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Human Health Risk Assessment of Trihalomethane through Multi-Pathway Exposure from Drinking Water of Baghdad, Iraq

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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 Baghdad are estimated. The USEPA risk assessment model approach as the hazard quotients (HQ) of CF, BDCM and DBCM in the investigated distribution networks is exceeded the WHO guideline value for several sites but the HQ of BF was within the WHO guideline value for all sites. The multi-pathway evaluations of lifetime cancer risks by 4 THMs (CF, BDCM, DBCM and BF) via three exposure routes (oral, dermal and inhalation) was 1.06×10-4, 0.87×10-4, 1.10×10-4 and 1.50×10-4 respectively, which exceeded the USEPA range of concern limit of 1.0×10-6. However, the DBCM was the higher cancer risk compare with the other THMs, and BF was the lower cancer risk.

***Keywords**: Water, Baghdad, Multipathway, Risk assessment, Trihalomethanes.*

Introduction

Water is very important to our existence in life. Potable water is the water that is free from disease producing microorganisms and chemical substances that are dangerous to health (Lamikaran, 1999). However, chlorination, which is a widely used as a disinfection process, has been reported to cause the formation of trihalomethanes (THMs) (Rook, 1974). THMs are formed due to the reactions between chlorine and the natural organic matter in water supplies, especially surface waters. Chloroform (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform (TBM) are the four main THM compounds termed THMs. These groups of compounds have been implicated in liver and kidney defects, central nervous system problems and increased risk of carcinogenicity and mutagenicity as Class B carcinogens (USEPA, 1990). Many cancer risk assessments have been conducted on DBPs, but most of them conveyed the risk as the excess cancer incidence through lifetime exposure (Lee et al., 2004; Uyak, 2006; Wang et al., 2007; Chowdhury and champagne, 2009; Chowdhury et al., 2011; Amjad et al., 2013; Gan et al., 2013).

Typical exposure routes to THMs in tap water are ingestion by the oral route, inhalation through breathing and dermal contact through skin during regular indoor activities, such as showering, bathing and cooking. 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 considered in the analysis of total human exposure to volatile contaminants in tap water (Weisel and Jo, 1996; Weisel et al., 1999; Hsu et al., 2001; Lee et al., 2004; Uyak, 2006; Mallika et al., 2008).

Therefore, the purpose of this study is to calculate the amount of exposure to THMs and the life time cancer risk by a multi pathway exposure assessment of selected public drinking waters among population in the study area of Baghdad.

Materials and Methods

Study area and sample collection

Study area

This study was examined two water treatment plants in Baghdad, as shown in figure1. Al-Wahda Treatment Plant which located at N 33º 17' 27 .73, E 44º 26' 38 .28, on the eastern bank of the Tigris River in the neighborhood of The General Company for Vegetable Oils Industry at the entrance of Al-Musbah Street. It was established in 1959. The operation capacity of the plant 27000m^3/day. Al-Qadisiyah Treatment Plant which plant located at N 33º 16' 00 .67, E 44º 22' 14 .8, on the western

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bank of the Tigris River in the region of Al-Qadisiyah. It was established in 1965 with design capacity 20 million gallons per day, then established new production line in 1975 with design capacity 10 million gallons per day. Maintenance of filters and pumps was in 2010. The operation capacity of the plant was 96000m^3/day. These two plants were containing two production lines which merged as one line and out of plant as one production line. Chlorine gas was used to disinfection of water.

Water sampling

The samples of drinking water were collected from the study sites from July 2015 to April 2016 for DBPs (THMs) examinations in Al-Wahda and Al-Qadisiyah plants within 2-3 days of each plant sequentially. Sites were distributed among residential areas. Distribution based on the distance from the plant from the nearest to the farthest point which feeding from the plant with drawn to cover plant. The points covered Al-Wahda plant is: W3 the middle point of water distribution network and W4 the farthest point of water distribution network from the plant. Where the points covered Al-Qadisiyah plant is Q3 the middle point of water distribution network and Q4 the farthest point of water distribution network from the plant. All samples were taken with three replicates seasonally: summer, autumn, winter and spring.

Water sampling for TTHMs tests

Samples were collected in Clean glass bottles (100 ml) with plastic screw caps and Teflon rubber and closed tightly under water to avoid any bubbles and taken to the laboratory within 2-3 hours in cooling box to conduct the tests (APHA, 2012).

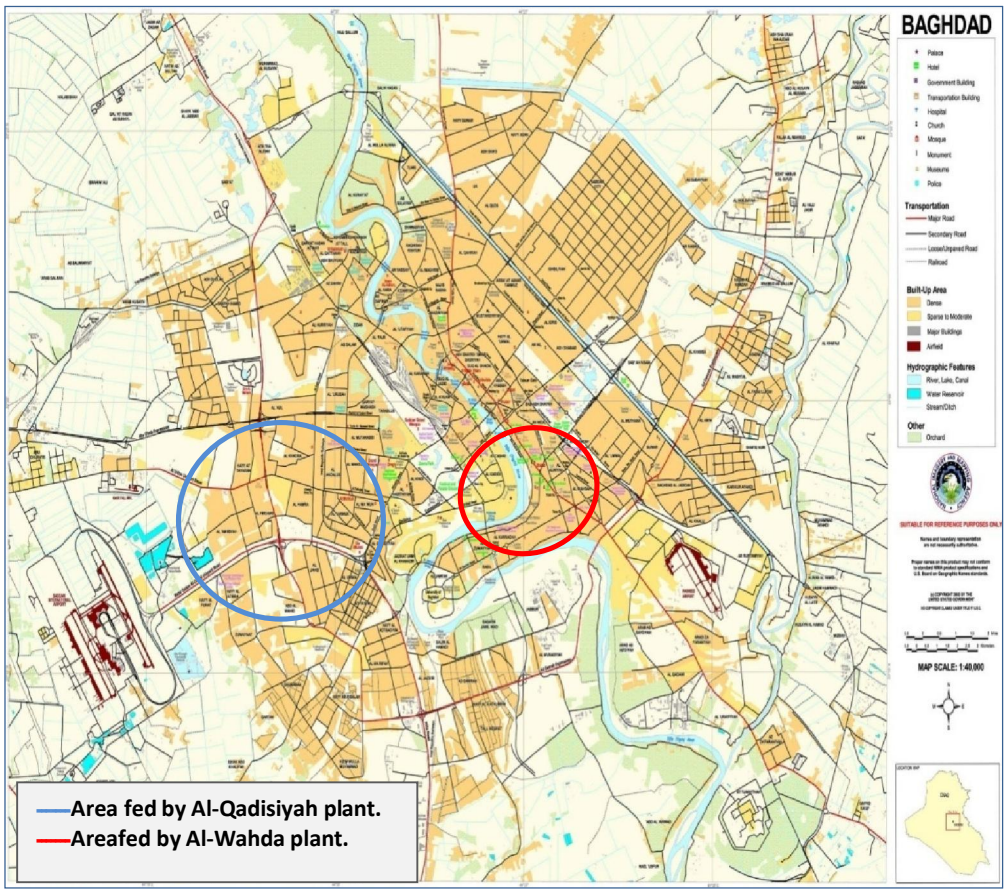

Figure 1. The study area that fed by both Al-Wahda and Al- Qadisiyah plants within the confines of Baghdad www.lib.utexas.edu

Analytical methods

This test was conducted in the Ministry of Science and Technology, Environmental and Water Directorate, Gas chromatography (GC) fitted with an electron capture detector (ECD) detector and a headspace sampler AOC- 5000 was used for the determination of four THMs.

The principle of the method is that the sample is placed in a sealed vial and allowed to equilibrate with its headspace vapour at 70ºC. Then the sample aliquot is taken from the vial headspace using a special syring and injected into the GC using a 0.25 mm i.d. 60 meter length fused silica capillary column. This method is derived from the UK reference booklet entitled "Determination of very low concentration of hydrocarbons and halogenated hydrocarbons in water" (1984-5), Methods for the Examination of Waters and Associated Materials, London Her Majesty's Stationary Office, ISBN (0117520047). The calibration graph was derived from a THM standard ampoule 1 ml mixture 2000 µg/ml each THM in methanol from Sigma-Aldrich or SUPELCO.

Risk assessment methodology

In this study, two approved risk assessment models were 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 ≤ 1 for compliance with WHO guidelines and was calculated as follows:

$$I_{WHO} = \frac{C_{CF}}{GV_{CF}} + \frac{C_{BDCM}}{GV_{BDCM}} + \frac{C_{DBCM}}{GV_{DBCM}} + \frac{C_{BF}}{GV_{BF}} \leq 1$$

Where C is the concentration of each THM in this study, and GV is the WHO guideline values have been established. The GV for CF is 200, BDCM 60, DBCM 100 and BF 100, all in mg/l (WHO, 2011).

The USEPA- approved Risk Assistant model which adopted by many researchers (Semerjian and Dennis 2007; Wang *et al*. 2007; Pardakhti, *et al*., 2011; Ferreira and Cunha, 2012; Karim, *et al*. 2013). The USEPA Risk assessment model is capable of estimating (1) Toxicological risks (toxic and non-carcinogenic risks) and Carcinogenic risks.

Toxicological risks, expressed as the hazard quotient (HQ), were calculated based on the comparison of actual exposure to the reference dose (RfD) as follows:

HQ = (Total amount ingested / body weight × exposure time × RfD)

Reference doses were extrapolated from toxicological studies of exposure that demonstrate a critical effect. They are expressed in units of mg/kg/day, and are available in the Integrated Risk Information System database (IRIS, 2009) database maintained by the USEPA (USEPA 2006).

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 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 1026 i.e.1:1,000,000 (USEPA, 2003).

The following relationships were used to calculate the cancer risks for THMs through ingestion, dermal absorption, and inhalation (Semerjian and Dennis 2007; Wang *et al*. 2007; Pardakhti, *et al*., 2011; Karim, *et al*. 2013).

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

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

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

Where $LADD_{oral}$ = (total amount ingested / body weight × life time)

= (Conc. THM in water × IR × EF × ED) / (BW× AT)

$LADD_{dermal}$ = (Conc.THM in water ×SA × PC×ET×EF× ED) / (BW×AT)

$LADD_{inhalation}$ = (Conc.CF in water ×AA × VF ×ET ×EF ×ED) / (BW× AT)

The lifetime cancer risk for people living in area study was calculated using the input parameters in Table (1) and the THM concentrations measured in this study.

Table (2) summarizes the reference doses (RfD), cancer group classifications for the THM components and cancer slope factors (CSF) for oral, dermal, and inhalation used for THM via different routes (RAIS, 2009).

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 (IRIS, 2009). For the 4THM components, the USEPA range of concern is for an increased carcinogenic risk of 1:1,000,000 (USEPA, 2003).

Table 1. Input factors and abbreviations for exposure assessment (Semerjian & Dennis, 2007).

Input parameter	Unit	Value	Reference
THM Conc. in water (C)	mg/l	See tables	This study
Exposure rate (ER)	L/day	2.0	US EPA (1997)
Exposure frequency (EF)	days/ year	365	Lee *et al*. (2004)
Exposure duration (ED)	year	70	US EPA (1997)
Average exposure time (AT)	days/year	70 × 365	Lee *et al*. (2004)
Body weight (BW)	kg	70	Lee *et al*. (2004)
Surface area (SA)	M^2	1.8	US EPA (1997)
Exposure time (ET)	min/day	35 min	RAIS (2009)
Aspirated air (AA)	m^3 per day	20 =0.83333m^3/hr	Semerjian and Dennis (2007)
Volatilization factor for chloroform (VF)	L/ m^3	0.5	Semerjian and Dennis (2007)
Permeability Coefficient (PC)	cm/h	0.00683 (CF) 0.00402(BDCM) 0.00289(DBCM) 0.00235(BF)	RAIS (2009)

Multi-pathways (oral, dermal, and inhalation) exposures were considered in the cancer risk assessment of THM in some of Baghdad population and were estimated based on the exposure factors in Tables (1) and (2).

Table (2) Carcinogenic slope factors (CSF), reference doses (RfD) and cancer group classifications for THM components (RAIS, 2009).

Chemicals	Cancer groups	Carcinogenic slope factors (CSF) (mg/kg day)		Reference dose (RfD)
		Oral/dermal	Inhalation	(mg/kg day)
CF	B1	3.1×10^{-2}	8.05×10^{-5}	0.01
BDCM	B2	6.2×10^{-2}		0.02
DBCM	C	8.4×10^{-2}		0.02
BF	B2	7.9×10^{-3}		0.02

B1: probable human carcinogen with limited human data.
B2: probable human carcinogen with sufficient animal data.
C: possible human carcinogen.

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 some of Baghdad region resulted inI_{WHO} values of less than 1 for all samples collected from the middle and farthest point of

distribution networks from both of plants. Computed I_{WHO} values ranged between 0.448–0.796, 0.352–0.510, 0.331–0.702 and 0.668–0.693 for samples collected during summer, autumn, winter and spring, respectively (table3).

The additive toxicity of recorded THM levels in the distribution networks of investigated sources is coincident with the (WHO, 2011) guideline value less than 1. Hence such concentrations do not pose any adverse toxic health impacts. Calculated I_{WHO} values for network THM levels recorded for individual locations are summarized in Table3. The highest I_{WHO} value of 0.796 and lowest I_{WHO} value of 0.331 were recorded in Al-Wahda treatment plants during summer and winter respectively.

The increase of I_{WHO} in summer at Al-Wahda plants due to its location in the south of city and the high temperature in summer that lead to polluted of raw water by organic matter which reacted with more amounts of alum and additive chlorine in treatment plant to precipitate and disinfect of water.

Table (3) Computed WHO additive toxicity values for network THM concentration

Variables	Season	Al-Wahda plant		Al-Qadisiyah plant	
		W3	W4	Q3	Q4
I_{WHO}	**Summer**	0.796	0.687	0.509	0.448
	Autumn	0.510	0.417	0.354	0.352
	Winter	0.390	0.331	0.702	0.693
	Spring	0.688	0.668	0.693	0.680

The USEPA risk assistant model approach

The hazard quotient (HQ) is calculated based on the comparison of actual exposure to the reference dose (RfD). The HQ estimations, ingestion was the only exposure route considered for a water consumption rate of 2 liters /day.

The HQ_{CF} value ranging between 0.013-0.058 during winter and summer respectively in Al-Wahda plant, the HQ_{BDCM} values ranged between 0.013-0.031 during winter and summer respectively in Al-Wahda plant, the HQ_{DBCM} ranged between 0.015-0.041during autumn at Al-Qadisiyah and summer at Al-Wahda respectively and the HQ_{BF} values ranged between 0.003-0.008 in several site for both plants (Table4) and (fig. 2,3,4,5).

The highest HQ value was CF and the lowest HQ value was BF during all seasons, so we can say that CF has more risk in health impacts from other components.

Table 4. Estimated non-carcinogenic risks of surveyed network THM levels in some of Baghdad population for a consumption rate of 2 liters/day

Variables	Season	Al-Wahda plant		Al-Qadisiyah plant	
		W3	W4	Q3	Q4
HQ_{CF}	**Summer**	0.051	0.058	0.032	0.036
	Autumn	0.035	0.029	0.029	0.025
	Winter	0.018	0.013	0.053	0.043
	Spring	0.056	0.05	0.055	0.052
HQ_{BDCM}	**Summer**	0.031	0.029	0.021	0.019
	Autumn	0.021	0.018	0.015	0.015
	Winter	0.016	0.013	0.03	0.029
	Spring	0.03	0.03	0.03	0.03
HQ_{DBCM}	**Summer**	0.041	0.031	0.025	0.02
	Autumn	0.024	0.019	0.015	0.016
	Winter	0.02	0.017	0.034	0.032
	Spring	0.031	0.029	0.031	0.03
HQ_{BF}	**Summer**	0.008	0.005	0.004	0.004
	Autumn	0.004	0.004	0.003	0.003
	Winter	0.005	0.005	0.004	0.008
	Spring	0.003	0.004	0.004	0.004

The toxicity of recorded THMs levels of CF, BDCM and DBCM in the investigated distribution networks is exceeded the guideline value of WHO(2011) for several sites, and consequently such concentrations have adverse toxic and non-carcinogenic risks in health impacts, but the HQ value of BF was within the WHO guideline value for all sites.

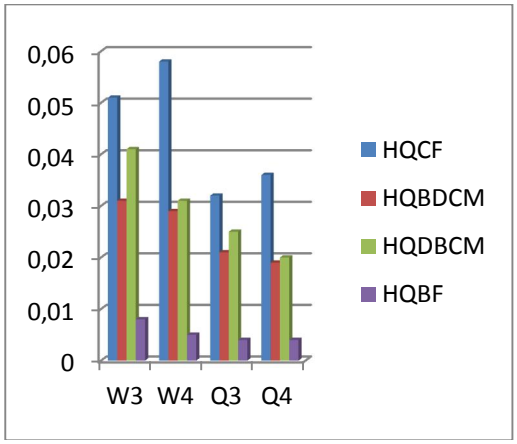

Figure 2. HQ values for 4THMs in summer

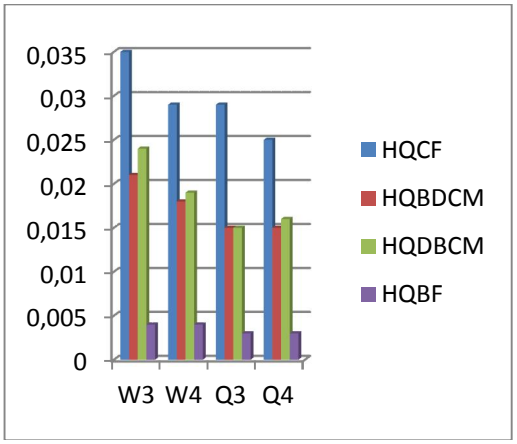

Figure 3. HQ values for 4THMs in autumn

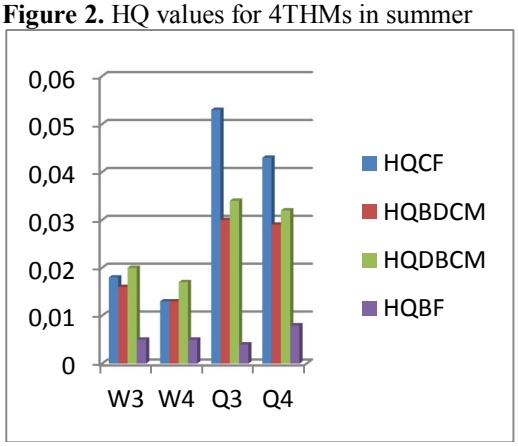

Figure 4. HQ values for 4THMs in winter

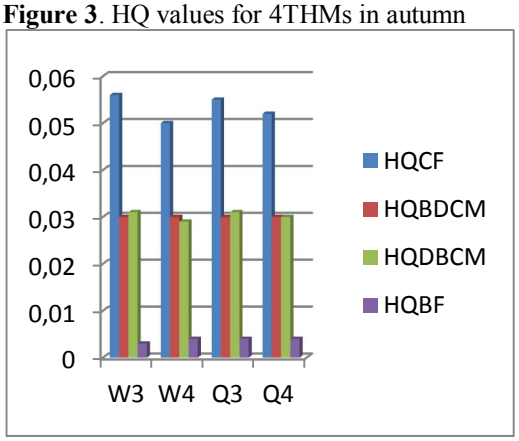

Figure 5. HQ values for 4THMs in spring

Table (5. The average lifetime cancer risk posed by 4THMs via three exposure routes

Variables	Season	cancer risk (10^{-4})				
		Summer	**Autumn**	**Winter**	**Spring**	**Annual**
CF	Oral	0.14	0.091	0.098	0.16	
	Inhalation	0.016	0.011	0.011	0.02	
	Dermal	0.029	0.19	0.21	0.35	
	Total	**0.185**	**0.292**	**0.319**	**0.53**	**0.3315**
BDCM	Oral	0.3	0.21	0.27	0.37	
	Inhalation	ND	ND	ND	ND	
	Dermal	0.039	0.027	0.034	0.047	
	Total	**0.339**	**0.237**	**0.304**	**0.417**	**0.3242**
DBCM	Oral	0.49	0.31	0.43	0.51	
	Inhalation	ND	ND	ND	ND	
	Dermal	0.044	0.028	0.039	0.046	
	Total	**0.534**	**0.338**	**0.469**	**0.556**	**0.4742**
BF	Oral	0.0082	0.0053	0.0086	0.0060	
	Inhalation	ND	ND	ND	ND	
	Dermal	0.00060	0.00039	0.00064	0.00044	
	Total	**0.0088**	**0.0056**	**0.0092**	**0.0064**	**0.0038**
TTHM	**Total**	**1.0668**	**0.8726**	**1.1012**	**1.5094**	**1.1337**

ND= Not Detected

Carcinogenic risks for THM

Multi-pathway evaluations of lifetime cancer risks for THM

The exposure metric used for carcinogenic risk assessment is the Lifetime Average Daily Dose (LADD). The average lifetime cancer risk posed by four THMs (CF, BDCM, DBCM

and BF) via three exposure routes was 1.06×10^{-4}, 0.87×10^{-4}, 1.10×10^{-4} and 1.50×10^{-4} for summer, autumn, winter and spring, respectively(table 5). The highest cancer risk was in spring then summer, winter and autumn respectively(Fig 6). DBCM was the higher cancer risk from other THMs, and BF was the lower cancer risk(Fig 7). Ingestion was found to be the most prominent exposure pathway followed by dermal absorption and inhalation.

The results showed that the lifetime cancer risk for the 4THM components are 1.13×10^{-4} which was higher than the USEPA range of concern limit of 1.0×10^{-6} (USEPA 2003), about 100 times (Table 5). That means approximately one of every 10,000 individuals in Baghdad could get cancer from the daily intake of water in his life span.

This result agrees with the study of Semerjian and Dennis (2007); Amjad *et al*. (2013); Ewaid (2015) and Siddique *et al*. (2015). These higher values of cancer risk may cause several diseases among the exposed population (Viana *et al*., 2009).

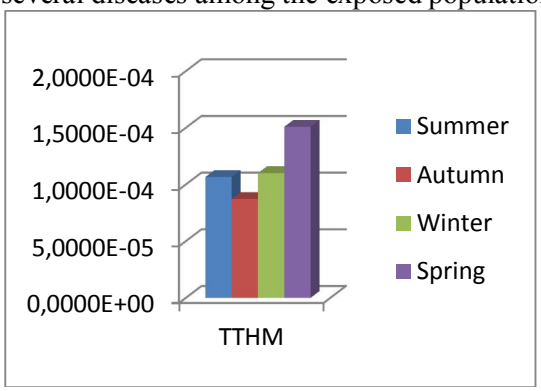

Figure 6. The average lifetime cancer risk posed by TTHMs via three exposure routes

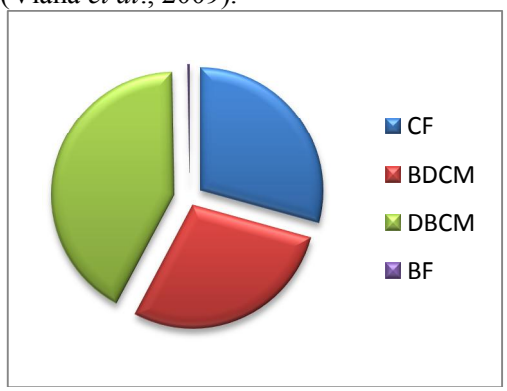

Figure 7. The annual percentage of the 4THMs components distribution in portable water of two plants

Conclusions

The concentration levels of THMs in drinking water samples from several Baghdad districts plants are generally within the allowable concentration recommended by the WHO and the Iraqi standards. The WHO additive toxicity of recorded 4THM levels in the distribution networks of investigated sources is coincident with the WHO guideline value (less than 1), such concentrations do not pose any adverse toxic health impacts.

The USEPA toxicity of recorded THMs levels of CF, BDCM and DBCM in the investigated distribution networks is exceeded the WHO guideline value for several sites, and consequently such concentrations have adverse toxic and non-carcinogenic risks in health impacts, but THMs levels of BF was within the WHO guideline value for all sites.

The lifetime cancer risk for the 4THM components via multi pathway exposure routes are 1.13×10^{-4} which was higher than the 1.0×10^{-6} which recommended by the USEPA, about 100 times. That means approximately one of every 10,000 Baghdad individuals could get cancer from the daily intake of drinking water in his life span.

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