QW} \times \text{Pv} \times \text{CW}(1 - \text{e} - \text{Ka.t})/\text{Ka.V} \quad (3a)$$

$$\text{Total Risk} = (\text{CDI Ingestion} \times \text{CSF Oral}) + (\text{CDI Inhalation} \times \text{CSF Inhalation}) + (\text{CDI Dermal} \times \text{CSF Dermal}) \quad (4)$$

The values of input parameters and cancer slope factors (CSF) are given in Table 1.

RESULTS AND DISCUSSION

THMs analytes mixture

The current study shows that the concentration of THMs is relatively higher as compared to other published literature, as shown in Figures 2–6. The occurrence of residual chlorine in the effluent in the drinking water chlorination treatment plant may also contribute to high levels of THMs in the drinking water distribution network (Imo et al. 2007). In this research, the sampling sites were fairly distant from the main water storage facilities and chlorination plant, which may be the probable reason for higher THMs concentration in drinking water. The higher values of total trihalomethanes (TTHMs) in Islamabad than in Rawalpindi could be elucidated on the basis of the main source of drinking water supply. Islamabad is mainly supplied with surface water, whereas Rawalpindi can access groundwater as well as a water supply from the chlorination plant. Groundwater does not have much NOM which contributes to the formation of higher concentrations of THMs (Imo et al. 2007). Thus it can be stated that the presence of higher concentrations of chloroform in Islamabad was mainly due to the supply of surface water. Surface water was susceptible to containing higher organic matter compared to well water due to excessive vegetation and high temperature throughout the year. Chlorine treatment of such surface water, with higher organic matter, leads to the formation of high concentrations of chloroform in the downstream water supply (Imo et al. 2007). This can be observed in the high concentration of TTHMs at sampling station 9. Again, the probable reason may be the distance of this station from the treatment plant and the high organic content in the main water reservoirs that is further treated at the main filtration plant supplying water to that area. Almost 95% of samples were contaminated with THMs. Chloroform was found to be a maximum in all samples from the entire drinking water supply network, i.e., underground tank, overhead reservoir, and filtration plants. The concentration of BDCM was between 0.5 and 33.2 mg/L for all locations, whereas the concentration of DBCM was detected in the range of 1.09–18 mg/L. Bromoform was only detected at four locations (data not shown). Thus, chloroform alone contributed more than 90% at all the sampling sites. BDCM and DBCM made a trivial contribution of 5–10% and 0–5%, respectively. Bromoform had presented an almost insignificant contribution to TTC which was found to be consistent with other studies (Basu et al. 2011). Chloroform was found to be the most common THMs in all drinking water samples followed by BDCM, DBCM, and bromoform. Chloroform and DBCM were found in all the samples thus presenting 100% occurrence frequency, whereas BDCM was detected in 90% samples. Bromoform was detected in only 10% of samples. The occurrence

Table 1 | Input parameters and abbreviations for cancer and non-cancer exposure assessment (Pardakhti *et al.* 2011; Lee *et al.* 2013)

Type of parameter	Parameter	Notation	Unit	Value	References
General	THM conc. in water	C_w	μg/L	Table	This study
	Average lifetime	AT	Days	64.2 × 365 (M), 67.9 × 365 (F)	Lee *et al.* (2004)
	Body weight	BW	kg	68.11 (M), 55.23 (F)	Aslam *et al.* (2010)
	Exposure duration	ED	Year	64.2 (M), 67.9 (F)	Lee *et al.* (2004)
	Exposure frequency	EF	Days/year	365	Lee *et al.* (2004)
	Exposure time	t	min/day	35	RAIS (2009)
Oral ingestion	Ingestion rate	IR	L/day	2	USEPA (1997)
Dermal	Skin surface area (4BW + 7)/(BW + 90)	SA	m^2	1.77 (M), 1.57 (F)	USEPA (1997)
	Shower frequency	F	Shower/day	1	
	Permeability coefficient	PC	m/min	2.85×10^{-6} (chloroform), 9.79×10^{-7} (BDCM), 1.68×10^{-6} (DBCM), 1.20×10^{-6} (bromoform)	Lee *et al.* (2013)
	Fraction absorbed water for THMs			1	Lee *et al.* (2013)
Inhalation absorption	THM concentration in air	C_a	μg/L	Equation	Little (1992)
	Air intake rate	R	m^3/min	0.015 (M), 0.012 (F)	Lee *et al.* (2013)
	Bathroom volume	V	m^3	4.78	Chen *et al.* (2003)
	Water flow rate	Q_W	L/min	10	Little (1992)
	Lag time per shower	τ	min/shower	30 (chloroform), 52.8 (BDCM), 94.2 (DBCM), 167.4 (bromoform)	Lee *et al.* (2013)
	Air change	Ka	min^{-1}	0.021	Lee *et al.* (2013)
	THM transformation rate from water to air	Pv	%	8.76	Chowdhury& Champagne (2009)
	Carcinogenic slope factor (oral/dermal)	CSF	mg kg^{-1} day^{-1}	0.031 (chloroform), 0.062 (BDCM), 0.084 (DBCM), 0.0079 (bromoform)	Pardakhti *et al.* (2011)
	Carcinogenic slope factor (inhalation)	CSF	mg kg^{-1} day^{-1}	8.05×10^{-5} (chloroform), 0.13 (BDCM), 0.095 (DBCM), 0.00385 (bromoform)	Pardakhti *et al.* (2011)
Hazard index	Reference dose	RfD	mg kg^{-1} day^{-1}	0.01 (chloroform), 0.02 (BDCM), 0.02 (DBCM), 0.02 (bromoform)	Lee *et al.* (2004)

M, male; F, female.

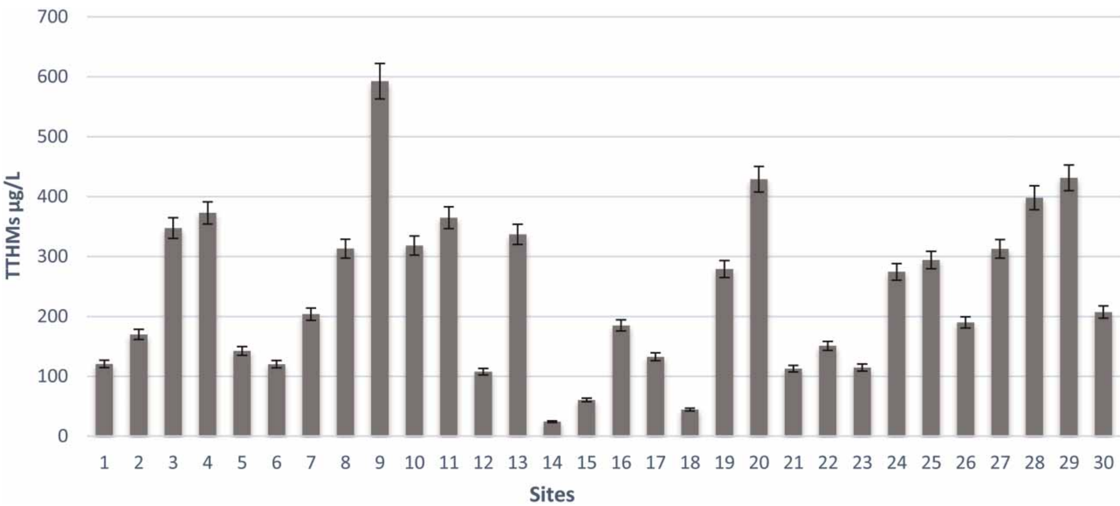

Figure 2 | Total concentration of THMs determined at different sites.

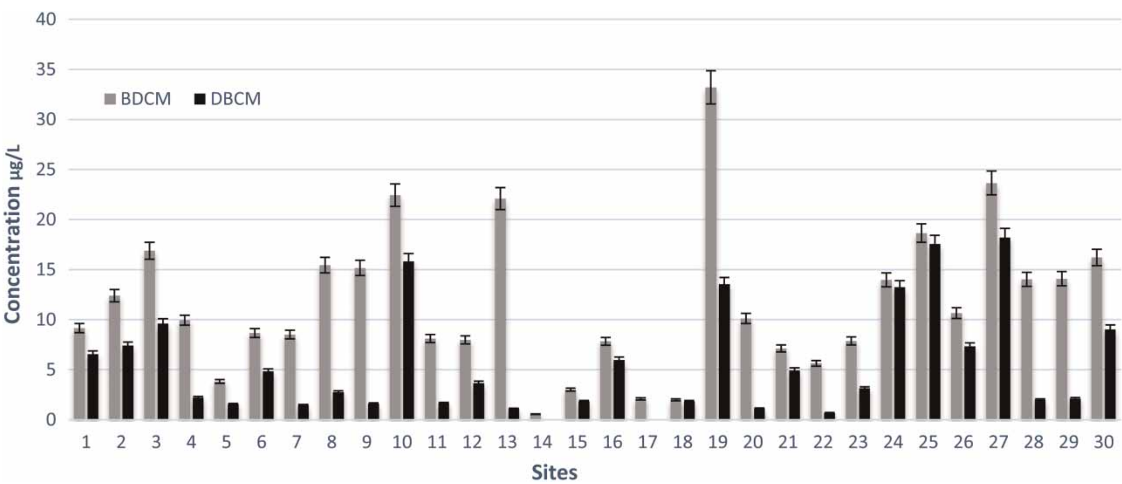

Figure 3 | Total concentration of BDCM and DBCM at different sites.

frequency trend of THMs is as follows: chloroform, BDCM, DBCM, bromoform. Similar observations have been reported in the literature (Karim *et al.* 2011; Pardakhti *et al.* 2011). Chloroform was comparatively lower in concentration in underground tanks and sampling station 14. Concentration of TTHMs ranged from 44.51 to 595.86 at different sampling stations (Table 1). These average concentrations of the reported species surpassed the authorized permissible limits of TTC in the USA (80 mg/L) and the EU (100 mg/L) as reported in the literature (Chowdhury *et al.* 2011a). The potential reason for contamination at different points is the presence of NOM. THMs formation occurs when chlorine is added to such water sources.

The results of this research showed that the total concentration of THMs found was much higher compared with other studies of a similar nature carried out in other countries (Chowdhury *et al.* 2011a; Fooladvand *et al.* 2011; Karim *et al.* 2011; Legay *et al.* 2011a, 2011b; Pardakhti *et al.* 2011). Out of 30 sampling sites only three sites met the USEPA drinking water quality standard values, and the remaining 27 sites exceeded the standard value of 80 µg/L.

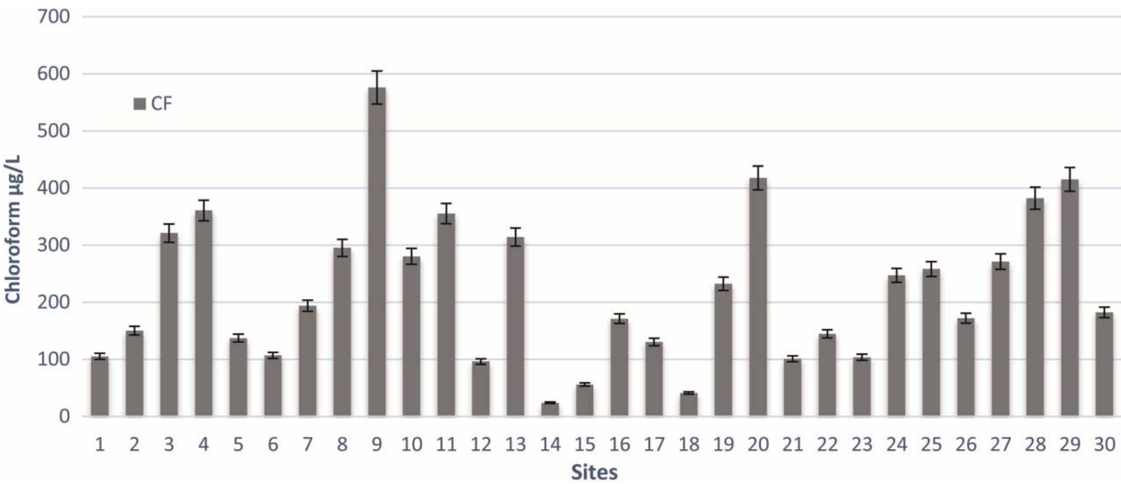

Figure 4 | Total concentration of chloroform determined at different sites.

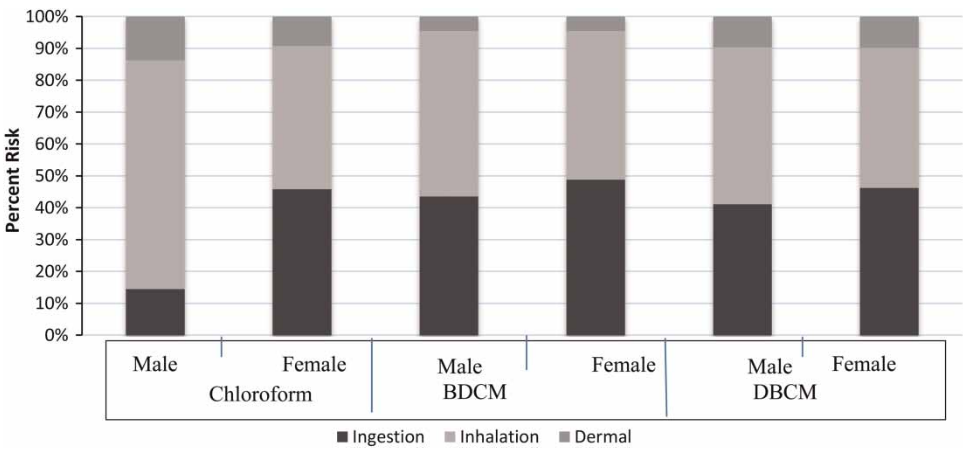

Figure 5 | Ratio of ingestion, inhalation, and dermal risk in males and females.

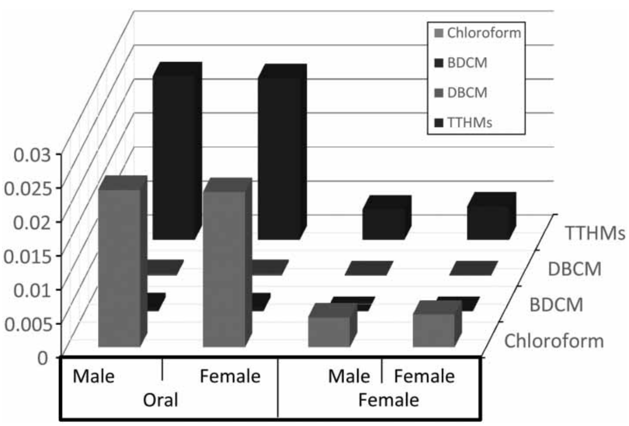

Figure 6 | Oral and dermal hazard index of THMs in drinking water samples of male and females.

Cancer risk analysis of THMs through different routes

Cancer risk through oral ingestion had been calculated for both males and females. The lifetime cancer risk was estimated from all possible routes of exposure using Equations (1–4) (Table 2). The average lifetime cancer risk posed by three THMs (CF, BDCM, and DBCM) via three exposure routes was calculated as 1.46×10^{-3}, 2.02×10^{-3}, 0.88×10^{-3} and 8.15×10^{-3}, 2.45×10^{-3}, 1.56×10^{-3} for both males and females (Tables 3 and 4) respectively. Ingestion was found to be the most prominent exposure pathway which contributed 65–85% to total cancer followed by halation and dermal absorption.

Table 2 | Physico-chemical quality parameters of drinking water samples

No.	Sampling point	Temp	pH	TDS	Turbidity	Conductivity	TOC	Free Cl	Monochloramines	Dichloramines	Total chlorine	$CHCl_3$	$CHClBr_2$	$CHCl_2Br$	$CHBr_3$	TTHMs
1	UGT # E-9 sector, Islamabad	15.5	7.5	332	0.91	671	5.1	0.1	0.1	0.1	0.3	105	6.55	9.15	BDL	120.7
2	H # D 22/6 E-9 sector, Islamabad	13.9	7.3	250	0.92	601	1.3	0.14	0.1	0.07	0.31	150	7.4	12.4	BDL	169.84
3	H # D 22/3 E-9 sector, Islamabad	12.1	7.6	389	1.1	501	4.2	0.11	0.06	0.05	0.22	321	9.6	16.88	7.78	355.26
4	H # D 21/6 E-9 sector, Islamabad	11.9	7.4	264	0.72	524	5.1	0.33	0.15	0.08	0.56	360.6	2.22	9.95	BDL	387.47
5	AHQ E-9 sector, Islamabad	13.2	7.5	230	0.89	456	3.5	0.12	0.14	0.04	0.3	137	1.55	3.83	BDL	142.38
6	Filtration plant, E-9, Islamabad	11.7	7.4	220	1	403	1.4	0.15	0.11	0.08	0.34	106.7	4.84	8.66	BDL	120.2
7	Filtration plant, E-8	13.4	7.4	210	0.88	472	1.7	BDL	0.09	0.6	0.69	193.8	1.45	8.5	BDL	203.7
8	H # 22, E-8, Islamabad	12.8	7.4	206	0.79	447	3.8	0.7	0.43	BDL	1.13	295	2.75	15.45	BDL	313.2
9	H # 25, E-8, Islamabad	14.6	7.3	367	1.23	416	5.1	0.06	0.09	0.05	0.2	575.9	1.59	15.16	3.11	595.86
10	H # 316, St # 39 G-9/1, Islamabad	11.3	7.3	210	1.55	415	0.8	0.17	0.3	0.02	0.49	280.2	15.81	22.44	BDL	318.47
11	H # 298, St # 39 G-9/1, Islamabad	15.1	7.1	208	0.66	443	8.9	0.09	0.04	0.06	0.19	355	1.65	8.11	BDL	364.76
12	H # 35, St # 8,F-7, Islamabad	13.1	7.4	215	0.53	513	0.5	BDL	0.04	0.02	0.06	96	3.67	7.98	BDL	107.65
13	Wasa office, Islamabad	12.6	7.6	233	0.76	407	0.4	0.21	BDL	BDL	0.21	314	1.09	22.09	BDL	337.18
14	H # 17-C, F-8, Islamabad	11.5	7.6	222	0.77	343	3	BDL	0.3	0.7	1	23.87	BDL	0.55	BDL	24.42
15	H # 6, street 66, F-7, Islamabad	12.5	7.6	234	0.55	403	1.4	BDL	0.05	0.03	0.08	55.66	1.84	3	BDL	60.5
16	H # 283, Gomal road, E-7, Islamabad	13.7	7.6	257	0.53	524	7	0.55	0.07	0.09	0.71	171.1	5.96	7.83	BDL	184.92
17	H # 309 Aurangzeb road, E-7, Islamabad	12.6	7.5	298	0.58	459	3.3	0.11	0.14	0.04	0.29	130.4	BDL	2.09	BDL	132.52

(*continued*)

Table 2 | continued

No.	Sampling point	Temp	pH	TDS	Turbidity	Conductivity	TOC	Free Cl	Monochloramines	Dichloramines	Total chlorine	$CHCl_3$	$CHClBr_2$	$CHCl_2Br$	$CHBr_3$	TTHMs
18	H # 205 Hill Side road, E-7, Islamabad	12.7	7.4	221	0.69	406	1	BDL	0.06	BDL	0.06	40.69	1.84	1.98	BDL	44.51
19	Filtration plant F-10, Islamabad	11.8	7.5	202	0.79	398	0.1	0.18	0.4	BDL	0.58	232.4	13.53	33.21	4.11	283.25
20	H # 202B Street 10, E-7, Islamabad	12.1	7.4	210	0.82	502	5.1	0.34	0.14	0.13	0.61	417.7	1.11	10.11	BDL	428.88
21	Filtration plant F-7, Islamabad	13.1	7.5	208	0.98	467	0.2	0.27	0.15	BDL	0.42	100.7	4.93	7.11	BDL	112.74
22	Treatment plant Simly Dam, Islamabad	12.2	7.4	223	1.05	416	8.2	0.82	0.21	0.15	1.18	144.5	0.69	5.64	BDL	150.83
23	Treatment plant Rawal Dam, Islamabad	13	7.4	219	0.99	505	4.6	0.19	BDL	0.06	0.25	103.6	3.14	7.87	BDL	114.57
24	Filtration plant Chaklala Base, Islamabad	11.3	7.5	265	0.87	517	0.1	0.14	0.12	BDL	0.26	247.1	13.24	13.98	BDL	274.32
25	Filtration plant Scheme III, Islamabad	12.1	7.1	232	0.89	508	0.1	0.15	0.09	BDL	0.24	258	17.55	18.64	BDL	294.19
26	Filtration plant Askari 4, Rawalpindi	12.6	7.3	203	0.75	426	6.9	0.48	0.11	0.07	0.66	172	7.32	10.65	BDL	189.97
27	H # 21-C Askari 4 Rawalpindi	11.8	7.6	242	0.79	411	0.1	0.14	0.08	BDL	0.22	271	18.21	23.65	5.72	189.97
28	Filtration plant Askari 3 Rawalpindi	13.4	7.5	215	0.97	343	5.9	0.44	0.2	0.1	0.74	382	1.99	14.02	BDL	398.02
29	H # 31-D Askari 3Rawalpindi	12.9	7.5	201	0.66	445	4	0.07	0.08	0.03	0.18	415.1	2.11	14.09	BDL	431.26
30	Filtration plant Askari 2 Rawalpindi	13.1	7.5	231	0.72	409	1.5	BDL	0.14	0.5	0.64	182	9.01	16.21	BDL	207.22

Table 3 | Cancer risk assessment for males ($\times 10^{-3}$ mg/kg.day)

	THMs				CF				BDCM				DBCM				
Site	**CF**	**BDCM**	**DBCM**	**TTHMs**	**Ing**	**Inh**	**Der**	**Total**	**Ing**	**Inh**	**Der**	**Total**	**Ing**	**Inh**	**Der**	**Total**	**TTHM**
1	105	9.15	6.55	120.7	3.08	3.68	0.70	0.12	0.27	0.32	0.03	0.06	0.19	0.23	0.05	0.04	0.22
2	150	12.4	7.4	169.84	0.00	5.25	1.00	0.03	0.36	0.43	0.04	0.08	0.22	0.26	0.05	0.05	0.16
3	321	16.88	9.6	347.48	0.01	11.24	2.13	0.07	0.50	0.59	0.05	0.11	0.28	0.34	0.07	0.06	0.24
4	360.6	9.95	2.22	372.76	0.01	12.63	2.39	0.08	0.29	0.35	0.03	0.07	0.07	0.08	0.02	0.01	0.15
5	137	3.83	1.55	142.38	0.00	4.80	0.91	0.03	0.11	0.13	0.01	0.03	0.05	0.05	0.01	0.01	0.06
6	106.7	8.66	4.84	120.2	0.00	3.74	0.71	0.02	0.25	0.30	0.03	0.06	0.14	0.17	0.03	0.03	0.11
7	193.8	8.5	1.45	203.7	0.01	6.78	1.29	0.04	0.25	0.30	0.03	0.06	0.04	0.05	0.01	0.01	0.11
8	295	15.45	2.75	313.2	0.01	10.33	1.96	0.06	0.45	0.54	0.05	0.10	0.08	0.10	0.02	0.02	0.18
9	575.9	15.16	1.59	592.63	0.02	20.16	3.82	0.12	0.45	0.53	0.05	0.10	0.05	0.06	0.01	0.01	0.23
10	280.2	22.44	15.81	318.47	0.01	9.81	1.86	0.06	0.66	0.79	0.07	0.15	0.46	0.55	0.11	0.10	0.31
11	355	8.11	1.65	364.76	0.01	12.43	2.36	0.07	0.24	0.28	0.02	0.05	0.05	0.06	0.01	0.01	0.14
12	96	7.98	3.67	107.65	0.00	3.36	0.64	0.02	0.23	0.28	0.02	0.05	0.11	0.13	0.03	0.02	0.10
13	314	22.09	1.09	337.18	0.01	10.99	2.08	0.07	0.65	0.77	0.07	0.14	0.03	0.04	0.01	0.01	0.22
14	23.87	0.55	0	24.42	0.00	0.84	0.16	0.00	0.02	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.01
15	55.66	3	1.84	60.5	0.00	1.95	0.37	0.01	0.09	0.11	0.01	0.02	0.05	0.06	0.01	0.01	0.04
16	171.1	7.83	5.96	184.92	0.01	5.99	1.14	0.04	0.23	0.27	0.02	0.05	0.18	0.21	0.04	0.04	0.13
17	130.4	2.09	0	132.52	0.00	4.57	0.87	0.03	0.06	0.07	0.01	0.01	0.00	0.00	0.00	0.00	0.04
18	40.69	1.98	1.84	44.51	0.00	1.42	0.27	0.01	0.06	0.07	0.01	0.01	0.05	0.06	0.01	0.01	0.03
19	232.4	33.21	13.53	279.14	0.01	8.14	1.54	0.05	0.98	1.16	0.10	0.22	0.40	0.47	0.09	0.09	0.35
20	417.7	10.11	1.11	428.88	0.01	14.62	2.77	0.09	0.30	0.35	0.03	0.07	0.03	0.04	0.01	0.01	0.16
21	100.7	7.11	4.93	112.74	0.00	3.53	0.67	0.02	0.21	0.25	0.02	0.05	0.14	0.17	0.03	0.03	0.10
22	144.5	5.64	0.69	150.83	0.00	5.06	0.96	0.03	0.17	0.20	0.02	0.04	0.02	0.02	0.00	0.00	0.07
23	103.6	7.87	3.14	114.57	0.00	3.63	0.69	0.02	0.23	0.28	0.02	0.05	0.09	0.11	0.02	0.02	0.09
24	247.1	13.98	13.24	274.32	0.01	8.65	1.64	0.05	0.41	0.49	0.04	0.09	0.39	0.46	0.09	0.08	0.23
25	258	18.64	17.55	294.19	0.01	9.03	1.71	0.05	0.55	0.65	0.06	0.12	0.52	0.61	0.12	0.11	0.29
26	172	10.65	7.32	189.97	0.01	6.02	1.14	0.04	0.31	0.37	0.03	0.07	0.21	0.26	0.05	0.05	0.15
27	271	23.65	18.21	312.86	0.01	9.49	1.80	0.06	0.69	0.83	0.07	0.16	0.53	0.64	0.13	0.12	0.33
28	382	14.02	1.99	398.02	0.01	13.38	2.53	0.08	0.41	0.49	0.04	0.09	0.06	0.07	0.01	0.01	0.18
29	415.1	14.09	2.11	431.26	0.01	14.53	2.75	0.09	0.41	0.49	0.04	0.09	0.06	0.07	0.01	0.01	0.19
30	182	16.21	9.01	207.22	0.01	6.37	1.21	0.04	0.48	0.57	0.05	0.11	0.26	0.32	0.06	0.06	0.20

Ing, ingestion; Inh, inhalation; Der, dermal.

Table 4 | Cancer risk assessment for females ($\times 10^{-3}$ mg/kg.day)

	THMs				CF				BDCM				DBCM				
Site	**CF**	**BDCM**	**DBCM**	**TTHMs**	**Ing**	**Inh**	**Der**	**Total**	**Ing**	**Inh**	**Der**	**Total**	**Ing**	**Inh**	**Der**	**Total**	**TTHM**
1	105	9.15	6.55	120.7	3.80	3.63	0.76	0.14	0.33	0.32	0.03	0.06	0.24	0.23	0.05	0.05	0.25
2	150	12.4	7.4	169.84	5.43	5.18	1.09	0.20	0.45	0.43	0.04	0.09	0.27	0.26	0.06	0.05	0.34
3	321	16.88	9.6	347.48	11.62	11.09	2.33	0.43	0.61	0.58	0.06	0.12	0.35	0.33	0.07	0.07	0.62
4	360.6	9.95	2.22	372.76	13.06	12.46	2.62	0.49	0.36	0.34	0.03	0.07	0.08	0.08	0.02	0.02	0.57
5	137	3.83	1.55	142.38	4.96	4.73	0.99	0.18	0.14	0.13	0.01	0.03	0.06	0.05	0.01	0.01	0.22
6	106.7	8.66	4.84	120.2	3.86	3.69	0.77	0.14	0.31	0.30	0.03	0.06	0.18	0.17	0.04	0.03	0.24
7	193.8	8.5	1.45	203.7	7.02	6.69	1.41	0.26	0.31	0.29	0.03	0.06	0.05	0.05	0.01	0.01	0.33
8	295	15.45	2.75	313.2	10.68	10.19	2.14	0.40	0.56	0.53	0.05	0.11	0.10	0.09	0.02	0.02	0.52
9	575.9	15.16	1.59	592.63	20.85	19.89	4.18	0.78	0.55	0.52	0.05	0.11	0.06	0.05	0.01	0.01	0.89
10	280.2	22.44	15.81	318.47	10.15	9.68	2.03	0.38	0.81	0.78	0.07	0.16	0.57	0.55	0.12	0.11	0.64
11	355	8.11	1.65	364.76	12.86	12.26	2.58	0.48	0.29	0.28	0.03	0.06	0.06	0.06	0.01	0.01	0.55
12	96	7.98	3.67	107.65	3.48	3.32	0.70	0.13	0.29	0.28	0.03	0.06	0.13	0.13	0.03	0.03	0.21
13	314	22.09	1.09	337.18	11.37	10.85	2.28	0.42	0.80	0.76	0.07	0.15	0.04	0.04	0.01	0.01	0.58
14	23.87	0.55	0	24.42	0.86	0.82	0.17	0.03	0.02	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.04
15	55.66	3	1.84	60.5	2.02	1.92	0.40	0.08	0.11	0.10	0.01	0.02	0.07	0.06	0.01	0.01	0.11
16	171.1	7.83	5.96	184.92	6.20	5.91	1.24	0.23	0.28	0.27	0.03	0.05	0.22	0.21	0.05	0.04	0.33
17	130.4	2.09	0	132.52	4.72	4.51	0.95	0.18	0.08	0.07	0.01	0.01	0.00	0.00	0.00	0.00	0.19
18	40.69	1.98	1.84	44.51	1.47	1.41	0.30	0.05	0.07	0.07	0.01	0.01	0.07	0.06	0.01	0.01	0.08
19	232.4	33.21	13.53	279.14	8.42	8.03	1.69	0.31	1.20	1.15	0.11	0.23	0.49	0.47	0.10	0.09	0.64
20	417.7	10.11	1.11	428.88	15.12	14.43	3.03	0.56	0.37	0.35	0.03	0.07	0.04	0.04	0.01	0.01	0.64
21	100.7	7.11	4.93	112.74	3.65	3.48	0.73	0.14	0.26	0.25	0.02	0.05	0.18	0.17	0.04	0.03	0.22
22	144.5	5.64	0.69	150.83	5.23	4.99	1.05	0.20	0.20	0.19	0.02	0.04	0.02	0.02	0.01	0.00	0.24
23	103.6	7.87	3.14	114.57	3.75	3.58	0.75	0.14	0.28	0.27	0.03	0.05	0.11	0.11	0.02	0.02	0.22
24	247.1	13.98	13.24	274.32	8.95	8.54	1.79	0.33	0.51	0.48	0.05	0.10	0.48	0.46	0.10	0.09	0.52
25	258	18.64	17.55	294.19	9.34	8.91	1.87	0.35	0.67	0.64	0.06	0.13	0.64	0.61	0.13	0.12	0.60
26	172	10.65	7.32	189.97	6.23	5.94	1.25	0.23	0.39	0.37	0.04	0.07	0.27	0.25	0.06	0.05	0.36
27	271	23.65	18.21	312.86	9.81	9.36	1.97	0.37	0.86	0.82	0.08	0.16	0.66	0.63	0.14	0.13	0.66
28	382	14.02	1.99	398.02	13.83	13.20	2.77	0.52	0.51	0.48	0.05	0.10	0.07	0.07	0.02	0.01	0.63
29	415.1	14.09	2.11	431.26	15.03	14.34	3.01	0.56	0.51	0.49	0.05	0.10	0.08	0.07	0.02	0.01	0.67
30	182	16.21	9.01	207.22	6.59	6.29	1.32	0.25	0.59	0.56	0.05	0.11	0.33	0.31	0.07	0.06	0.42

Ing, ingestion; Inh, inhalation; Der, dermal.

Table 5 | Hazard indices for males and females based on THMs

	Chloroform				BDCM				DBCM				Total			
	Male		Female		Male		Female		Male		Female		Male		Female	
Site	Oral	Dermal	Oral	Dermal	Oral	Dermal	Oral	Dermal	Oral	Dermal	Oral	Dermal	Oral	Dermal	Oral	Dermal
1	367.63	69.67	362.69	76.21	16.02	1.38	15.80	1.51	11.47	2.27	11.31	2.48	395.11	73.32	389.81	80.20
2	525.33	99.55	518.27	108.90	21.71	1.87	21.42	2.05	12.95	2.56	12.78	2.81	559.99	103.99	552.46	113.75
3	1123.90	212.98	1108.80	232.98	29.55	2.55	29.15	2.79	16.81	3.33	16.58	3.64	1170.25	218.86	1154.53	239.41
4	1262.51	239.25	1245.55	261.71	17.42	1.50	17.18	1.65	3.89	0.77	3.83	0.84	1283.82	241.53	1266.57	264.20
5	479.67	90.90	473.23	99.43	6.70	0.58	6.61	0.63	2.71	0.54	2.68	0.59	489.09	92.02	482.52	100.65
6	373.58	70.80	368.56	77.44	15.16	1.31	14.96	1.43	8.47	1.68	8.36	1.83	397.22	73.78	391.88	80.71
7	678.36	128.55	669.25	140.62	14.88	1.29	14.68	1.41	2.54	0.50	2.50	0.55	695.78	130.34	686.44	142.58
8	1032.86	195.73	1018.99	214.11	27.05	2.34	26.68	2.56	4.81	0.95	4.75	1.04	1064.73	199.02	1050.42	217.70
9	2016.29	382.10	1989.20	417.96	26.54	2.29	26.18	2.51	2.78	0.55	2.75	0.60	2045.62	384.94	2018.13	421.07
10	981.12	185.93	967.94	203.38	39.28	3.39	38.76	3.71	27.68	5.48	27.31	5.99	1048.08	194.80	1034.00	213.08
11	1242.94	235.54	1226.24	257.65	14.20	1.23	14.01	1.34	2.89	0.57	2.85	0.63	1260.03	237.34	1243.10	259.62
12	336.12	63.70	331.60	69.67	13.97	1.21	13.78	1.32	6.42	1.27	6.34	1.39	356.51	66.17	351.72	72.39
13	1099.39	208.34	1084.62	227.89	38.67	3.34	38.15	3.65	1.91	0.38	1.88	0.41	1139.97	212.06	1124.65	231.96
14	83.57	15.84	82.45	17.32	0.96	0.08	0.95	0.09	0.00	0.00	0.00	0.00	84.54	15.92	83.40	17.42
15	194.88	36.93	192.26	40.40	5.25	0.45	5.18	0.50	3.22	0.64	3.18	0.70	203.35	38.02	200.62	41.59
16	599.17	113.55	591.12	124.20	13.71	1.18	13.52	1.29	10.43	2.07	10.29	2.26	623.31	116.79	614.93	127.76
17	456.67	86.54	450.53	94.66	3.66	0.32	3.61	0.35	0.00	0.00	0.00	0.00	460.33	86.86	454.14	95.01
18	142.47	27.00	140.55	29.53	3.47	0.30	3.42	0.33	3.22	0.64	3.18	0.70	149.15	27.93	147.15	30.56
19	813.69	154.20	802.76	168.67	58.14	5.02	57.36	5.49	23.69	4.69	23.37	5.13	895.51	163.91	883.48	179.29
20	1462.33	277.12	1442.68	303.13	17.70	1.53	17.46	1.67	1.94	0.38	1.92	0.42	1481.97	279.03	1462.06	305.22
21	352.57	66.81	347.84	73.09	12.45	1.07	12.28	1.18	8.63	1.71	8.51	1.87	373.65	69.60	368.63	76.13
22	505.93	95.88	499.13	104.88	9.87	0.85	9.74	0.93	1.21	0.24	1.19	0.26	517.01	96.97	510.06	106.07
23	362.59	68.71	357.72	75.16	13.78	1.19	13.59	1.30	5.50	1.09	5.42	1.19	381.86	70.99	376.73	77.65
24	865.16	163.95	853.53	179.34	24.47	2.11	24.14	2.31	23.18	4.59	22.87	5.02	912.81	170.65	900.54	186.67
25	903.32	171.18	891.18	187.25	32.63	2.82	32.19	3.08	30.72	6.08	30.31	6.65	966.67	180.08	953.69	196.99
26	602.21	114.12	594.12	124.83	18.64	1.61	18.39	1.76	12.81	2.54	12.64	2.77	633.67	118.27	625.16	129.37
27	948.84	179.81	936.09	196.69	41.40	3.58	40.85	3.91	31.88	6.31	31.45	6.90	1022.12	189.70	1008.38	207.50
28	1337.51	253.46	1319.54	277.26	24.54	2.12	24.21	2.32	3.48	0.69	3.44	0.75	1365.53	256.27	1347.19	280.33
29	1453.22	275.39	1433.70	301.24	24.67	2.13	24.33	2.33	3.69	0.73	3.64	0.80	1481.58	278.25	1461.68	304.37
30	637.23	120.76	628.66	132.09	28.38	2.45	28.00	2.68	15.77	3.12	15.56	3.42	681.38	126.33	672.22	138.19

This sequence of exposure was found to be in agreement with previous studies carried out by Lee et al. (2004), whereas Pardakhti et al. (2011) reported inhalation to be the major route of exposure. Among THMs species, chloroform presented the higher cancer risk due to its highest concentration in all the water samples, which was followed by BDCM and DBCM. These findings were similar to other reports (Viana et al. 2009; Karim et al. 2011). Chloroform imparted the lowest cancer risk among all the THMs in both males and females through oral ingestion which is higher by about 163 in females and 140 in males than the acceptable level (10^{-6}). The average cancer risk for THMs for both males and females was in the order of bromoform, BDCM, DBCM, and chloroform (high to low). Hazard indices for both and female are shown in Table 5. According to the current study, females were found to be more susceptible to cancer risk compared to males in all cases. These results clearly indicated that public drinking water supplies pose a serious cancer threat. These results were much higher than those found in studies reported by other researchers (Wang et al. 2007; Karim et al. 2011; Pardakhti et al. 2011). Thus, the higher values of cancer risk may contribute to potential life-threatening diseases among the exposed population (Viana et al. 2009).

In this study, both BDCM and DBCM are found to be significant contributors to total cancer risk which is, to a certain extent, similar to the results obtained by Uyak (2006), who reported DBCM to be the principal component of total cancer risk through the oral route. Chloroform was found to be, if not the main, a significant contributor to cancer risk, but in one study it was found to be the lowest, which may be due to the predominance of Br-THMs over Cl^- THMs in the studied water carried out by Basu et al. (2011), which is contrary to this research.

Rawalpindi and Islamabad have a population of nearly three million people (Demographia 2011) with an assumed average age of 70 years. The lifetime cancer cases for both males and females, based on average TTHM risk (1.48×10^{-3}) and 3 million people, could be expected to be up to 254 cases. This scenario suggests that approximately three to four cases of cancer could be expected each year, and consequently the number of cancer cases among females being more than that of males according to this study. According to this scenario we can make an assumption that one-third of the three million population lives in the target cities. We expect approximately 120 and 150 lifetime cancer cases among males and females.

CONCLUSION

The current study compared DBPs occurrence, formation, and their health risk among the male and female population in different parts of the twin cities. The statistics indicated that most of the samples were contaminated. The applied analytical method including SPME for sampling THMs from water and determination by GC-ECD was successfully applied.

The study of THMs from the distribution system of both municipalities revealed that the maximum concentration of TTHMs and chloroform was 575 μg/L and 595 μg/L, respectively, which is above the prescribed limit of USEPA drinking water quality standards of 80 and 70 μg/L.

Unfortunately, nearly 95% samples were contaminated with THMs. In such circumstances, where DBP values are narrowly approached or exceed the standard values, water authorities need to evaluate water treatment practices with a view to improving the elimination of organic contents of the water sources prior to disinfection as well as reducing water age in distribution systems. The average lifetime cancer risk for males and females was 0.51×10^{-3} and 1.22×10^{-3}, respectively. The expected number of cancer cases per year could reach two to three cases for each city. The highest risk from THMs seems to be from the inhalation route followed by ingestion and dermal contact.

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