ORIGINAL PAPER

Disinfection byproducts in swimming pool water in Sanandj, Iran

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ABSTRACT

The present study aimed to determine the concentrations of several disinfection byproducts (DBPs), including trihalomethanes (THMs), haloacetic acids (HAAs), and haloacetonitriles (HANs), in the public and private swimming pools in Sanandaj, Iran (n=16). Correlations between DBP levels with water quality parameters (free chlorine, pH, total organic carbon, temperature, number of swimmers, and gender of swimmers) and various DBP categories were investigated and compared. According to the results, concentrations of THMs in public and private pools were lower than the recommended limit in Iran (200 µg/L). In addition, HAAs had the highest concentrations, followed by THMs and HANs, respectively. Among the HAAs, THMs, and HANs, trichloroacetic acid was the most dominant species, followed by chloroform and dichloroacetonitrile, respectively. DBP levels were not affected by the gender of swimmers, number of swimmers, pH, temperature, and free chlorine. However, total organic carbon showed a fairly good correlation with TTHM, THAA, and DBP levels (r=0.45-0.78; $P<0.05$). Some correlations were also observed between various DBP categories.

Keywords: Disinfection Byproducts, Swimming Pool, Sanandaj, Iran

Introduction

Swimming in pools is a popular activity for millions of people around the world.[1] It is crucial to prevent the infectious diseases caused by microbial pathogens in water through the disinfection of swimming pools.[2] With its residual disinfection effects, chlorine is the most commonly used disinfectant to control the quality of pool water and prevent the spread of water-borne diseases in swimmers.[3]

Disinfection byproducts (DBPs) are formed during the chlorination of pool water as a result of the reactions between chlorine and organic compounds from the water source or swimmers (e.g., hair, sweat, lotion, mucus, sunscreen, cosmetics, urine, and skin particles).[3] So far, more than 600 DBPs have been identified,[1, 4] among which haloacetic acids (HAAs), trihalomethanes (THMs), and haloacetonitriles (HANs) are the most frequent chlorinated byproducts.[5] In addition, the most abundant THMs are bromodichloromethane (BDCM), chloroform, bromoform, and dibromochloromethane (DBCM).[6] According to the International Agency for Research on Cancer (IARC), chloroform and BDCM are possible carcinogens for humans (group B2).[7] Several epidemiological studies have reported an association between THM exposure and adverse health consequences, such as asthma, irritation of the eyes, skin, and respiratory tract, and adverse reproductive effects.[8, 9]

Dichloroacetic acids (DCAA) and trichloroacetic acids (TCAA) are the most abundant HAAs.[10] According to several toxicological studies, HAAs exhibit cytotoxic, mutagenic, genotoxic, and teratogenic effects on body cells.[11] These compounds are classified as group C and group B2 carcinogens.[12] HANs encompass trichlroacetonitrile (TCAN), bromochloroacetonitrile (BCAN), dichloroacetonitrile (DCAN), and dibromoacetinitrile (DBAN). According to the literature, DCAN exhibits mutagenicity on assays, while carcinogenic or mutagenic effects have been attributed to DBAN and BCAN in mice.[13]

Exposure to DBPs in swimming pools may occur through the inhalation of volatile DBPs,

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skin absorption, and ingestion of pool water, especially in children.[14] Some studies have reported a high risk of DBP exposure through inhalation and dermal pathways while swimming.[15] Moreover, the presence of DBPs in pool water has been linked to the increased risk of colon, bladder, and rectal cancers.[10] In Spain, Villanueva *et al.* observed a correlation between bladder cancer and THM exposure via water ingestion, inhalation, and thermal absorption while showering or bathing.[15] Some studies have also demonstrated that HAAs are more carcinogenic compared to THMs, and DCAA and TCAA could cause liver tumors in rodents.[16] Additionally, some animal studies have denoted that THM exposure is linked to tumor formation in the kidneys and liver.[17]

In 1998, the United States Environmental Protection Agency (USEPA) announced the maximum allowed content of these compounds to be 80 µg/L in drinking water.[18] The Institute of Standard and Industrial Research in Iran has determined the maximum acceptable content of these compounds to be 200 µg/L based on the standard No. 11203.[19] Since swimming pools use a water recirculation system for relatively long intervals, DBP concentrations in swimming pools may be higher than drinking water.[20] The results obtained by Weisel *et al.* confirmed that the chloroform concentration in pool water is 2-20 times higher than tap water.[21]

The amount of formed DBPs in pool water is influenced by several factors, including free residual chlorine, number of swimmers, gender of swimmers, water temperature, total organic carbon (TOC), water resource, and pH.[13] To the best of our knowledge, the present study is the first report in Sanandaj, Iran regarding the concentrations of DBPs (THMs, HAAs, and HANs) in the swimming pools treated with chlorine. Correlations between DBP concentrations with water quality parameters and various DBP categories were also investigated.

Materials and Methods

Materials

Standard solutions of four THMs (BDCM, chloroform, DBCM, and bromoform), two HAAs (TCAA and DCAA), and four HANs (DCAN, TCAN, DBAN, and BCAN) were supplied by Supelco Company (USA). In addition, sodium thiosulfate was purchased from PanReac Company. Methyl tert-butyl ether (MtBE) and HPLC-grade methanol were supplied by Tedia Company. The other chemicals were provided from Merck Company.

Study Design

Swimming pool water samples were collected from 16 private and public swimming pools in Sanandaj, Iran. The water in all the swimming pools was disinfected with chlorination. Sample collection was performed during June-November 2016. To assess the variations in DBP concentrations, four water samples were obtained monthly from each swimming pool in the morning or afternoon for two months in the presence of male and female swimmers. In total, 128 samples were collected at the depth of 20-30 centimeters from the water surface and a 40-centimeter distance from the edge of the pool. In order to investigate the effective parameters in water contamination, several variables were measured, including water temperature, pH, free chlorine, and TOC. During sampling, the number of swimmers was recorded as well.

Sampling and Analysis

To measure the THM levels, swimming pool samples were collected in 50-millilitre bottles. Before sampling, approximately two milligrams of sodium thiosulfate was added to the bottles in order to neutralize residual chlorine and hinder the formation of the THMs. To determine HAA and HAN compounds, the swimming pool samples were transferred to 50-millilitre glass bottles. Before sampling, about six milligrams of ammonium chloride was added to the bottles as the dechlorination agent for HAA measurements. In addition, in HAN measurements, a mixture (0.8 g) containing 99% potassium phosphate monobasic (KH_2PO_4) and 1% sodium phosphate monobasic (Na_2HPO_4), was used to reduce the pH to 4-5 and inhibit HAN degradation.

For TOC analysis, the water samples were collected in one-liter glass bottles. In each sampling, one control blank sample was analyzed as well. All the water samples were stored at the temperature of 4 ºC until further assessment. The EPA-500 method (US EPA methods 524.2, 552.2, 551.1; 1995) was used to evaluate the concentrations of HAA, THM, and HAN, respectively.[22-24]

THMs and HANs were extracted using pentane and MtBE, respectively and analyzed using a gas chromatograph (GC) (VARIAN CP-3800) equipped with an electron capture detector (ECD). For peak separation, a capillary column (30 m×0.25 mm×3.0 µm) was applied. The oven temperature program included 35 ºC for five minutes, the first ramp of 10 ºC/min to 80 ºC for one minute, and the second ramp of 20 ºC/min to 200 ºC for one minute. The temperature was 280 ºC and 300 ºC in the injector and detector, respectively. The detection limit for each THM was 0.1 µg/L, while the detection limits for DCAN, TCAN, BCAN, and DBAN were 0.1, 0.2, 0.2, and 0.1 µg/L, respectively.

HAA samples were adjusted at the pH of ≤0.5 through the addition of sulfuric acid (98%). Afterwards, extraction was performed with MtBE, and the organic phase containing the HAAs was treated with methanol. HAAs were measured using a GC equipped with an ECD, and a capillary column was employed for peak separation. As for the oven temperature for GC, primary temperature was 40 ºC (0.5 minute), and the first ramp was 10 ºC/min to 230 ºC (one minute). The temperature was set at 300 ºC and 270 ºC in the injector and detector, respectively. The detection limit for each HAA was one µg/L.

A TOC analyzer (TOC 5000A, Shimadzu, Japan) was applied to examine the TOC concentration in the filtered samples, and the detection limit for TOC was 0.1 mg/L. Free residual chloride was determined on the site using the colorimetric method by adding DPD reagent. Following that, water temperature and pH were analyzed on the site using a thermometer (PDQ400, USA) and portable pH meter (3510, Jenway), respectively.

Statistical Analysis

Data analysis was performed using Spearman's correlation-coefficient to evaluate the correlations between various DBP categories (TTHMs, THAAs, and THANs), as well as the associations of DBPs and water quality parameters.

Results and Discussion

Quality Parameters

Quality parameters of the studied swimming pools (n=16) are presented in Table 1. In total, four swimming pools (25%) were private, and 12 pools (75%) were public. Mean concentration of free residual chlorine in the public and private pools was 1.01 and 2.03 mg/L, respectively, which was within the standard range (1-3 mg/L).[19] However, mean free chlorine concentration was observed to be higher than the recommended level (0.2 mg/L) for microbial inactivation.[13] Regarding the temporal distribution of chlorine, the effect of sampling time on the concentration of free chlorine in the public and private pools is depicted in Fig. 1.

In the public and private pools, chlorine concentration was relatively high in the morning (8:30-9:30 AM) and low/undetectable at noon (around 12 PM), while it slightly increased in the evening (17:30-19:30 PM). According to the findings, the number of swimmers may influence the chlorine concentration in pools as the number of swimmers was low in the morning and early afternoon, while it significantly increased in the evening. In the case of extremely low or undetectable free chlorine concentration, the risk of microbial growth and outbreaks of infections could increase for swimmers,[25] while the formation of DBPs would decrease.

Mean TOC concentration in the public and private pools was 4.12 and 9.42 mg/L, respectively, which was lower than the levels reported by Manasfi *et al.* (11.52 mg/L).[8] A significant amount of organic constituents (e.g., hair, sun lotion, and urine) may enter the pool water from swimmers, especially in summer, thereby increasing the TOC concentration in pool water.[26] It is notable that the previous studies in this regard have reported higher levels of TOC in swimming pools.[8, 9]

In the current research, pH ranges were 7.2-7.7 and 6.7-7.6 in the public and private pools, respectively. The pH of the pool water samples was consistent with the recommended level in Iran (pH=7.2-7.8). Previous studies have denoted that hypochlorite acid is predominant at the pH level of <7.5, whereas hypochlorite ions are frequent at the pH of >7.5. Therefore, addition of poor acids to pool water is essential to for effective disinfection.[13] In the present study, temperature was 26.3 ºC in the water of public swimming pools and 27.2 ºC in the water of private swimming pools, which was in line with the standard limit in Iran (26-28 ºC).

Table 1. Concentrations of physicochemical parameters in water samples from public and private swimming pools

	Public pools	Private pools
Free chlorine (mg/L)	1.01±0.90 (0.4-2.3)	2.03±0.60 (0.8-4.02)
pH	7.43±0.14 (7.2-7.7)	7.1±0.80 (6.7-7.6)
Temperature (ºC)	26.3±0.5 (26.2-28.8)	27.2±0.3 (26.1-30.4)
TOC (mg/L)	4.12±1.32 (2.1-6.3)	9.42±3.71 (7.3-11.8)

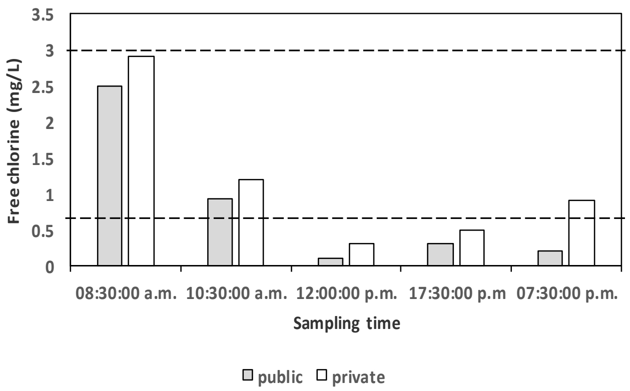

Fig. 1. Free chlorine level at different intervals in water samples from public and private swimming pools (horizontal dashes indicate the free chlorine range based on Iranian standards)

DBPs in Swimming Pool Water

Table 2 shows the concentrations of various THMs (chloroform, DBCM, BDCM, and bromoform), HANs (TCAN, DCAN, BCAN, and DBAN), and HAAs (DCAA and TCAA) in the water samples obtained from the public and private pools treated with chlorine (n=16). Accordingly, DBP concentrations in the pool water depended on the qualities and impurities in the water used to fill the pools.

Total THM concentrations in the public and private pools were 144 and 173.3 µg/L, respectively, which were significantly higher compared to some of the previous studies in this regard,[2, 6, 7, 27] while lower than the reported levels in several other studies.[4, 9] Several factors could influence THM formation in pool water, including the gender of swimmers, number of swimmers, temperature, pH, residual chlorine concentration, and TOC.[2] According to the information in Table 2, chloroform had the highest concentration compared to other THMs (80-90%) in the public and private pools, followed by DBCM and BDCM (18.4 and 2.2 µg/l in public pools and 22.1 and 3.5 µg/l in private pools, respectively). On the other hand, concentration of bromoform was lower than the detection limit (0.2 µg/L).

In the present study, the most dominant HAA species was TCAA (74% of total HAAs) in the public and private pools, followed by DCAA (26% of total HAAs). This finding might be attributed to the low concentration of bromide in the pool water.[27] In general, mean concentration of total HAAs was 266.3 µg/L in the public pools and 279.8 µg/L in the private pools; in this regard, Yang *et al*. reported a higher HAA concentration (798 µg/L) compared to our findings. However, significantly lower concentrations of HAAs (120.9 µg/L) were reported in 86 swimming pools in Korea.[13] The similarity or discrepancy of the results obtained in the mentioned studies could be due to the variable disinfection conditions of swimming pools.[27]

Formation of HANs in pool water depends on the presence of nitrogen and organic compounds. Reaction of nitrogen compounds, such as sweat and urine with chlorine, seems to contribute to the generation of HAN compounds in swimming pools.[28] According to the current research, mean concentration of total HANs in the public and private pools was 5 and 8.4 μg/L, respectively. Consistent with the previous studies in this regard,[4, 8, 13] DCAN had the highest concentration in the selected pools in the present study compared to other HANs (maximum: 76% and 62% of total HANs in the public and private pools, respectively).

As can be seen in Fig. 2, THAAs were the most frequent DBPs with the highest concentrations (64.3% and 61.07% of total DBPs in the public and private pools, respectively), followed by TTHMs (34.8% and 37.8% in the public and private pools, respectively) and THANs (0.9% and 1.13% in the public and private pools, respectively).

In order to prevent the outbreaks of water-borne diseases, swimming pool water should be frequently disinfected.[13] Consequently, chlorine concentration in swimming pools often exceeds that of drinking water. However, previous studies have indicated that high chlorine concentrations are directly associated with the increased formation of HAAs rather than THMs.[4] In addition, THMs could be easily volatilized from water into air, whereas HAAs are comparatively nonvolatile.[4,13] Therefore, THAA concentrations in pool water are higher than TTHM concentrations. On the other hand, recent studies have reported THAAs to have the highest concentrations, followed by TTHMs and THANs.[5]

In the present study, concentrations of HAAs and THMs in the pools exceeded the EPA regulations for drinking water (1998) (THM≤80, HAAs≤60 μg/L).[18] However, the concentration of THMs in the public and private pools was lower than the standard limit in Iran (200 μg/L). In a study by Heydari *et al.* investigating the water in the swimming pools in Fars province (Iran), THM concentration was observed to be higher than the standard level in Iran.[3] In general, our findings regarding the distribution of DBPs are consistent with the previous studies in this regard, which denoted chloroform, TCAA, and DCAN to be the prominent species among THMs, HAAs, and HANs, respectively.[8, 13]

Table 2. DBP concentrations (μg/L) in samples from public and private swimming pools

	Public pools	Private pools
Chloroform	123.4±83.4 (82-198.3)	147.7±76.9 (96.3-214.8)
BDCM	18.4±32.1 (1.2-24.1)	22.1±41.4 (7.3-46.2)
DBCM	2.2±4.0 (1.3-4.3)	3.5±6.1 (1.7-6.4)
Bromoform	N.D.	N.D.
TCAA	197.4±102.6 (56.2-421.2)	206.5±113.1 (48.2-543.1)
DCAA	68.9±27.8 (15.1-274)	73.3±97.5 (22.1-298.4)
DCAN	3.8±4.0 (0.4-11.2)	5.2±6.1 (4.1-8.5)
TCAN	1.2±1.3 (0.3-6.3)	3.2±4.1 (1.1-7.3)
BCAN	N.D.	N.D.
DBAN	N.D.	N.D.

N.D. not detected

Fig. 2. Distribution of DBPs in samples from public and private swimming pools

Correlations between Various DBPs

Correlations between various DBP categories are shown in Table 3. In the current research, positive correlations were observed between TTHMs and THAAs (r=0.32; P<0.05). Furthermore, a significant correlation was denoted between THAAs and DBPs, which could be attributed to the domination of HAAs among DBP categories (r=0.82; P<0.05). These findings are in congruence with the results obtained by Lee *et al.*, which indicated a significant correlation between DBP and THAA concentrations in pool water (r=0.95; P<0.0001).[13] On the other hand, DBPs showed a relatively poor correlation with THMs (P<0.05), which might be associated with THM volatilization from water into air. Consequently, THM concentration may not be an efficient indicator of DBP levels in swimming pool water.[8, 13]

Table 3. The correlation coefficients for TTHMs, THAAs, THANs, and DBPs in the water samples from swimming pools

	TTHMs	THAAs	THANs	DBPs
TTHMs	1	0.32*	0.04	0.061*
THAAs		1	0.03	0.82*
THANs			1	0.07
DBPs				1

*P<0.05

Table 4. The correlation coefficients between classes of DBPs and physicochemical parameters in the swimming pool water samples

	TTHMs	THAAs	THANs	DBPs
TOC	0.45*	0.63*	0.21	0.78*
Free residual chlorine	0.11	0.32	0.30	0.20
pH	0.34	0.05	0.34	0.43
Temperature	0.03	0.07	0.46	0.03
Number of swimmers	0.04	0.41	0.52	0.52
Sex of swimmers	0.032	0.38	0.63	0.50

*P<0.05

Correlations between DBPs and Quality Parameters

Table 4 demonstrates the correlations between DBPs and quality parameters of pool water, including TOC, free chlorine, pH, temperature, number of swimmers, and gender of swimmers. Accordingly, no correlations were observed between DBPs and water temperature, pH, and free residual chlorine. Moreover, the results indicated no correlations between DBP levels and the number and gender of swimmers. On the other hand, TOC had fairly significant correlations with TTHM, THAA, and DBP concentrations in pool water (r=0.45-0.78). Therefore, it could be concluded that TOC is the most significant determinant of DBP formation in swimming pools, and TOC level should be lowered to inhibit the formation of DBPs in swimming pool water.[13]

Conclusion

In the present study, DBP concentrations were surveyed in the public and private swimming pools treated with chlorine (n=16). The correlations between quality parameters and DBPs, as well as the associations of various DBP categories, were also examined. According to the results, THM and HAA levels in the swimming pools exceeded the values recommended by intentional organizations for drinking water, while THM levels were lower than the standard levels in Iran. In addition, HAAs had the highest concentrations, followed by THMs and HANs, respectively. These findings could be attributed to the use of high disinfectant doses for pool water treatment to prevent infection outbreaks. In the current research, TCAA, chloroform, and DCAN were identified as the dominant species among HAAs, THMs, and HANs, respectively. Additionally, TOC and HAA exhibited a positive association with total DBPs. However, free residual chlorine, pH, temperature, number of swimmers, and gender of swimmers had no effects on DBP concentrations. Some correlations were also observed between various DBP categories. In conclusion, it is suggested that the formation of DBPs be controlled through applying new treatment technologies, such as filtration methods and new oxidation techniques, and promoting hygiene among swimmers.

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