Evaluation of Human Health Risks Associated with Exposure to Disinfection by-Products (Dbps) in Drinking Water of Wassit Province Southeast Iraq

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Abstract:

The toxicological risks and lifetime cancer risks associated with exposure to disinfection by-products (DBPs) including Haloacetic acids (HAAs) and trihalomethanes (THMs) compounds by drinking water in several districts in Wassit Province were estimated. The seasonal variation of HAAs and THMs compounds in drinking water have indicated that the mean values for total HAAs (THAAs) and total THMs (TTHMs) ranged from 43.2 to 72.4 mg/l and from 40 to 115.5 mg/l, respectively.

The World health organization index for additive toxicity approach was non-compliant with the WHO guideline value in summer and autumn seasons and this means that THMs concentration has adverse toxic health effects.

The multi-pathway of lifetime human health risk of cancer credited to THMs and HAAs in drinking water via three exposure routes for THMs and only one exposure route for HAAs was evaluated and found to be 6.13×10^{-4} and 1.78×10^{-4} respectively and these values were higher than the US.EPA range of concern limit of 1×10^{-6}. The risk ratio of THAAs to TTHMs was 3.44. Also, the highest cancer risk was recorded for BDCM followed by DBCM, CF, TCAA, DCAA, and BF.

Keywords: Disinfection by-products, Drinking water, HAAs, Health risks, THMs.

Introduction:

Water is a vital part of the food chain and its quality is a priority for human consumption. Drinking water is the water that is free from chemicals that are hazardous to public health and microorganisms producing disease (1).

Disinfection of drinking water is essential to eliminate pathogenic microorganisms and the chlorine is widely used in disinfection process due to its potency and relative ease of use. But at the same time, it reacts with natural organic matter (NOM) and/or inorganic substances in water forming various disinfection by-products (DBPs) like haloacetic acids (HAAs), trihalomethanes (THMs) and other undesirable compounds (2, 3).

The occurrence of DBPs in potable water has become a health issue having potential adverse effects on human health. Many of DBPs compounds have been implicated in kidney and liver defects, central nervous system problems and increased risk of carcinogenicity (4, 5). Among these products, THMs and HAAs are the most common and well-documented DBPs compounds in drinking water and they are generally considered as indicators of DBPs exposure in epidemiological examinations (6).

Several DPB components were classified by the United States Environmental Protection Agency (USEPA) depending on their carcinogenicity (7). As a result, four THM compounds {Chloroform (CF), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform (BF)} and two HAA compounds {Dichloroacetic acid (DCAA) and Trichloroacetic acid (TCAA)} are considered as probable or possible human carcinogenic.

Exposures to DBPs components may occur during a lifetime by three different routes which are oral, inhaling and dermal routes. These chronic exposures may impose various risks to human health (8, 9,10).

Therefore, the aim of this work is to assess the amount of four THMs and two HAAs species due to chronic exposure during a lifetime that may result in a cancer risk by a multi-pathway exposure evaluation of chosen public drinking water among the population in the study area of Wassit Province exposed to DBPs compounds.
Materials and Methods:

Water sampling

Five water treatment plants which are Al-Kut, Al-Karama, Al-Muwfaquia, Al-Haay, and Al-Bashaer within Wissit province were selected and subjected to the current study from January to December 2017. Each water plant was represented by randomly 3 locations situated within the residential sites. Drinking water sample was collected 3 times at each season and thus each location of each water plant has 12 water samples where each water sample was taken from tap water with three replications were taken from tap water of various homes after a flow of tap water for about 2-3 minutes. So, the size of this work was 540 water samples.

All drinking water samples were collected in glass bottles (100 ml) with plastic screw caps and Teflon rubber and ensure the bottles are free from any bubble. To prevent DBPs formation after sampling, a dechlorination solution (sodium thiosulphate 3%) was added to each bottle and stored in a cool box at 4°C and delivered to the laboratory which tested not more than 3 hours as suggested by (11) to be tested for THMs and HAAs content.

Analytical methods

Standard method of 6232B (11) was used to measure THMs using Gas chromatography (GC). The column was an HP-5 fused silica capillary column of 30 m×0.25 mm I.D. with 0.25 μm film thickness. The instrument temperature program was set to initial temperature of 35 °C with increasing temperature rate of 6 °C/min up to 180 °C. detector and Injector temperature were 250°C and 230°C, respectively. The carrier gas, nitrogen, was set in constant flow mode at 60 psi to the GC column. The calibration graph was derived from a THM standard ampoule 1 ml mixture 2000 μg/ml each THM in methanol.

HAAs were measured using a liquid-liquid microextraction gas chromatography (GC) according to USEPA Method 552.3 (12). The GC capillary column type ZB-1, 30m × 0. 25mm i.d., a 0.25μm film thickness. The instrument temperature program was set to 40-100 °C hold for 2 min at 10°C/min. Injector and detector temperature were 250 and 290, respectively. The calibration graph was derived from HAAs standard ampoule 1ml mixture 2000 μg/ml and each HAA in MTBE was stored in a cool box at 4 ̊C and delivered to the laboratory which tested not more than 3 hours as suggested by (11) to be tested for THMs and HAAs content.

Risk evaluation methodology

HAAs have nine kinds. But, the cancer potency information is only available for trichloroacetic acid (TCAA) and dichloroacetic acid (DCAA) (7, 13). So this study will be focused on four THMs and two HAAs being the most hazardous compounds of DBPs in drinking water for risk evaluation using adopted two approved risk assessment models. These models were firstly WHO index for additive toxicity (I\text{WHO}) guideline to estimate the toxic (non-carcinogenic and developmental) risk linked with chlorinated drinking water and calculated as follows:

\[
I_{\text{WHO}} = \frac{C(\text{CF})}{G(V(\text{CF}))} + \frac{C(\text{DBCM})}{G(V(\text{DBCM}))} + \frac{C(\text{DBCM})}{G(V(\text{DBCM}))} + \frac{C(\text{BF})}{G(V(\text{BF}))} \leq 1
\]

Where C represents the concentration of each THMs or HAAs in this study and GV is the WHO guideline values that have been established. The GV for CF is 200, DBCM is 60, DBCM is 100 and BF is 100 (1).

The second approved risk assistant model was the US.EPA approved risk assistant model which adopted by many researchers (14, 15, 16, 17). Carcinogenic risks resulted from the exposure to both THMs and HAAs concentrations were calculated using the US.EPA method.

Carcinogenic compounds are different from toxic compounds where there is no lower limit for risk existence. Thus, carcinogenic risk assessment models are generally being the risk that is proportional to total lifetime dose. The exposure metric used for carcinogenic risk assessment is the Lifetime Average Daily Dose (LADD). Depending on the DBP distributions, an exposure assessment was conducted to evaluate their potential intake during multiple pathways. THMs are volatile organic compounds which are known to have health risks via inhalation and dermal exposures during regular indoor activities which cannot be ignored as suggested by previous works (18, 19) while HAAs are non-volatile compounds (20). Therefore, ingestion, inhalation, and dermal contact exposures were measured for THMs whereas only ingestion (oral) exposure was measured for HAAs.

The following relationships were used to calculate the cancer risks for THMs and HAAs through ingestion, dermal contact, and inhalation following previous studies (20, 15, 21, 22, 17, 23). THMs or HAAs carcinogenic risk of oral route = LADD\text{oral} × CSF\text{oral}

THMs carcinogenic risk of dermal absorption = LADD\text{dermal} × CSF\text{oral}

THMs carcinogenic risk of inhalation = LADD\text{inhalation} × CSF\text{inhalation}

Where LADD is the Lifetime daily dose (mg/kg/day) and CSF is cancer slope factor (mg/kg/day) of THMs and HAAs species. The values assumed for CSF are summarized in Table 1. LADD\text{oral} = (C × IR × EF × ED) / (BW× AT)

Where: C is the THMs or HAAs concentration in drinking water (mg/l) in this study; IR is the drinking water ingestion rate of 2 L/day (24); EF is the exposure frequency 365 days/year (20); ED is the exposure duration, which was assumed to be 70 years based on (24); BW is body weight, which
averages 70 kg (20); AT is the Average exposure time \(70 \times 365\) days/year (20).

\[
\text{LADD}_{\text{dermal}} = \left( C \times \text{SA} \times PC \times \text{ET} \times \text{EF} \times \text{ED} \right) / (\text{BW} \times \text{AT})
\]

Where: SA is skin-surface area \(1.8\text{m}^2\) (24), PC is a permeability coefficient (cm/h) which was 0.00683, 0.00402, 0.00289, and 0.00235 for CF, BDCM, DBCM, and BF respectively (13), ET is exposure time 35/\text{min/day} (13).

\[
\text{LADD}_{\text{inhalation}} = \left( C_{\text{CF}} \times \text{AA} \times \text{VF} \times \text{ET} \times \text{EF} \times \text{ED} \right) / (\text{BW} \times \text{AT})
\]

Where: \(C_{\text{CF}}\) is the concentration of chloroform in drinking water, AA is aspirated air \(20\text{m}^3/\text{per day}\) (14), VF is volatilization factor for chloroform 0.5 L/\text{m}^3 (14).

**Table 1. The carcinogenic slope factors (CSF) of 4 THMs and 2 HAAs (13)**

| Chemicals   | Carcinogenic slope factors (CSF) (mg/kg day) |
|-------------|---------------------------------------------|
|             | Oral | Dermal | Inhalation |
| CF          | \(3.1 \times 10^{-2}\) | \(3.1 \times 10^{-2}\) | \(8.05 \times 10^{-3}\) |
| BDCM        | \(6.2 \times 10^{-2}\) | \(6.2 \times 10^{-2}\) | \(3.1 \times 10^{-2}\) |
| DBCM        | \(8.4 \times 10^{-2}\) | \(8.4 \times 10^{-2}\) | \(7.9 \times 10^{-3}\) |
| BF          | \(7.9 \times 10^{-3}\) | \(7.9 \times 10^{-3}\) |
| DCAA        | \(5 \times 10^{-2}\) |
| TCAA        | \(7 \times 10^{-2}\) |

**Results and Discussion:**

The seasonal mean values of the THMs and HAAs in drinking water sampled from each of water plant are shown in Table 2. The obtained results of total THMs (TTHMs) and total HAAs (THAAs) in drinking water sampled from different sites of each plant showed that the highest mean value was recorded in summer (115.5 mg/l and 72.4 mg/l respectively) in Al-Hayy plant drinking water while the lowest mean value was recorded in winter (40 mg/l and 43.2 mg/l respectively) in Al-Muwfaqia water plant. The TTHMs and THAAs concentrations during summer were higher than those of autumn, spring, and winter and this may be because of the increase of reaction rate between the NOM and added chlorine as water temperature increased while the increase in DBPs formation during summer may be due to the raw water quality (mostly the high TOC concentration in the summer season) and operational conditions such as increasing chlorine dose as reported by previous works (25, 26). Also, the study results have indicated that DCBM, DBCM, TCAA, and DCAA were the major constituent of THMs and HAAs in all samples. In case of comparing the obtained values of TTHMs with regulatory standards, none of drinking water samples have exceeded the permissible limit of Iraqi standards for drinking water (150 μg/l). But, many other water samples have exceeded the permissible limit of US.EPA (80 μg/l) particularly in summer and autumn. In Iraq, only DCAA and TCAA are regulated for drinking water at 50 and 100 mg/l respectively. In case of comparing these obtained values of DCAA and TCAA with Iraqi regulatory standards, none of the samples from the distribution systems have exceeded the regulated limits. Conversely, when comparing the values of total HAA obtained with US.EPA regulatory standards found that many samples have exceeded the regulated limits (60 μg/l). The results recorded in the present study have agreed with those of other studies (23, 27).

**Table 2. Seasonal-mean of THMs and HAAs concentration in Wassit districts drinking tap water of each plant during four seasons**

| Variables          | Season | Al-Kut | Al-Karama | Al-Muwfaqia | Al-Hayy | Al-Bashaer | Annual |
|--------------------|--------|--------|-----------|-------------|---------|------------|--------|
| Chloroform (CF) μg/l | Summer | 36.9   | 24.2      | 20          | 24.8    | 22.6       | 25.7   |
|                    | Spring | 21.4   | 18.2      | 15.7        | 19.3    | 16.1       | 18.14  |
|                    | Winter | 11.7   | 13.1      | 8.5         | 12.6    | 10.1       | 11.2   |
|                    | Autumn | 27.5   | 27.1      | 24.3        | 22.1    | 20.4       | 24.28  |
|                    | Summer | 43.2   | 44.9      | 47.2        | 47.6    | 42.9       | 45.16  |
| Bromodichloromethane (BDCM) μg/l | Spring | 21.8   | 24.8      | 18.3        | 21.1    | 25         | 22.2   |
|                    | Winter | 15.3   | 19.9      | 15          | 20.6    | 19         | 17.96  |
|                    | Autumn | 36.2   | 40.2      | 37.3        | 32.5    | 36.2       | 36.46  |
|                    | Summer | 33.3   | 33.8      | 38.3        | 35.4    | 28         | 33.76  |
| Dibromochloromethane (DBCM) μg/l | Spring | 17.1   | 21.4      | 14.6        | 15.9    | 18.8       | 17.56  |
|                    | Winter | 12.1   | 15.6      | 12.3        | 15.7    | 13.8       | 13.9   |
|                    | Autumn | 27.8   | 26.2      | 28          | 21.6    | 21.7       | 25.06  |
|                    | Summer | 8.2    | 10.8      | 10.2        | 7.7     | 8.6        | 9.1    |
| Bromoform (BF) μg/l | Spring | 3.7    | 4.3       | 7           | 4.6     | 4.5        | 4.82   |
|                    | Winter | 4.6    | 5.3       | 4.2         | 5       | 4.9        | 4.8    |
|                    | Autumn | 8.7    | 10        | 8.5         | 9       | 6.5        | 8.54   |
| Dichloro acetic acid | Summer | 13.9   | 14.1      | 13.1        | 17.4    | 13         | 14.3   |
Applying the approach of World Health Organization (WHO) index for additive to distribution system pipes \((23)\), THM levels in several Wassit regions have resulted in \(I_{\text{WHO}}\) values of more than 1 for all samples collected from the distribution systems of all water plants in summer. Computed \(I_{\text{WHO}}\) values of THM levels for distribution system was recorded for individual locations as presented in Table 3.

The obtained results have indicated that the highest \(I_{\text{WHO}}\) values of 1.37 and lowest \(I_{\text{WHO}}\) value of 0.458 was recorded in Al-Muwfaqia treatment plant during summer and winter, respectively. The increase of \(I_{\text{WHO}}\) value during summer season at Al-Muwfaqia water plant may be due to its location in the south of the city and high temperature in summer which may lead to polluting of the water by organic matter which reacted with more amounts of additive chlorine in the treatment plant during water disinfection and then formed high concentration of THMs. This method was conducted only on the THMs components. The additive toxicity of recorded THMs concentrations in the distribution systems of investigated sources was not coincident with that of WHO (2011) guideline value (less than 1). Thus, such concentrations have high adverse toxic health impacts.

### Table 3. Computed WHO additive toxicity values for distribution system THMs concentration

| Variables                  | Spring | Winter | Autumn |
|----------------------------|--------|--------|--------|
| (DCAA) \(\mu g/l\)       | 10.2   | 7.8    | 10.6   |
| (TCAA) \(\mu g/l\)       | 15.7   | 11.1   | 15.7   |
| (MCAA) \(\mu g/l\)       | 9.8    | 7.5    | 15.7   |
| (MBAA) \(\mu g/l\)       | 8.5    | 6.9    | 8.6    |
| (BCAA) \(\mu g/l\)       | 4.9    | 4.6    | 4.7    |
| (BDCAA) \(\mu g/l\)      | 5.3    | 3.9    | 4.7    |
| (CDBA) \(\mu g/l\)       | 4.9    | 5.5    | 4.7    |
| (TBAA) \(\mu g/l\)       | ND     | ND     | ND     |

ND=Not Detected

\(I_{\text{WHO}}\) values for distribution system THMs concentration...
2HAAs via one exposure route are given in Table 4 and presented in Fig. 1.

The obtained results of the average lifetime cancer risk posed by 4THMs via three exposure routes and

### Table 4. The average lifetime cancer risks posed of 4THMs and 2HAAs via different exposure routes.

| Variables | Season | Oral   | Spring | Summer | Autumn | Winter | Total |
|-----------|--------|--------|--------|--------|--------|--------|-------|
|           |        | 2.27   | 1.16   | 9.92   | 3.74   | 8.16   | CF    |
| Inhalation|        | 1.03   | 7.30   | 4.50   | 9.80   | 3.19   |       |
| Dermal    |        | 4.98   | 3.45   | 2.13   | 4.62   | 1.52   |       |
| Total     |        | 3.80   | 2.23   | 1.65   | 5.18   | 1.28   |       |
| Oral      |        | 7.99   | 3.93   | 3.18   | 6.46   | 2.16   |       |
| BDCM      |        |        |        |        |        |        |       |
| Inhalation|        |        |        |        |        |        |       |
| Dermal    |        | 1.01   | 4.98   | 4.03   | 8.18   | 2.74   |       |
| Total     |        | 9.5    | 4.43   | 3.58   | 7.28   | 2.43   |       |
| Oral      |        | 8.01   | 4.21   | 3.34   | 6.01   | 2.16   |       |
| DCAA      |        |        |        |        |        |        |       |
| Inhalation|        |        |        |        |        |        |       |
| Dermal    |        |        |        |        |        |        |       |
| Total     |        | 8.75   | 4.59   | 3.64   | 6.56   | 2.35   |       |
| Oral      |        | 2.10   | 1.90   | 1.90   | 1.22   | 7.12   |       |
| BF        |        | 1.52   | 8.05   | 8.02   | 1.43   | 4.56   |       |
| Total     |        | 2.25   | 1.98   | 1.98   | 1.36   | 7.57   |       |
| TTHMs     | Total  | 2.17   | 1.14   | 9.1     | 1.91   | 6.13   |       |
| DCAA      | Oral   | 2.04   | 1.58   | 1.11   | 1.44   | 6.13   |       |
| Inhalation|        |        |        |        |        |        |       |
| Dermal    |        |        |        |        |        |        |       |
| Total     |        | 2.04   | 1.58   | 1.11   | 1.44   | 6.13   |       |
| Oral      |        | 3.66   | 3.19   | 2.21   | 2.58   |        |       |
| TCAA      | Inhalation| ND  | ND  | ND  | ND  |        |       |
| Dermal    |        |        |        |        |        |        |       |
| Total     |        | 3.66   | 3.19   | 2.21   | 2.58   | 1.16   |       |
| THAAs     | Total  | 5.7    | 4.8    | 3.3    | 4.0    | 1.78   |       |
| Total DBPs|        | 2.74   | 1.62   | 1.24   | 2.31   | 7.9    |       |

ND: Not Detected

The lifetime cancer risks during oral ingestion of 4THMs and 2HAAs from tap water of all districts were higher than $1 \times 10^{-6}$ which is the negligible risk level defined by the USEPA. The highest lifetime cancer risk was in summer with $8.01 \times 10^{-5}$ for DBCM, while the lowest cancer risk was in winter with $1.22 \times 10^{-6}$ for BF. The average lifetime cancer risk for both THMs and HAAs were varied from high to low in the arrange of DBCM, BDCM, TCAA, CF, DCAA, and BF.

The lifetime cancer risks of four THMs compound through dermal contact with tap water in almost districts were higher than $10^{-6}$ which is the lower of the range of acceptable risk by the US.EPA...
and below this range for BF. The highest lifetime cancer risk recorded the value of 1.01×10^{-5} for BDCM during summer and the lowest cancer risk recorded the value of 8.02×10^{-8} for BF during winter. Among the four THMs, BDCM had the highest lifetime cancer risk, followed by DBCM, CF, and BF in order.

The computed cancer risk of THMs through inhalation (during bathing and showering) was only carried out for chloroform compound. The chloroform from tap water of the districts was above the 10^{-6} risk level and the highest lifetime cancer risk of CF recorded during summer was 1.03×10^{-5} and the lowest cancer risk of CF recorded was 4.50×10^{-6} during winter.

The average lifetime cancer risk posed by four THMs and two HAAs compounds in drinking water by three exposure routes of THMs and one exposure route of HAAs were 6.13×10^{-4} and 1.78×10^{-4}, respectively. The risk ratio of THAA to TTHMs was 3.44. The highest total cancer risk was in summer with 2.17×10^{-4} and 5.7×10^{-5}, while the lowest cancer risk was in winter with 9.1×10^{-5} and 3.3×10^{-5} for THMs and HAAs, respectively, Table 4.

Figures 2 and 3 show that the exposed population has a higher risk of cancer through oral ingestion followed by dermal absorption and inhalation and that BDCM appears to be the highest percentage contribution to the average lifetime cancer risk (30.7%) followed by DBCM, CF, TCAA, DCAA, and BF.

Also, the results have revealed that total cancer risk of HAAs and THMs during multiple pathways of exposure was 7.9×10^{-4} which was higher than the USEPA range of concern limit of 1×10^{-6} (14) by around 100 times. This indicates that about eight of every 10,000 individuals in Wassit Province could get cancer from the daily intake of drinking water in his lifespan. The Iraqi standard for TTHMs 150 mg/l must be reduced.

The results recorded in the present study have agreed with those of other studies (27, 28, 29, 30). These high values of cancer risk may cause many diseases among the exposed population (31).

The significance of the three exposure pathways was ranked differently in the studies and may be attributed to various concentrations and speciation of THMs and HAAs present in the water (15, 31, 22, 17, 32).

Conclusion:

The concentration levels of HAAs and THMs in drinking water samples from several Wassit districts water plants were generally within the allowable concentration recommended by the Iraqi standards, but many samples have exceeded the permissible limit of USEPA. The recorded values of the WHO index for additive toxicity (I_{WHO}) has indicated that THMs concentrations in drinking water of residential districts of Wassit Province pose adverse toxic health effects and non-carcinogenic risks in the Wassit population. The average lifetime cancer risk posed by two HAAs and four THMs during multiple pathways of exposure was higher than the USEPA range of concern limit by around 100 times. This indicates that about eight of every 10,000 individuals in Wassit could get cancer from the daily intake of water in his lifespan. Also, the contribution of risk was observed in the following order: oral ingestion (84%) > dermal absorption (10.8%) > inhalation (5.2%).

Conflicts of Interest: None.

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تقييم المخاطر الصحية البشرية المرتبطة بالتعرض للمنتجات العرضية الناتجة من تعقيم مياه الشرب في محافظة واسط جنوب شرق العراق

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الخلاصة:
قدرت المخاطر السمية ومخاطر السرطان خلال فترة الحياة المرتبطة بالتعرض لمركبات الهالوستك أسد والتراي هالوميثان الناتجة من عملية تعقيم مياه الشرب في عدة مناطق في محافظة واسط. أشارت نتائج التغيرات الفصلية لمركبات الهالوستك أسد والتراي هالوميثان في مياه الشرب إلى أن قيم معدلات الهالوستك أسد الكلية وقيم التراي هالوميثان الكلية تراوحت من 43.2 إلى 72.4 مايكروغرام/لتر ومن 40.0 إلى 115.5 مايكروغرام/لتر على التوالي. منهج دليل منظمة الصحة العالمية للمواد السامة حيث كانت قيمة النتائج في الصيف والخريف غيرمتوافقة مع الحدود المسموح بها، وهذا يعني أن تراكيز التراي هالوميثان لها تأثيرات سمية على الصحة. أما التقييم متعدد الطرق لخطورة السرطان على صحة الإنسان خلال العمر والتي تؤدي إلى تراكيز التراي هالوميثان في مياه الشرب، وبواسطة طريق تعرض واحد للهالوستك أسد وتلك تأثير الكرياتي هالوميثان كانت قيم التراي هوالوستك أسد والتي أهمية حماية البيئة الأمريكية بحوالي مائة مرة. وكانت نسبة خطر التراي هالوميثان أشد الكلي إلى التراي هالوميثان الكلي 3.44 وسجت على نسبة خطر السرطان للثاني كلووبروموثان وليبيا كل من الثلاثي البروم كلروfwورام والكلوروفورم والثلاطي كلوووسك أسد والثاني كلوووسك أسد والثاني كلوووسك أسد والثاني كلوووسك أسد والثاني كلوووسك أسد.

الكلمات المفتاحية: المنتجات العرضية للتعقيم، مياه شرب، هالوستك أسد، المخاطر الصحية، تراي هالوميثان.