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Research Article

Occurrence of bromide and bromate in chlorinated indoor swimming pools, and associated health risks

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ABSTRACT

Swimming is a physical activity that is accessible to people of all ages in all seasons. However, continuous organic and inorganic precursor load and disinfectant dosing make pool water chemistry much more complex than other disinfected waters. Carcinogenic bromate compound is one of the hundreds of disinfection by-products in pool water. The occurrence of bromate in pool waters depends on the precursor content of filling water, the disinfection process, operating parameters, and the purity of disinfectants. While the average filling water bromide concentrations of University Campus indoor swimming pool in Gülbahçe –Urla (SP1) and Buca public indoor swimming pool (SP2) were determined to be 182 µg/L and 11.0 µg/L, respectively, the average bromate concentrations of SP1 and SP2 were 59.4 µg/L and 68.3 µg/L. Estimated chronic-toxic health risks of accidental ingestion of pool water during swimming (between 10^{-3} and 10^{-1}) were lower than the threshold level ('1'). Although the carcinogenic risks in central tendency scenario ($<10^{-6}$) indicate negligible risks for swimmers, worst case scenario indicates carcinogenic risks (medians were ranged from 1.61×10^{-6} to 9.42×10^{-6}) for highly exposed specific swimmer groups. Bromate accumulation in swimming pools needs attention for mitigating the health risks for swimmers.

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INTRODUCTION

Swimming is one of the most common sports activities that people of all age groups can do. With the '*Yüzme Bilmeyen Kalmasın*' project initiated by the Ministry of Youth and Sports in Türkiye; people are encouraged to increase their interest in swimming [1]. Within the scope of this project, the total number of pools (Olympic, semi-Olympic and non-Olympic) in Türkiye which was 46 in 2002 increased to 610 in 2022. Although the number of indoor and outdoor swimming pools in Türkiye is not known, the increase in the number of swimming pools opened in recent years can be considered as an indicator of people's interest in swimming. Swimming pool disinfection is vital to maintaining a safe and healthy swimming environment. Swimming pools must be disinfected continuously to prevent microbial activities that cause waterborne outbreaks and infections caused by swimmer-borne pathogens (viruses, bacteria and fungi) [2, 3]. Chlorine is the

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most widely used disinfectant for swimming pools, as it is effective at killing a wide range of microorganisms and due to its low cost [4, 5]. Besides chlorine; sodium hypochlorite, chloramine, ozone, chlorine dioxide, ultraviolet radiation (UV) etc. are also used effectively [6, 7].

Disinfection of swimming pools may generate potentially hazardous byproducts known as disinfection byproducts (DBPs). Organic or inorganic disinfection by-products are formed due to the reaction in the pool water during disinfection. Organic DBPs occur when disinfectant reacts with organic matter in the pool water, such as sweat and urine [8, 9]. These byproducts can cause irritation of the eyes and lungs, as well as other health problems [10, 11].

Pool operators should carefully adjust the amount of chlorine required for disinfection to reduce the risk of DBPs, monitor the amount of residual chlorine in the pool to ensure continuity of disinfection, and regularly test pool water for DBPs. To ensure that sufficient disinfection efficiency is maintained in the pool water, residual chlorine concentration in swimming pool is regulated. In Türkiye, the concentration of free chlorine in the chlorinated pool water is regulated to be between 1-1.5 mg/L in indoor swimming pools, and 1-3 mg/L in outdoor swimming pools, while free chlorine concentrations in pools where other disinfection methods (UV, ozone, chlorine dioxide, and others) are used are limited to 0.3-0.6 mg/L [12].

Inorganic disinfection byproducts (bromate, chlorite and chlorate) are also detected in the water [13]. Bromate is classified as a possibly 2B carcinogen (possibly carcinogenic) to human [14]. There are several reasons for the formation of bromate detected in disinfected water. Bromate is formed when ozone is udes as the disinfectant in bromide-containing waters [15, 16]. It also forms with the use of disinfectants containing chlorine. Chlorine is an oxidizing substance. When the water is disinfected with chemicals containing chlorine, if the water containing residual chlorine contains a certain amount of bromide ion, this ion is converted to bromate by chlorine oxidation. In Türkiye, the most commonly used chlorination methods in swimming pools are sodium hypochlorite dosing and hydrolysis to separate free chlorine molecules from the saline solution. When disinfected with chemicals containing chlorine, the chlorine mixes with the water and forms hypochlorous acid, which dissociates into hydrogen atoms and hypochlorite ions. It is the sum of the chlorine residue (free chlorine), hypochlorous acid, hypochlorite ion and aqueous chlorine concentrations in the pool as a result of disinfection [17]. Bromate formation on chlorination of bromide ingredient waters is very slow process due to the low disproportionation rate constant of hypobromous acid (HOBr). Bromate formation in chlorinated water starts with the formation of HOBr with reaction between hypochlorous acid (HOCl) and bromide ion results bromide oxidation by HOCl to form HOBr and Chloride (Cl^-). HOBr is in equilibrium with hypobromite ion (OBr^-) (acid dissociation constant, pKa=8.8). Further, disproportionation of HOBr forms the reduced species (-1 and +1 valance) of Br and oxidized species (Bromate, BrO_2^-, Br^{+3} and Br^{+5}) [18]. Bromide ion to bromate conversion rate was less than 1.3% at the overdosing chlorine condition (5 mg/L) [19].

Electrolysis could be used for production of sodium hypochloride (NaOCl) from saline solution. During the electrolysis process, bromate could be formed as an unintentionally produced by-product due to the chemical conversion of bromide to bromate [20-23]. Bromate formation rate in electrolysis process depends to impurities in chlorine precursor salt [24, 13].

Bromate formation is not a primary concern in water chlorination as the formation process is slow in disinfection. However, it might be a significant concern in chlorinated waters due to the continuous disinfectant dosing and the non-volatile structure of bromate [25] that may result in bromate accumulation in swimming pools. Considering that swimming pools are closed systems, bromate levels tend to increase continuously since bromate is not volatile and water-soluble compound, and bromate entry into the pool due to continuous disinfectant dosing. For those reasons, neglected bromate levels in chlorinated water could be a significant risk factor for swimmers. Swimmers may be exposed by aerosol inhalation, dermal absorption (depending on skin permeability) or accidental ingestion pathways [26, 27]. In the article presented at the EUROTOX 2021 symposium, Röhl reported that 73-98% of the bromate exposure of swimmers occurred through the oral pathway [28].

After the NaOCl solution is produced, it is stored for sale or application after sale. During storage, the chlorine concentration in the disinfectant decreases under high temperature conditions. No significant change is observed in cold seasons. Bromate in solution exhibits a stable state unaffected by temperature [21]. The decrease in the chlorine concentration in the disinfectant during storage causes more disinfectant to be fed into the water to arrange regulatory levels and results co-dosing of bromate to the pool waters.

Reports of potential hazards and environmental levels of bromate in pool water are scarce in the literature. The aim of this study is to investigate the presence of bromate in two different indoor swimming pools and their filling waters, accidental oral exposure, and associated health risks. To the best of our knowledge this is the first study that investigate the occurrence of bromate in Turkish public pools and estimates associated health risks.

MATERIALS AND METHODS

Sampling and Analyses

Sampling campaign was conducted simultaneously in two Semi-Olympic indoor swimming pools that those were in Gülbahçe-Urla (University Campus-SP1) and Buca (Public Pool-SP2) from April-2019 to May-2019 (Figure 1).

While the pool water volume of SP1 was 900 m^3, pool water volume of SP2 was 579 m^3. They were filled with municipal water, and continuously circulated. Coarse particles, human body residues (hair, skin, etc.), and textile fibers in pool water were filtered in sequential coarse and fine sand filters and returned to pool from bottom nozzles after disinfectant (sodium hypochlorite) dosing. Temperature, pH, and free chlorine levels were continuously controlled to ensure regulatory limits.

Both pool water and filling water were sampled. While the physico-chemical parameters (water temperature, pH, bromide and bromate) were analyzed in pool waters, only bromide and bromate were the targeted parameters in filling water. Pool water samples were taken at 3 different points at a depth of 20 cm and 40 cm from the edge of the pool and combined. Water samples for bromate analysis were collected in Teflon-faced amber bottles (40 mL). Sodium sulfite was spiked in all samples for quenching the chlorine. Samples were kept at +4°C and analyzed within 48 hours.

pH, electrical conductivity (EC), and temperature parameters were measured with the Hach Lange HQ40D device. Bromide (Br^-) and bromate (BrO_3^-) ions were analyzed based on SM 4110 [29] using Ion chromatography coupled with CD20 conductivity detector (Dionex ICS-5000) in Environmental Research Center of Izmir Institute of Technology.

Exposure and Risk Assessment

Chronic-toxic health risks and carcinogenic risks associated with accidental ingestion of bromate pool water were estimated deterministically using Equations (1), (2) and (3). Exposure scenarios were constructed for four age groups, 6 to <11 years, 11 to <16 years, 16 to <21 years, and adults, in central tendency and worst cases (Table 1). In central tendency scenarios, median levels of pool water ingestion rate, body weight, and swimming duration were used for estimation of health risks, while those were upper-bound values of ingestion rate and swimming time and lower-bound for

Figure 1. Sampling points.

Table 1. Variables of exposure and risk assessment

Parameter	Central Tendency	Worst Case	Reference
C	**Individual**	**Individual**	**This study**
IR	6 to < 11 year: 0.025 L/h	6 to < 11 year: 0.096 L/h	[30]
	11 to < 16 year: 0.029 L/h	11 to < 16 year: 0.152 L/h	
	16 to < 21 year: 0.019 L/h	16 to < 21 year: 0.105 L/h	
	Adults: 0.013 L/h	Adults: 0.092 L/h	
A x EF	6 to < 11 year: 30.2 h/year	6 to < 11 year: 96 h/year*	[30, *34]
	11 to < 16 year: 27.8 h/year	11 to < 16 year: 96 h/year*	
	16 to < 21 year: 29 h/year	16 to < 21 year: 96 h/year*	
	Adults: 9 h/year	Adults: 96 h/year*	
ED	30 years for adults and 5 for other age groups		Assumed
CF	0.001		This study
BW	6 to < 11 year: 26.5 kg	6 to < 11 year: 21.3 kg	[31, 32]
	11 to < 16 year: 49.2 kg	11 to < 16 year: 39.4 kg	
	16 to < 21 year: 58.3 kg	16 to < 21 year: 53.53 kg	
	Adults: 65 kg	Adults: 48 kg	
AT	27375 days		[35]
SF	7×10^{-1} per mg/kg-day		[33]
RfD	4×10^{-3} mg/kg-day		[33]

body weight in worst case scenario. While the pool water ingestion rates and activity times of swimmers were taken from Exposure Factors Handbook [30], body weights of combined female and male Turkish Citizens [31, 32] were used for assessment of bromate exposure. Product of exposure duration, and exposure frequency was assumed to be equal to averaging time in CTR estimation. Reference dose (RfD) and slope factor (SF) of bromate were taken from the Integrated Risk Information System [33].

$$CDI = \frac{C \times IR \times A \times EF \times ED \times CF}{BW \times AT} \quad (1)$$

$$CR = CDI \times SF \quad (2)$$

$$CTR = \frac{CDI}{RfD} \quad (3)$$

where, CDI: chronic daily intake (mg/kg-day), C: bromate concentration in pool water (µg/L), IR: accidental ingestion rate of pool water (L/hr), A: activity time (hr/day), EF: exposure frequency (day/year), ED: exposure duration, CF: conversion factor (µg/L to mg/L), BW: body weight (kg), AT: average time (day), CR: carcinogenic risk (unitless), SF: slope factor (per mg/kg-day), CTR: chronic toxic risk (unitless), RfD: reference dose (mg/kg-day).

RESULTS

Physico-chemical parameters of filling water and pool water

Both of the studied swimming pools were filled with municipal water. Pool water temperature and pH was in agreement with the Türkiye guidelines setting physical parameters for swimming pools [12] (Table 2). The average bromide ion concentrations of filling water of SP1 and SP2 were determined to be 182 and 11.0 µg/L, respectively, with ranges of 162-203 µg/L and 8.58-12.9 µg/L. Filling water bromide concentrations of SP1 were determined to be around two order of magnitude higher than SP2. Relatively higher bromide levels of filling water of SP1, located in coastal zone of Karaburun Penninsula, might be due to the effects of seawater intrusion to the groundwater and/or difference of geogenic factors [36]. Coastal freshwater bromide ion could reach as high as 4 000 µg/L [37]. Bromide ions of swimming pool samples could not be analyzed due to overlapping of the peaks of inorganic chlorinated compounds and bromide.

The average bromate concentrations of SP1 and SP2 were determined to be 59.4 and 68.3 µg/L, respectively. Bromate concentrations in swimming pools were determined to be higher than the regulatory levels for Türkiye

Table 2. Physico-chemical characteristics of filling waters and pool waters

Parameters	Sample	Average	Minimum	Maximum
pH	FW1	6.98	6.95	7.01
	SP1	7.46	7.02	7.76
	FW2	*N.D.	*N.D.	*N.D.
	SP2	7.45	7.40	7.50
Bromide (µg/L)	FW1	182	162	203
	SP1	*N.D.	*N.D.	*N.D.
	FW2	11.0	8.58	12.9
	SP2	*N.D.	*N.D.	*N.D.
Bromate (µg/L)	FW1	*N.D.	*N.D.	*N.D.
	SP1	59.4	53.8	62.2
	FW2	2.13	0.55	2.96
	SP2	68.3	59.4	62.2

*N.D.: not determined

or European drinking water (10 µg/L) [38, 39]. Although there was a two order of magnitude difference of bromide concentrations between both sample pools, no significant difference was observed between the two pools, the difference in the bromate concentrations was not significant. While the bromate was not detected in the filling water in Italy, pool water concentrations were determined to be in the range of 10-48 µg/L [40]. Righi reported that the bromate could be formed in chlorinated bromide ingredient waters [40]. However, our findings support the bromate occurrence in chlorinated pools might be significantly affected by the co-dosing of pre-occurrence bromate with sodium hypochlorite for arranging the regulatory free chlorine levels in pool water. Also, bromate was detected in commercial sodium hypochlorite solutions that were used in disinfection of potable waters and pool waters [41]. Due to the continuous dosing of sodium hypochlorite disinfectant to ensure the regulatory disinfectant levels in pool water, co-dosing of bromate within sodium hypochlorite solution might be the most important bromate accumulation factor in chlorinated swimming pools. Non-volatile structure and chemical resistance to degradation results accumulation of bromate in swimming pools. Accidental ingestion of bromate-contaminated pool water during swimming activity, especially high levels for low age swimmers, might be pose a significant health risk for swimmers. Carcinogenic risk associated with bromate exposure of swimmers should be considered on public health mitigation efforts.

Bromate Health Risks in Swimming Pools

CTR levels of bromate exposure through accidental ingestion of pool water are shown in Figure 2. Exposure scenarios were constructed as described in the section on Material and Methods. In the central tendency scenario, CTR levels of bromate exposure were estimated to be lower than 3.00×10^{-2}, even in the maximum level with the medians of 1.60×10^{-2}, 9.84×10^{-3} , 5.46×10^{-3}, and 3.31×10^{-3} for the age groups of 6 to <11, 11 to <16, 16 to < 21, and adults, respectively. Those median CTR levels of bromate exposure in the worst-case scenario were 6.31×10^{-2}, 6.43×10^{-2}, 3.72×10^{-2}, and 3.20×10^{-2}, respectively. The maximum CTR levels in the worst-case scenario were estimated to be ranged from 5.61×10^{-2} to 1.13×10^{-1} for the considered age groups. While the maximum CTR levels were estimated to be lower than the threshold level of '1', synergistic effects of multi-compound exposure might be occurring with significant CTR for especially younger swimmers.

Central tendency scenario CR levels of bromate exposure due to the swimming activity are shown in Figure 3.

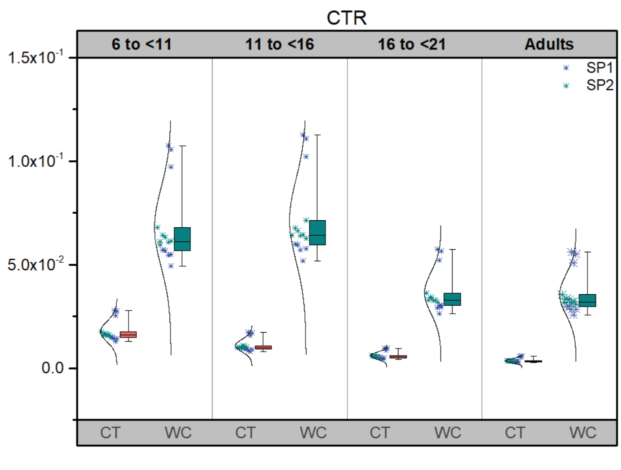

Figure 2. Bromate-associated CTR levels of accidental ingestion of pool water (CT: central tendency scenario, WC: worst case scenario).

CR levels are divided into four categories [32, 42]: safe zone was CR≤10^{-6}, acceptable risk zone was 10^{-6}<CR<10^{-5}, low priority risk zone was 10^{-5}<CR<10^{-4}, and high priority risk zone is CR≥10^{-4}. The CR of the bromate in the central tendency exposure scenario ranged in the safe zone for all age groups with the median levels of 2.46×10^{-7} for 6 to <11 age group, 1.40×10^{-7} for 11 to <16 age group, 8.09×10^{-8} for 16 to <21 age group, and 9.15×10^{-9} for adults (Figure 4). CR levels of bromate ingestion during swimming in the worst-case scenario were estimated to be higher than the safe zone level. The median CR levels of 6 to <11, 11 to <16, 16 to <21, and adult groups in the worst-case scenario were estimated to be 3.01×10^{-6}, 3.16×10^{-6}, 1.61×10^{-6}, and 9.42×10^{-6}, respectively. While the maximum CR was estimated for adults with the level of 1.65×10^{-5}, the lowest level was estimated for the 16 to <21 age group. 16 to <21 age group had a lower CR than adults, which might be due to the shorter exposure duration despite swallowing more pool water than adults. While the CR levels of < 21 age swimmers were estimated to be in the acceptable risk zone, upper quartile CR levels of adults were in the low priority zone. CR and CTR levels in SP2 tended to be close to the median levels due to the concentration variation being lower than the SP1. Disinfectant dosing system stability, treatment system efficiency, and partial refilling strategies might affect pool water bromate levels, which directly affects the health risk levels. While the bromate formation potential of chlorinated bromide content waters is near the negligible levels, pool waters (SP1) affected by seawater intrusion needs to be studied in detail on toxic bromate and brominated organic by-product formation due to the high bromide ions in the filling water. Variation of bromate concentrations in SP1 might be the result of the seawater intrusion of source water. Detailed long-term studies are needed to cover the gap in bromate levels in chlorinated swimming pools.

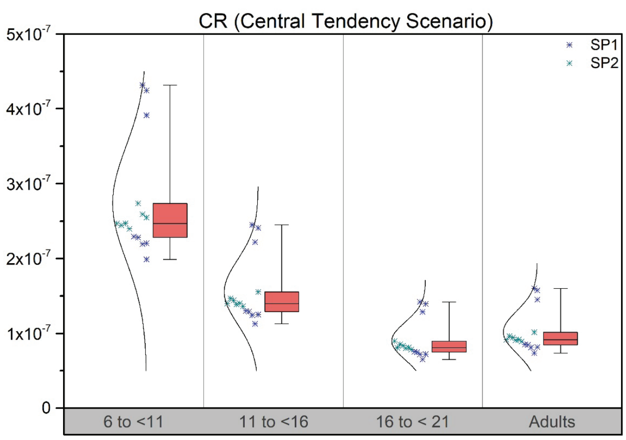

Figure 3. Bromate-associated CR levels of accidental ingestion of pool water (Central Tendency Scenario).

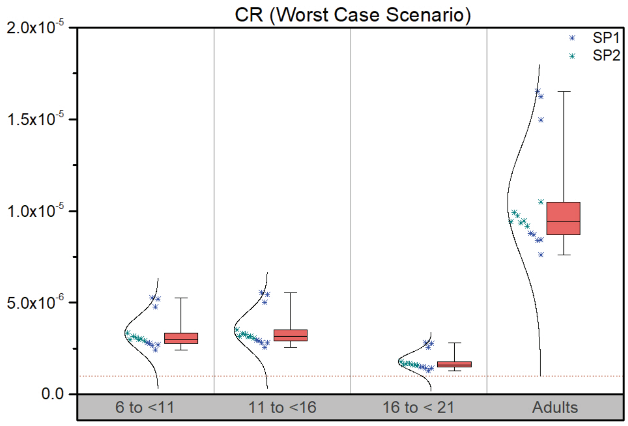

Figure 4. Bromate-associated CR levels of accidental ingestion of pool water (Worst Case Scenario).

CONCLUSION

Bromate ion is not taken into account in the TS EN 901 standard [43] for the chemical substances (sodium hypochlorite) used in the treatment of water. Depending on the manufacturing process and purity of chlorine salt, bromate contents of commercial sodium hypochlorite solutions may vary in broad range. The results of this monitoring study indicate that the bromate concentrations in pool waters are not dependent to bromide concentrations of filling waters, whereas co-dosing with commercial sodium hypochlorite solution might be the possible significant bromate contamination pathway in chlorinated pool waters.

Chronic-toxic risk (CTR) and carcinogenic risk (CR) levels associated with accidental ingestion of pool water were estimated for four age groups in central tendency and upper-bound (worst case) scenarios. Estimated CTR levels were lower than the threshold level even in maximum exposure case. However, synergistic effects of co-exposure to toxic compounds in swimming pools may result in increased chronic-toxic effects of bromate. While the CR levels in central tendency scenario were estimated to be in the safe zone (<10^{-6}), worst case scenario shows the bromate exposure pose a significant CR for specific (highly exposed) groups. The CR levels of lower age groups being higher than those of elders was due to their higher accidental ingestion rate and lower body weight. However, the highest CR levels were estimated for the adults due to the higher exposure frequency and exposure duration. Although lower CR were estimated in central tendency scenario, co-exposure to toxic compounds in pool waters might be pose a significant health risks due to the synergistic effects of co-exposure.

Bromate occurrence and associated health risk assessments shows that bromate should be considered in regulation of pool waters considering chemistry. Bromate contents of commercial sodium hypochlorite solutions should be regulated to avoid in-situ exposures from disinfected waters. Pool water replacement strategies and treatment systems should be optimized to reduce accumulation of non-volatile toxic compounds. Also, swimming pools in coastal zones, that are effected by seawater intrusion, should be specifically designed, and managed due to the highly toxic brominated by-product formation potentials of bromide ions.

AUTHORSHIP CONTRIBUTIONS

Authors equally contributed to this work.

DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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