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# Comparison of trihalomethanes in the air of two indoor swimming pool facilities using different type of chlorination and different types of water

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#### ABSTRACT

Certain aspects of the distribution of disinfection by-products (DBPs) in the air of indoor swimming pools, the exposure of the users, and possible health effects have not been well documented. To determine the distribution of trihalomethanes (THM), measurements were performed at 0.05 m, 0.60 m and 1.50 m above the water surface. These heights were chosen to measure the exposure in the breathing zone of the users. Air samples were collected from two indoor swimming pool facilities in Norway. Facility 1 uses calcium hypochlorite and facility 2 uses sodium hypochlorite for water treatment. In facility 2, one of the swimming pools is filled with 33% seawater, while the other pools in this study were filled with freshwater. Higher values were measured at 0.05 m compared to 1.50 m. Negligible differences between the measurements at 0.60 m and 1.50 m above floor levels were obtained. In average, 282% higher concentrations of total THM (tTHM) were measured in facility 2. Different disinfection products and ventilation concepts are possible explanations. Swimmers are exposed to higher concentrations compared to users by the poolside. For future studies, it is crucial to measure as close to the water surface as possible.

Keywords; Distribution of THM, seawater, sodium hypochlorite, calcium hypochlorite, brominated THM, ventilation

# **INTRODUCTION**

Chlorine-based water treatment is the most common disinfectant used in swimming pool water. The method reduces the risk of exposure to pathogens present in the water such as bacteria, parasites and fungus. If these pathogens are not removed, they can cause severe health effects (World Health Organization, 2006). The reaction between chlorine and organic matter from the occupants such as hair, lotions, mucks, skin extraction and urine creates disinfection by-products (DBPs). The presence of DBPs in the poolroom has been linked to a variety of health issues (World Health Organization, 2006; Kogevinas, 2010; Manasfi et al., 2017). Among the most important groups within DBPs are trihalomethanes (THMs), where chloroform (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM) and bromoform (TBM) are most abundant. Of the four THMs, TCM is the compound generally present in the greatest concentration (World Health Organization, 2006). However, adding seawater to the pool increases the amount of brominated DBPs making TBM the most dominant THM ( Parinet et

al., 2012; Ged & Boyer, 2014; Chowdhury, 2015; Manasfi et al., 2016). Studies show that increased formation of brominated THMs occurs in chlorinated bromide rich waters (Hua et al., 2006; Ged & Boyer, 2014). Another factor associated with increased amount of brominated DBPs in swimming pool water is the use of sodium hypochlorite for water disinfection if the brine solution used for the production contain bromide ions (Bouland et al., 2005). The brominated THMs are in general more genotoxic and mutagenic compared to their chlorinated analogues (Kogevinas, 2010, Manasfi et al., 2017).

Exposure to DBPs in pools can take place through inhalation, dermal contact and to a lesser extent by accidental swallowing of water (Chen et al., 2011; Chowdhury, 2015). Occupants in the pool inhale more concentrated air in comparison to employees and visitors staying by the poolside. Besides that, the employees and athletes will be more exposed due to longer exposure time. To determine health effects related to DBPs, it is crucial to measure the concentration of THMs directly above the surface of the water, meaning the breathing zone of the swimmer, as high activity provides high pulmonary ventilation (World Health Organization, 2006). The aim of the present study was to measure the concentration of THMs 0.05 m, 0.60 m and 1.50 m above the pool water surface, representing the breathing zone of the swimmers, as well as children and adults standing by the pool side, and to compare the distribution of THMs from three freshwater pools and one seawater pool using different disinfection principles.

# METHOD

Four swimming pools in Norway, located in two public indoor swimming facilities, were selected for this study. They were chosen to make a comparison between different disinfection methods possible, as facility 1 (S1) uses granulated calcium hypochlorite ( $Ca(OCl)_2$ ) while facility 2 (S2) uses liquid sodium hypochlorite (NaOCl). The latter includes also a pool where the water is a mixture of 67% freshwater and 33% seawater. Samples were collected between February 13, 2017 and March 10, 2017. Above each pool, between 15 and 26 samples were collected. In table 1, physicochemical parameters and number of bathers for the different swimming pools are listed. The two swimming facilities are referred to as S1 and S2:

	S	51	<u>\$2</u>		
	P1	Р2	Р3	P4	
Water volume [in m <sup>3</sup> ]	600	200	2450	210	
Disinfectant	Ca(OCl) <sub>2</sub>	$Ca(OCl)_2 + UV$	NaOCl + UV	NaOCl + UV	
Water temperature [in °C]	26.5-26.8	32.9 - 33.1	28.3 - 28.9	33.9 - 34.6	
Air temperature [in °C]	27.0 - 28.5	27.5 - 29.0	28.0 - 30.3	29.6 - 32.3	
рН	7.22 - 7.45	7.37 - 7.87	7.20 - 7.35	7.15 - 7.23	
Free chlorine [in mg/L]	0.77 - 0.89	1.03 - 1.44	0.45 - 0.51	0.95 - 1.04	
Combined chlorine [in mg/L]	0.15 - 0.25	0.02 - 0.42	0.02 - 0.06	0.11 - 0.31	
Number of users	5 - 25	0 - 22	5 - 25	22 - 50	
Fresh/seawater ratio	1/0	1/0	0.67/0.33	1/0	

Table 1: Physicochemical parameters and number of bathers at the different swimming pools

In S1 pool 1 and 2 are in the same room. In addition, there is also a whirlpool and a small padding pool. Approximately 130 000 visitors are visiting S1 per year. Both pool 1 and 2 use drinking water from the public water supply. The results are based on 41 air samples collected from S1 during the four weeks of sampling. In S2, pool 3 is in the same hall as four other

swimming pools; including a wave pool, a slide pool and several whirlpools and fountains. Pool 4 is in its own room with its own ventilation system but is supplied with disinfectant water from the same circulation system as pool 3. In total, approximately 360 000 people visit S2 per year. Besides 33% of seawater being used in pool 3, both pool 3 and 4 use drinking water from the public water supply. The results are based on 41 air samples collected from S2 during the four weeks of sampling.

Both S1 and S2 uses the same type of ventilation system and the ventilation flow rate is constant. Supplied amount of fresh air is controlled according to the humidity and the temperature in the rooms.

#### Sampling

Each sampling day, stationary air samples were collected during morning swimming (0630 - 0930 AM), baby swimming (1430 - 1730 PM) and public swimming (1730 - 1830 PM). Both the morning swimming and baby swimming took place in the therapy pools (pool 2 and 4) while public swimming occurred in the sport pools (pool 1 and 3) of the two facilities. Three samples were collected simultaneously using a test stand with three different heights, 0.05 m, 0.60 m and 1.50 m above the pool water. The test stand was placed by the pool side and in the middle of the long side of the pool, at the same place during all measurements.

Air samples where obtained by pulling air through stainless steel tubes containing 0.20 g of Tenax TA 35/60 (Markes International), using low flow pumps with an average flowrate of 50 ml/min for 20 min to obtain air volumes of 1 liter. The pumps were calibrated in situ, before and after each sample. The tubes were immediately recapped after collection using Swagelok caps combined PTFE ferrules to avoid losses. After sampling, the tubes were wrapped in uncoated aluminum foil and placed in an airtight container with charcoal to avoid contamination. This is in accordance with recommendations of the standard US EPA Method TO-17 (United States Environmental Protection Agency, 1999). Tube number and placement, pool, activity, free chlorine, combined chlorine, pH, air- and water temperature, number of users and relative humidity where recorded when the samples were collected.

Information on water temperature, free and combined chlorine and pH are recorded automatically and continuously in the two pool facilities and these values were read immediately after sampling. The air temperature was measured in situ with a portable instrument (AirMeterTM TestT001, Fluke).

# Laboratory analysis

The sampling and analysis of THM levels in air were based on the US EPA Method TO-17 (US EPA, 1999) and ISO 16017 (International Organization for Standardization, 2000). Determination of THMs in the air was performed by using Unity thermal desorber (Markes International) coupled with Agilent Technologies 5975T Low Thermal Mass (LTM) Gas Chromatography/Mass Selective Detector (GC/MSD). Thermal desorption was carried out for 10 min at 284 °C with a flow rate of 30 ml/min and to a cold trap packed with Tenax TA. Secondary desorption was carried out with a carrier gas flow rate of 20 ml/min from the trap. The THMs where submitted to a 3.7:0.7 split ratio. The separation was performed on a capillary column (DB-1; ID 0.25 mm and 0.25  $\mu$ m film thickness). The oven temperature was running with a temperature program from 35 °C to 90 °C with 5 °C/min steps with post run at 230 °C. Identification and quantification of THMs were performed in selection ion monitoring (SIM) mode. The results are based on 82 of 87 collected samples. Five samples were excluded due to tube leakage during the pressure test before desorption. The analysis of THM was performed

in the laboratory of the Department of Health, Safety and Environment at the institute of industrial economy and technology management at NTNU.

#### Method validation and quality assurance

Both external and internal calibration method were performed. For internal calibration, the sorbent tubes were spiked with 250 ng 8260 Internal Standard Mix 2 (Supelco) containing fluorobenzene, chlorobenzene-d5, 1.4- dichlorobenzene-d4 in methanol. For external calibration, a five-point calibration curve was made, ranging from 0.5 ng to 500 ng, for each of the four THMs using Trihalomethanes Calibration Mix (Supelco) diluted with methanol (n = 30). Using this method, a limit of quantification (LOQ) of 0.5  $\mu$ g/m<sup>3</sup> and a linear range from 0.5  $\mu$ g/m<sup>3</sup> to 500  $\mu$ g/m<sup>3</sup> was obtained for all four THMs. In accordance with US EPA Method TO-17, all duplicate measures and volume pairs of tubes have been within the precision of 5%. The test for breakthrough was carried out weekly to verify that less than 5% of the target analytes were observed on any of the back-up tubes (US EPA, 1999). The calibration check was performed during and after the measure campaign was finished.

# Statistical analysis

For descriptive purpose, arithmetic mean (AM) and standard deviation (SD) were calculated for the THM concentrations using Statistical Package for Social Sciences (SPSS) 24.00. Spearman rank correlation was used to estimate the correlation between the various variables and a one-way analysis of variance (ANOVA) model was used to estimate the swimming pool variance components. Independent T- tests were used to investigate if the two facilities were significantly ( $p \le 0.05$ ) different from each other.

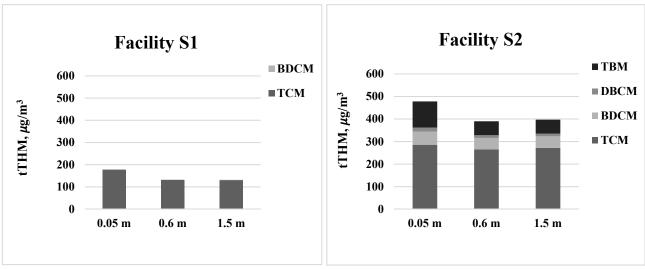
# **RESULTS AND DISCUSSION**

A summary of the measured parameters is shown in table 2. The concentration of combined chlorine was always in agreement with the Norwegian guidelines (< 0.50 mg/L). The final day of sampling, the measured level of pH in S1 exceeded the required value in the Norwegian guideline (7.20 - 7.60). In pool 3, one sample of free chlorine showed 0.45 mg/L which is lower than the limit value of 0.50 mg/L (Norwegian Ministry of Health, 1996).

Pool, Facility	Activity	n	RH (%)	рН	F <sub>chlorine</sub> (mg/L)	B <sub>chlorine</sub> (mg/L)	No. of Bathers	T <sub>air</sub> (°C)	T <sub>water</sub> (°C)	tTHM, (μg/m <sup>3</sup> )
Pool 2, S1	Morning swimming	11	66.4	7.5	1.26	0.20	2.2	27.5	33.0	145,8
Pool 2, S1	Baby swimming	12	60.3	7.5	1.15	0.23	14.3	28.3	33.1	122,4
Pool 1, S1	Public swimming	18	57.2	7.3	0.82	0.21	13.4	27.9	26.7	168,4
Pool 4, S2	Morning swimming	14	57.5	7.2	0.99	0.20	25.2	31.8	34.2	435,5
Pool 4, S2	Baby swimming	12	58.8	7.2	0.99	0.20	41.4	31.2	34.1	267,3
Pool 3, S2	Public swimming	15	66.8	7.3	0.49	0.04	15.6	29.0	28.6	528,1

*Table 2: Average chemical-physical parameters and number of bathers of swimming pool waters for each activity in S1 and S2* 

Abbreviations: n; samples of tTHM, RH; relative humidity, F<sub>chlorine</sub>; Free chlorine, B<sub>chlorine</sub>; Combined chlorine, T<sub>air</sub>; Air temperature, T<sub>water</sub>; Water temperature, tTHM: Average concentration (all heights)



### Concentration of THM 0.05 m, 0.60m and 1.5 m above water surface

Figure 1: The average tTHM for both pools and both facilities.

Figure 1 shows that significantly higher levels of tTHM were measured in S2. Only TCM and BDCM were quantified on all samples in S1 (pool 1 and 2), while DBCM and TBM was either not detected or under the limit of quantification (LOQ). In S1, at 0.05 m above the water surface, the concentration of tTHM was in the range of 162.5  $\mu$ g/m<sup>3</sup> - 316.4  $\mu$ g/m<sup>3</sup> and 95.4  $\mu$ g/m<sup>3</sup> - 202.7  $\mu$ g/m<sup>3</sup> for pool 1 and 2 respectively. For S2 the concentration of tTHM was in the range of 448.3  $\mu$ g/m<sup>3</sup> - 781.7  $\mu$ g/m<sup>3</sup> and 210.6  $\mu$ g/m<sup>3</sup> - 638.7  $\mu$ g/m<sup>3</sup> for pool 3 and 4 respectively.

Both S1 and S2, the concentrations measured at 0.60 m and 1.50 m above the water surface were almost identical. In S1, the concentrations were measured from  $89.2 \ \mu g/m^3 - 199.7 \ \mu g/m^3$ . Former studies which measured the concentration of tTHM at the same heights found values between 44.0  $\mu g/m^3 - 260.8 \ \mu g/m^3$  (Fantuzziet al., 2010; Richardson et al., 2010; Lourencetti et al., 2012; Chowdhury, 2015; Marco et al., 2015; Manasfi et al., 2017). In S2, higher values were found, ranging from 205.0  $\mu g/m^3 - 676.1 \ \mu g/m^3$ . High concentrations were also measured in other studies (Bessonneau et al., 2011; Tardif et al., 2016). However, the mean concentrations of tTHM obtained in these studies were lower compared to the mean concentrations measured in S2. For S1 and S2, average concentrations of tTHM measured at 0.05 m above the water surface showed 37% and 20% higher values than the corresponding value found 1.50 m above water surface.

As shown in table 2, TCM was found to be the dominant compound in S1 (between 97% - 99%), followed by BDCM. In S2, all four THMs were present in all samples and like for S1, TCM was found to be the dominant compound. However, in pool 4 (100% freshwater pool), tTHM in the air was comprised of 25% brominated THMs. Higher fractions of brominated THMs in the air above pool 3 were anticipated due to the source water quality (mixture of 33% seawater and 67% freshwater). For this seawater pool (pool 3), the fraction of brominated THM in air samples was 55.6% (0.05 m), 40.9% (0.60 m) and 39.0% (1.50 m). Past studies have reported approximately 95% brominated THMs in seawater pools (Parinet et al., 2012; Chowdhury, 2015). In a former study where THMs above one pool containing seawater were measured, TBM was found to account for 85% (0.60 m) of tTHM (Chowdhury, 2015). In pool 3, a smaller amount of brominated THMs was found possible due to only 33% seawater in the pool. A former study found that even small amounts of salt can increase the amount of bromide

present in the water significantly (Ged & Boyer, 2014). As brominated THMs were found to represent approximately 25% of tTHMs in pool 4 (freshwater pool), brominated THMs seems to be associated with the use of sodium hypochlorite for water treatment, possibly due to bromide ions in the brine used to produce sodium hypochlorite.

# The distribution of THMs

Table 2 shows the average concentration of combined chlorine in pool 3 (seawater pool) measured to be 0.04 mg/L, which is lower compared to the concentration of combined chlorine measured in the other pools. In pool 3, UV- treatment is used as a secondary disinfectant and it is well known that UV- treatment lamps might reduce the concentration of combined chlorine in the pool water (Cassan et al., 2006). This could, to some extent, explain the low concentration of combined chlorine was much greater (0.20 mg/L - 0.23 mg/L). Since pool 3 contains 33% seawater, it is likely that high concentrations of bromide are present in the water. When chlorine is added to bromide rich waters, bromide ions oxidises yielding hypobromous acid (Taylor, 2006). If hypobromous acid react with ammonia in the water, this might form bromamines. It is therefore questioned whether combined chlorine is a good marker for haloamines present in the pool water, as combined bromine might also be present. Most European guidelines only regulate the concentration of free and combined chlorine in the water, it may be questioned if brominated compounds also should be regulated in bromide rich waters.

For pool 3, little difference between the concentration of TCM was measured between the three heights, while the concentration of TBM was measured to be 226% higher at 0.05 m compared to 1.50 m above the water surface. TBM is less volatile than TCM (Beech et al., 1980), and it was therefore expected to find higher concentrations of TBM directly above the pool water surface. These findings emphasize how important it is to measure as close to the water surface as possible, especially considering the increased toxicity related to brominated compounds. As stated above, significantly higher concentrations of tTHM were measured in S2 compared to S1 (on average 282%). In a former study, a strong correlation was found between the number of users in the swimming pool and the concentration of tTHM (Chu & Nieuwenhuijsen, 2002). However, for S2, the correlation was found to be significantly negative. In S1, no correlation was obtained. Presence of brominated compounds is found to increase the amount of tTHMs (Hua et al., 2006; Ged & Boyer, 2014), this could, to some extent, explain the high concentration of tTHM in S2. However, this hypothesis should be further tested before any conclusions are made. Different in ventilation is another possible explanation for this wide range of tTHM concentration. S1 and S2 uses the same type of ventilation system and the air changes per hours of fresh air is almost equal for both facilities. However, slides, fountains, wave pool and higher air- and water temperature in S2 are activities likely to promote the dispersion of tTHMs to the air. A higher amount of fresh air may be needed to compensate for the increased disturbance in the water surface and increased air- and water temperature.

In S1 and S2, samples collected during morning swimming showed in average 19% and 60% higher values of tTHM compared to corresponding values measured during baby swimming in the afternoon. These values are calculated based on the days were no samples were excluded. This finding can be associated with the use of a night mode in the ventilation system. Night mode ventilation equals approximately 70% of day mode air flow ventilation, and during night less fresh air is supplied.

Measurements carried out on Mondays were found to be higher compared to Wednesdays and Fridays. This is associated with high occupancy during weekends and constant ventilation flowrate, regardless of the number of visitors. This underlines the importance of a flexible and good ventilation strategy.

### CONCLUSION

In this study, concentrations of brominated THMs in the air appear to be much higher when sodium hypochlorite is used for water treatment. Regardless of whether calcium hypochlorite or sodium hypochlorite is used for water treatment, the concentration of tTHMs were measured to be higher in the breathing zone of the swimmers in the pool compared to 1.50 m above the floor. The levels of TBM above the pool with 33% seawater were measured to be much higher (226%) at 0.05 m compared to 1.50 m. Considering the increased toxicity of this compound, it is crucial to measure as close to the water surface as possible, especially when brominated THMs are present. More attention should be payed to TBM in swimming pool air as this compound potentially could cause severe effects to human health. Increased attention should also be payed to the effect of supplied amount of fresh air and air distribution to reduce the concentration of the volatile DBPs.

Pool 1, S1 (s	ports pool)		-			
Height (m)	THM	n	AM ( $\mu g/m^3$ )	SD (μg/m <sup>3</sup> )	Min ( $\mu g/m^3$ )	Max (µg/m <sup>3</sup> )
0.05	tTHM	6	230.9	57.7	162.5	316.4
	TCM	6	224.3	55.0	160.0	306.6
	BDCM	6	6.6	2.8	2.5	9.7
0.60	tTHM	6	139.5	36.9	99.7	199.7
	TCM	6	137.5	36.2	98.4	196.5
	BDCM	6	2.1	0.7	1.4	3.2
1.50	tTHM	6	134.9	28.1	99.4	179.4
	TCM	6	133.2	28.5	97.9	179.4
	BDCM	6	2.2	0.8	1.4	3.4
Pool 2, S1 (tl	nerapy pool)		•			
Height (m)	THM	n	AM ( $\mu g/m^3$ )	SD ( $\mu g/m^3$ )	Min ( $\mu g/m^3$ )	Max (µg/m <sup>3</sup> )
0.05	tTHM	8	141.9	39.5	95.9	202.7
	TCM	8	139.4	38.6	94.8	198.8
	BDCM	8	2.4	1.2	1.1	3.9
0.60	tTHM	7	129.3	27.7	96.7	173.2
	TCM	7	127.1	27.3	95.1	171.2
	BDCM	7	2.2	1.9	0.6	6.2
1.50	tTHM	8	129.1	39.5	89.2	170.4
	TCM	8	127.2	29.6	89.2	168.4
	BDCM	8	1.9	1.4	< LOQ	4.3
Pool 3, S2 (s					<u> </u>	
Height (m)	THM	n	AM ( $\mu g/m^3$ )	SD ( $\mu g/m^3$ )	Min (µg/m <sup>3</sup> )	Max (µg/m <sup>3</sup> )
0.05	tTHM	5	606.0	128.5	448.3	781.7
0.00	TCM	5	268.4	96.5	158.8	415.7
	BDCM	5	55.3	12.7	38.4	70.4
	DBCM	5	30.4	6.1	24.4	38.4
	TBM	5	251.9	54.8	174.0	318.9
0.60	tTHM	5	492.5	114.2	366.5	676.1
0.00	TCM	5	290.7	108.4	201.7	476.2
	BDCM	5	58.1	14.8	42.0	77.4
	DBCM	5	19.9	3.6	15.3	24.4
	TBM	5	123.8	25.8	101.0	165.2
1.50	tTHM	5	485.9	112.5	372.4	657.4
1.50	TCM	5	296.1	103.2	205.8	468.6
	BDCM	5	59.5	103.2	46.5	75.8
	DBCM	5	18.9	3.2	15.5	22.5
	TBM	5	111.3	23.3	92.5	318.9
Pool 4, S2 (tl		5	111.5	23.3	92.3	510.9
Height (m)	THM	n	<b>AM (μg/m<sup>3</sup>)</b>	SD ( $\mu g/m^3$ )	Min (µg/m <sup>3</sup> )	Max (µg/m <sup>3</sup> )
0.05	tTHM	8	397.3	145.5	210.6	638.4
0.05	TCM	8 8	296.7	143.3	153.5	476.6
	BDCM	8 8	58.9	21.6	31.4	94.1
	DBCM	8 8	10.6	4.1	4.6	16.8
	TBM	8 8	31.2	4.1	21.1	51.1
0.60	1	_	338.4	107.4		508.8
	tTHM TCM	10		86.0	205.0	308.8 400.0
		10	251.5		140.7 32.8	
	BDCM	10	47.5	11.8		69.3
	DBCM	10	8.7	3.0	4.9	12.9
1.50	TBM	10	30.6	10.7	17.9	50.4
1.50	tTHM	8	342.9	109.8	210.9	492.9
	TCM	8	258.4	86.9	152.9	372.6
	BDCM	8	46.0	13.5	28.0	68.8
	DBCM	8	8.5	3.4	4.0	16.8
	TBM	8	31.2	11.3	21.1	49.9

Table 3: THM concentrations in  $\mu g/m^3$  for the different swimming pools and different heights

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