

HUMAN HEALTH RISK ASSESSMENT OF TRIHALOMETHANES IN OUTDOOR SWIMMING POOL WATERS IN MONTENEGRO

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SUMMARY

Objectives: This paper deals with the characterization of outdoor swimming pool waters to study the occurrence of trihalomethanes (THMs) and to assess the health risk of THMs exposure for three population groups (male, female, and children). The concentration of total THMs and individual species (chloroform, bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform) were monitored during the seven months of 2022 in outdoor swimming pools in the coastal area of Montenegro.

Methods: A total of 73 water samples from outdoor swimming pools were collected and tested for the content of THMs using an Agilent 7890 A GC. Carcinogenic and non-carcinogenic health risks of total THMs and each species were assessed through lifetime cancer risk (CR) and hazard index (HI), respectively. Health risk assessment was assessed through the oral and dermal exposure pathways.

Results: The concentration of total THMs in investigated water samples ranged between 0.19 and 237.18 µg/L when 26% of tested water samples had values above the maximal permitted value (100 µg/L) established by Montenegrin legislation. Chloroform concentration was in a range of 1.13–217.16 µg/L, while contents of BDCM and DBCM were in a range of 1.01–29.43 µg/L and 0.18–16.19 µg/L, respectively. Calculated values of dermal CR were higher than 10^{-6} , thus CR values of total THMs exposure were higher than 10^{-6} for all population groups. On the other hand, CR via ingestion values were meagre, HI values of total THMs exposure for three population groups were calculated to be below 1.

Conclusion: There is no non-carcinogenic risk for the children population while male and female populations were faced with non-carcinogenic health risk of total THMs exposure. On the other hand, all investigated population groups were faced with unacceptable cancer health risks and dermal contact was recognized as the main exposure route for all investigated populations.

Key words: swimming pool, trihalomethanes, health risk assessment, ingestion, dermal contact

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INTRODUCTION

Since swimming is a popular recreational sport, disinfection of swimming pools is of extreme importance from the standpoint of human health protection. Among the different ways of pool disinfection, chlorination is the most popular and cost-effective way of pool water disinfection. Moreover, the residual chlorine from the disinfection process additionally protects water from further contamination (1). However, free chlorine reacts with a humic and fulvic acids present in water and form trihalomethanes (THMs) (2). THMs include chloroform, bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform which present potential danger to humans.

According to the International Agency for Research on Cancer (IARC), chloroform and bromodichloromethane are categorized as probably carcinogenic to humans (2B group) while dibromochloromethane and bromoform are categorized as possible human carcinogens (C group). Thus, special attention should be paid to the THMs content in swimming pool waters and possible adverse

effects on human health. The toxicological effect of THMs on human health is achieved through their absorption during human exposure to water and their bioaccumulation in adipose tissue, the liver, kidneys, and lungs (1, 3), which may lead to serious health problems like bladder, colon and rectal cancers (4). Due to the carcinogenic nature of THMs, different regulations and guidelines have been proposed for the THMs content in drinking water but there are no regulations concerning their content in swimming pool waters. However, the guideline values for drinking water set by the World Health Organization (WHO) can be used to assess potential risks arising from disinfection by-products from swimming pools (5). The guideline values for individual THM in drinking water set by the WHO (6) are 300 µg/L for chloroform, 60 µg/L for BDCM, and 100 µg/L for DBCM. However, Montenegrin regulations consider the value of total THMs of 100 µg/L in recreational water (swimming pool) and drinking water (7). The proposed values by Denmark, Germany and the U.S. Environmental Protection Agency (USEPA) are 50 µg/L, 20 µg/L and 80 µg/L of total THMs in drinking water, respectively (8, 9).

The formation of THMs in water depends on water parameters like pH, temperature and free chlorine. It was found that the increase of THMs concentrations in swimming pool waters is associated with the increase in temperature (10), residual chlorine and pH value of water (11).

According to the WHO, humans can be exposed to the THMs present in water by three exposure routes: ingestion (oral), dermal contact and inhalation (5). The ingestion route is greatly dependent on the age and skill experience and duration of exposure while the extent of uptake through the dermal contact depends on the temperature of water and the period of exposure (5), and generally, it is the same for indoor and outdoor pools. However, exposure through the inhalation route is greatly dependent on the type of pool environment. The inhalation route is greatly associated with the dispersion of THMs vapour in the air which is considerably high in outdoor swimming pools. Therefore, the concentration of THMs in the air above outdoor pools is low, so ingestion and dermal contact are the main routes of exposure to THMs in outdoor swimming pool water (12).

Over the years monitoring of THMs concentration and associated health risks were considered for drinking water, however, a limited number of studies were conducted for swimming pools mainly for indoor pools. To the best of our knowledge, there are no such studies for Montenegrin swimming pool waters at all. Thus, this study aimed to assess the THMs content in outdoor swimming pool waters in coastal area of Montenegro and the associated health risk.

MATERIALS AND METHODS

Data Collection

The study of THMs in hotel outdoor swimming pool waters was appraised in a 13 km² coastal area of Montenegro which included 6 towns (Ulcinj, Bar, Budva, Tivat, Herceg Novi, and Kotor) which are popular tourist hubs. The water samples were collected from 73 hotel outdoor swimming pools, during the period from March until October 2022.

Sampling of swimming pool waters was done using glass Winkler vessels capacity of 100 to 150 ml. For sampling, the Winkler vessel was immersed in a pool to a depth of about 30 cm. The vessel was filled to the top, then removed from the pool and closed tightly with a stopper, so that the water was forced out to remove air bubbles. The samples were stored at room temperature. The sampling and analyses were done in duplicate, for each swimming pool. The sampling was performed in a time interval from 2 p.m. to 3 p.m. with general high attendance of hotel guests in pools.

Determination of THMs Content in Swimming Pool Water Samples

Reagents and Solutions

Trihalomethanes mixture in methanol, used for the preparation of standard solutions, was purchased from ULTRA Scientific (USA). The concentration of each respective analyte in the mixture (CHCl₃, CHBrCl₂, CHBr₂Cl, and CHBr₃) was 100 mg/L. Stock standard solution of the mixture (1,000 µg/L) was prepared

in methanol (for HPLC, gradient grade ≥ 99.9%, Sigma-Aldrich, Germany) and used for making aqueous standard solutions (distilled water, HPLC purity) for calibration. The original mixture and standard stock solution were stored in a freezer at −17°C (± 3°C). Calibration was performed weekly. Calibration levels were: 0 µg/L, 2 µg/L, 5 µg/L, 10 µg/L, 20 µg/L, and 50 µg/L. The standard solutions were prepared in 10 ml glass volumetric flasks (class A, ISOLAB, Germany) using Eppendorf Research Plus single channel pipettes, 10–100 µl and 100–1,000 µl, respectively.

HS-GC-ECD Method

Agilent 7890 A GC with an electron capture detector (ECD) and static headspace was used for the determination of THMs content. THMs were determined directly from pool water samples, without any specific pre-preparation. Using the previously mentioned 100–1,000 µl pipette, 10 ml of water sample was placed in a 20 ml crimp-top glass headspace vial (Agilent, USA). Nitrogen was used as a carrier gas, with a 35 ml/min flow. For the separation of THM compounds a SLB-5 ms capillary column (Sigma-Aldrich, Germany) with 30 m × 0.25 mm × 0.25 µm dimensions, was used. The total time of a single GC cycle was 18 minutes, of which 10 minutes were for vial equilibration. The duration of the injection was 0.3 minutes. The injected volume was 2 µl. The temperature of the injection inlet was 170°C and the split mode was used, with a 50 : 1 split ratio. The starting oven temperature was 40°C and was programmed to rise at: 15°C/min to 65°C, 5°C/min to 120°C and finally 30°C/min to 180°C, without prolonged maintenance at any specific temperature. The temperature of ECD was held at 300°C. Before each analysis, the column was heated at 300°C for 15 min period to remove any possible remaining contamination.

Health Risk Assessment

The health risk assessment for THMs in swimming pool water was estimated through oral ingestion and dermal exposure pathways. The chronic daily intake via ingestion and dermal contact for three population groups (male, female, and children aged 11–14 years) were estimated according to the next equations.

$$CDI_{ing} = \frac{C \cdot IR \cdot ET \cdot EF \cdot ED}{BW \cdot AT} \cdot CF \quad (1)$$

$$CDI_{der} = \frac{C \cdot SA \cdot Kp \cdot ET \cdot EF \cdot ED}{BW \cdot AT} \quad (2)$$

where CDI_{ing} is chronic daily intake via ingestion (mg/kg/day), C is the concentration of THMs in water (µg/L), IR is water intake, ET is the exposure time, EF is the exposure frequency, ED is the exposure duration, BW is body weight, AT is the average exposure time, CF is the conversion factor from micrograms to milligrams (0.001), SA is the skin surface area exposed to THMs, and Kp is the dermal permeability coefficient.

Carcinogenic and non-carcinogenic health risks of each species were assessed as lifetime cancer risk (CR) and hazard index (HI), respectively, according to Eq. 3 and 4.

$$CR = \sum_{i=1}^m \sum_{j=1}^n CDI_{ij} \cdot Sfi \quad (3)$$

$$HI = \sum_{i=1}^m \sum_{j=1}^m CDi / RfDi \quad (4)$$

where m presents each one of the three considered THMs; n is a specific exposure route; $RfDi$ and Sfi are the reference daily dose and cancer slope factor for each specie. Factors used in the health risk assessment equations are given in Table 1. Values of IR , ET , ED , BW , AT , and SA were taken from a USEPA source (13), and values of Kp , RfD and Sf were obtained from the Integrated Risk Information System (IRIS) (14). If the HI value is calculated to be less than 1, the risk exposure is considered to be controlled at the given exposure scenario. If the calculated HI value is more than 1, human health is of concern. Cancer risk is defined in four classes: negligible risk ($CR < 10^{-6}$), acceptable low risk ($1 \times 10^{-6} \leq CR < 5.1 \times 10^{-5}$), acceptable high risk ($5.1 \times 10^{-5} \leq CR < 10^{-4}$), and unacceptable risk ($CR \geq 10^{-4}$) (15).

RESULTS

THMs Concentration in Swimming Pool Waters

The concentration of THMs and their compounds (chloroform, BDCM and DBCM) in hotel outdoor swimming pool water samples collected from different sampling sites in the coastal area of Montenegro are given in Figure 1 and Table 2. Bromoform was detected in negligible concentrations only in 9 out of 73 locations of water samples collected from outdoor swimming pools (Fig. 1). In other water samples bromoform concentration was below the limits of detection and thus, bromoform was not considered in the evaluation of health risk.

The results presented in Figure 1 and Table 2 indicate that chloroform accounts for 89.6% of the average total THMs concentration in swimming pool waters with a concentration ranging from 1.13 to 217.16 $\mu\text{g/L}$ and an average concentration of 58.69 $\mu\text{g/L}$. BDCM and DBCM contribute to total THMs content with 10.9% and 3.7% with an average concentration

Table 1. Factors used in the risk assessment equations

Factor	Value		
	Males	Females	Children
IR ($\text{L} \cdot \text{h}^{-1}$)	0.025	0.025	0.05
ET (h/event)	1	1	1
EF (events/year)	120	120	120
ED (year) non-carcinogenic health risk	1	1	1
ED (year) carcinogenic health risk	70	70	–
BW (kg)	78.1	65.4	48.17
AT (days)	$ED \times EF$	$ED \times EF$	$ED \times EF$
SA (cm^2)	1.94	1.69	1.42
Kp (chloroform), (cm/h)	8.9×10^{-3}		
Kp (BDCM), (cm/h)	5.8×10^{-3}		
Kp (DBCM), (cm/h)	3.9×10^{-3}		
RfD (chloroform), (mg/kg/day)	0.01		
RfD (BDCM) ingestion, (mg/kg/day)	0.02		
RfD (DBCM) ingestion, (mg/kg/day)	0.02		
Sf (chloroform) ingestion, (mg/kg/day)	6.1×10^{-3}		
Sf (BDCM) ingestion, (mg/kg/day)	6.2×10^{-2}		
Sf (DBCM) ingestion, (mg/kg/day)	8.4×10^{-2}		
Sf (chloroform) dermal, (mg/kg/day)	3.1×10^{-2}		
Sf (BDCM) dermal, (mg/kg/day)	6.33×10^{-2}		
Sf (DBCM) dermal, (mg/kg/day)	1.40×10^{-1}		

of 7.11 $\mu\text{g/L}$ and 2.48 $\mu\text{g/L}$, respectively. The concentration of total THMs in evaluated outdoor swimming pool water samples was in the range of 0.19 to 237.18 $\mu\text{g/L}$ with an average value of 65.47 $\mu\text{g/L}$ (Table 2).

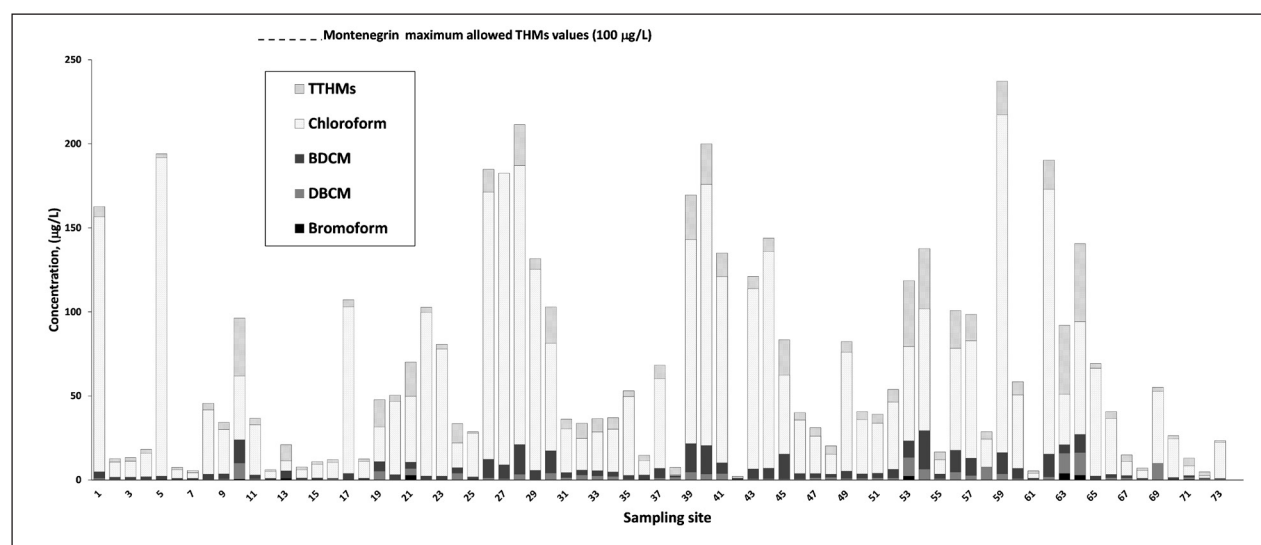


Fig. 1. Concentration of total THMs and their compounds (chloroform, BDCM, DBCM and bromoform) in outdoor swimming pool water samples collected from different sampling sites.

TTHMs – total trihalomethanes; BDCM – bromodichloromethane; DBCM – dibromochloromethane

Table 2. THMs ($\mu\text{g/L}$) content in outdoor swimming pool water (coastal area of Montenegro 2022)

Statistical indicator	Total THMs	Chloroform	BDCM	DBCM
Average	65.47	58.69	7.11	2.48
Mediana	40.71	36.02	3.93	1.03
Min	0.19	1.13	1.01	0.18
Max	237.18	217.16	29.43	16.19
SD	59.16	55.19	7.21	3.57

THMs – trihalomethanes; BDCM – bromodichloromethane; DBCM – dibromochloromethane

Carcinogenic Health Assessment

The results of carcinogenic health risks caused by the exposure to the THMs in outdoor swimming pool waters for men and women via ingestion and dermal contact are given in Table 3. The results show that both groups of population were faced with

certain carcinogenic health risks, as calculated CR values were higher than 10^{-6} via both exposure routes. The women were faced with slightly higher overall cancer risk in comparison to men since the obtained CR values (5.26×10^{-6}) for female part of population were higher than CR values (5.01×10^{-6}) obtained for male part of population. In similar fashion CR values for each of exposure pathways (dermal CR value and oral CR value) were marginally higher for women. Dermal contact was recognized as the major exposure route for both populations with a contribution of 94% for men and 93% for women, while contributions of ingestion route were 6% and 7%, respectively (Fig. 2 a-c).

The results presented in Table 3 indicate that cancer risk for total THMs through ingestion was negligible ($\text{CR} < 1 \times 10^{-6}$), given that average calculated values were 3.17×10^{-7} for men and 3.79×10^{-7} for women. The highest risk through ingestion route for both populations comes from BDCM (44%) followed by chloroform (36%) and DBCM (20%) (Table 3). Calculated cancer risks of BDCM through ingestion route for women and

Table 3. Lifetime cancer risk of THMs through two exposure routes and contribution of THM species to total THMs

Population	Exposure pathway	Chloroform	BDCM	DBCM	Total THMs
Male	Ingestion	1.15×10^{-7} (36%)	1.38×10^{-7} (44%)	6.45×10^{-8} (20%)	3.17×10^{-7}
	Dermal	4.02×10^{-6} (86%)	6.34×10^{-7} (14%)	3.26×10^{-8} (1%)	4.69×10^{-6}
	Total	4.14×10^{-6}	7.72×10^{-7}	9.71×10^{-8}	5.01×10^{-6}
Female	Ingestion	1.37×10^{-7} (36%)	1.65×10^{-7} (44%)	7.71×10^{-8} (20%)	3.79×10^{-7}
	Dermal	4.18×10^{-6} (86%)	6.59×10^{-7} (14%)	3.39×10^{-8} (1%)	4.88×10^{-6}
	Total	4.32×10^{-6}	8.24×10^{-7}	1.11×10^{-7}	5.26×10^{-6}

THMs – trihalomethanes; BDCM – bromodichloromethane; DBCM – dibromochloromethane

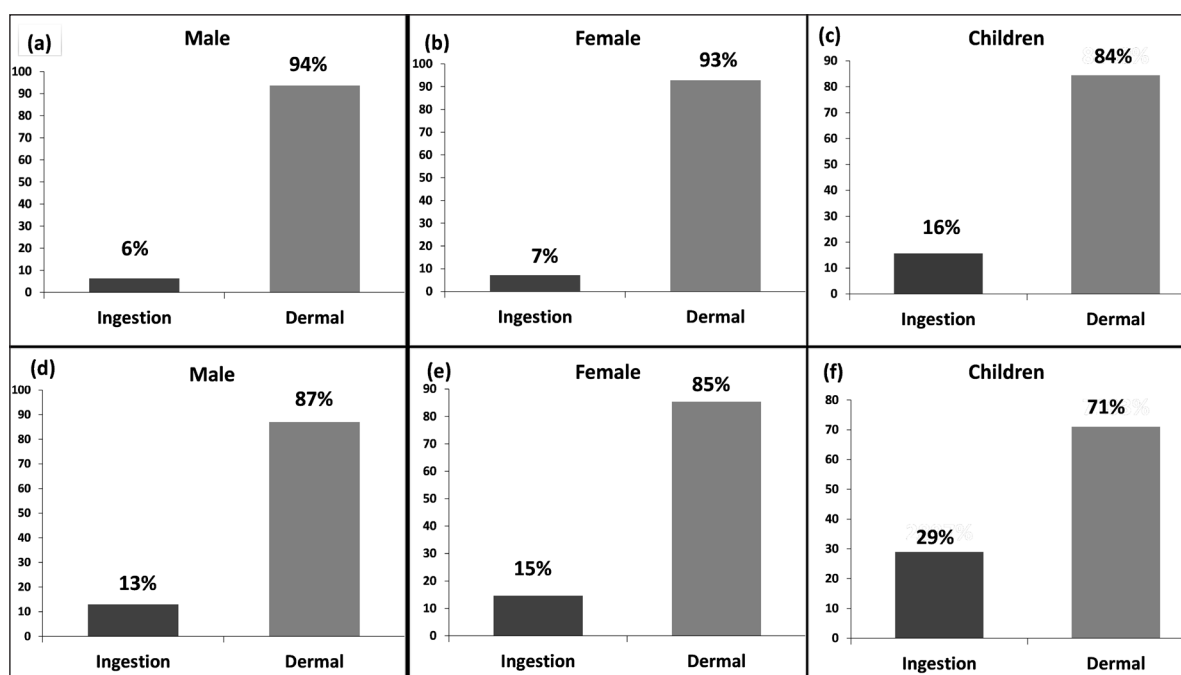


Fig. 2. Contribution of each exposure pathway to carcinogenic health risk (a), (b) and (c), and contribution of each exposure pathway to non-carcinogenic health risk (d), (e) and (f).

men were 1.65×10^{-7} and 1.38×10^{-7} , respectively. The cancer risks through ingestion of chloroform in swimming pool waters for women and men were 1.37×10^{-7} and 1.15×10^{-7} , respectively. The lowest cancer risk via ingestion for both populations was related to DBCM (10^{-8} orders of magnitude).

The cancer risk of total THMs through dermal contact was acceptably low for both women (4.88×10^{-6}) and men (4.69×10^{-6}) (Table 3). We can see that these values are remarkably higher compared to CR values via ingestion. The highest risk through dermal permeability for two populations present by far chloroform (86%) with values of 4.02×10^{-6} for men and 4.18×10^{-6} for women, followed by BDCM (14%) and almost insignificantly DBCM (1%). The contribution of BDCM to the cancer risk of total THMs via dermal contact was considerably lower (14%) in comparison to the contribution via the ingestion route (44%) for two population categories. Calculated dermal cancer risk values of BDCM and DBCM were 6.34×10^{-7} and 3.26×10^{-8} individually for men along with 6.59×10^{-7} and 3.39×10^{-8} for women, respectively.

Non-carcinogenic Health Risk

The obtained results for non-carcinogenic health risk presented in Table 4 indicate that non-carcinogenic risk was not of apparent concern for three population categories (men, women and children), since hazard indexes of THMs through both exposure routes were below the acceptable maximum value of 1. Contribution of dermal contact to the total non-carcinogenic health risk of THMs exposure was 87%, 85% and 71% for men, women and children, while contributions of ingestion route for these three populations were 13%, 15% and 29%, respectively (Fig 2. d-f). The HI values for the male and female population were quite similar, but somewhat higher for women (1.56×10^{-2} for males and 1.66×10^{-2} for females), with remarkably higher values of dermal HI (1.36×10^{-2} for males and 1.41×10^{-2} for females) compared to those of HI via ingestion (2.03×10^{-3} for males and 2.42×10^{-3} for females) in total HI sum. When it comes to the children, we can see that total HI value of 2.27×10^{-2} is clearly the highest out of the three, which makes children the most at-risk population group in this situation. In parallel manner dermal HI value (1.61×10^{-2}) is more substantial than HI via ingestion value (6.58×10^{-3}).

When it comes to the risk of individual THMs, chloroform presented the highest risk through dermal contact, with the same contribution value to total THMs for all three populations (Fig.

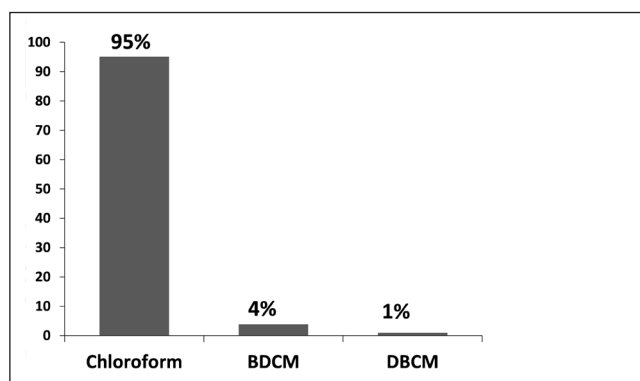


Fig. 3. Contributions of each species to non-carcinogenic risk to total THMs via dermal contact for male and female populations.

THMs – trihalomethanes; BDCM – bromodichloromethane; DBCM – dibromochloromethane

3). Chloroform contributed to total THMs with 95% while the contribution of BDCM and DBCM was 4% and 1%, respectively.

DISCUSSION

This study aimed to determine the concentration of THMs in the hotel outdoor swimming pools of the coastal area of Montenegro and to assess the health risk of total and individual THMs exposure through ingestion and dermal contact for three subgroups of swimmers: adults (men and women) and children.

As mentioned in Figure 1 and Table 2, chloroform was the most abundant trihalomethane in analysed samples. This finding is in accordance with the results of other studies with similar topic (2, 12, 16–18). The average concentration of total THMs obtained in this study was in the range reported for outdoor swimming pools (68–131 µg/L) (19).

The results presented in Figure 1 indicate that the concentration of total THMs in 26.0% of water samples exceeded proposed permissible value of 100 µg/L according to the Montenegrin legislation (7), while 42.4%, 72.6% and 34.2% of water samples exceeded the limits proposed by Danish (50 µg/L), German (20 µg/L) and US (80 µg/L) standards (8, 9).

On the other hand, the concentration of each individual trihalomethane was below the prescribed respective limit proposed

Table 4. Hazard index of THMs through two exposure routes and contribution of THM species to total THMs

Population	Exposure pathway	Chloroform	BDCM	DBCM	Total THM
Male	Ingestion	1.88×10^{-3}	1.11×10^{-4}	3.84×10^{-5}	2.03×10^{-3}
	Dermal	1.30×10^{-2}	5.01×10^{-4}	1.16×10^{-4}	1.36×10^{-2}
	Total	1.49×10^{-2}	6.12×10^{-4}	1.55×10^{-4}	1.56×10^{-2}
Female	Ingestion	2.24×10^{-3}	1.33×10^{-4}	4.59×10^{-5}	2.42×10^{-3}
	Dermal	1.35×10^{-2}	5.21×10^{-4}	1.21×10^{-4}	1.41×10^{-2}
	Total	1.57×10^{-2}	6.54×10^{-4}	1.67×10^{-4}	1.66×10^{-2}
Children	Ingestion	6.09×10^{-3}	3.61×10^{-4}	1.25×10^{-4}	6.58×10^{-3}
	Dermal	1.54×10^{-2}	5.94×10^{-4}	1.38×10^{-4}	1.61×10^{-2}
	Total	2.15×10^{-2}	9.55×10^{-4}	2.63×10^{-4}	2.27×10^{-2}

THMs – trihalomethanes; BDCM – bromodichloromethane; DBCM – dibromochloromethane

by the WHO (300 µg/L for chloroform, 60 µg/L for BDCM, and 100 µg/L for DBCM) (6). These results indicate the necessity of improving the treatment of pool water and controlling the concentration of THMs in swimming pool water.

When it comes to the contribution of each considered exposure pathway (dermal and oral), according to obtained results skin penetration plays the main role, with approximately 90% of total contribution to both total carcinogenic and total non-carcinogenic health risk for THMs. This pattern is in agreement with other literature sources (19, 20).

When it comes to the carcinogenic health risk, displayed results revealed an existence of acceptable, low carcinogenic health risk of exposure to total THMs for male and female populations. The highest cancer risk for both populations through exposure via ingestion is caused by BDCM, while the highest risk through dermal contact presents chloroform. It was determined that dermal contact was the main exposure route for both of evaluated populations. Also, it is important to mention that the female swimmers were slightly more at the risk of exposure to THMs than the male swimmers. Overall, we can see that while total cancer risk exists for both groups, the values are not extreme nor unacceptable. However, despite this, they still should not be taken lightly.

When it comes to non-carcinogenic health risk, it was determined that existing risk was not overly extreme for any of the three population groups. More notable risk was observed for children, compared to the male and female population, where we had similar values, narrowly higher for women. Dermal contact was identified as the major contributor to non-carcinogenic health risk.

Also, it is necessary to note the fact that another major pathway of exposure to the THMs in swimming pools, inhalation, was not a part of consideration in this study. It is safe to assume that had it been considered the obtained results for both CR and HI would most likely have been higher. Clearly, more progress in the fields of measuring, monitoring and controlling DBPs through legislation and prevention is necessary in Montenegro, as well as the rest of the world.

CONCLUSIONS

This study aimed to determine the concentration of THMs in the hotel outdoor swimming pools of the coastal area of Montenegro and to assess the health risk of total and individual THMs exposure through ingestion and dermal contact for three populations of swimmers: males, females and children. The results indicated that in the 26% samples of swimming pool waters, the measured concentration of total THMs exceeded the values of 100 µg/L which is prescribed by Montenegrin legislation. The study revealed an existence of acceptable and low cancer risk of exposure to total THMs for male and female populations. It was determined that dermal contact was the main exposure route for all investigated populations. The highest cancer risk for all populations through exposure via ingestion is caused by BDCM, while the highest risk through dermal contact presents chloroform.

When it comes to non-carcinogenic health risk, it was determined that existing risk was not overly extreme for none of the three population groups. More notable risk was observed for children, compared to the male and female population, where we had similar values, narrowly higher for women. However, for all

three populations total non-carcinogenic risk via two key routes (dermal and oral) was also determined for both adults and children, with dermal contact being identified as the major contributor to non-carcinogenic health risk.

In conclusion, knowledge about the concentration of THMs in swimming pool waters and the factors affecting their levels may help public health authorities with the improvement of the hygiene and safety conditions of swimming facilities. Additionally, it is necessary to consider the fact that analysed THMs represent only four of several hundred (so far confirmed) disinfection by-products found in swimming pools and that they are far less complex and dangerous compounds than the vast majority of other DBPs. Therefore, they represent just tiny portion of the total risk posed by DBPs to swimmers. Clearly, more progress in the fields of measuring, monitoring and controlling DBPs through legislation and prevention is necessary in Montenegro, as well as the rest of the world.

Conflicts of Interest

None declared

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