

DETERMINANTS OF EXPOSURE AND VARIABILITY OF TRIHALOMETHANES IN THE AIR OF THREE INDOOR SWIMMING POOLS

Therese Bergh Nitter¹, Kristin v Hirsch Svendsen²

¹Institute for Civil and Environmental Engineering, Centre for Sport Facilities and Technology (SIAT),
Norwegian University of Science and Technology, Trondheim Norway

²Department of Industrial Economics and Technology Management, Norwegian University of Science and
Technology, Trondheim Norway

Correspondence to:

Therese Bergh Nitter,

Høgskoleringen 7A,

7491 Trondheim, Norway

E-mail: therese.nitter@ntnu.no

Telephone: 0047 417 28 412

Abstract

Introduction: Negative health effects related to long-term exposure to volatile trihalomethanes (THMs) formed during the chlorination of pool water is recognized, but the determinants causing the concentrations to vary within and between sampling locations have not received much attention.

Methods: 120 air samples of four THMs were examined in three Norwegian indoor pool facilities. In each facility, repeated samples were collected above a sports pool and a therapy pool, 0.05 m and 0.60 m above the water's surface. A linear mixed model (LMM) was used to identify determinates of exposure and the variability in THM concentration within and between sampling locations, days, and heights in pool facilities.

Results: The within variability of days, sampling locations, and heights was greater than between days, sampling locations, and heights. Determinates contributing significantly to the exposure were pool facility, height, swimming pool, day of the week, time during the day, and number of bathers. These findings limits how exposure categories should be defined to be able to identify the real long-term exposure and to propose suitable exposure limit values.

Discussion: These determinants could help future research be designed with effective sampling strategies and to collect information about the real long-term exposure, which is important in terms of establishing a dose-response relationship and exposure limit values.

Conclusions: If unbiased exposure assessments are to be conducted amongst the different users of the pool facility, air samples should be collected over time and for different exposure scenarios.

Introduction

To prevent the growth of hazardous microorganisms in swimming pool waters, it is common to disinfect the pool water using chlorine. However, when chlorine reacts with organic and inorganic materials in the pool water, unwanted disinfection by-products (DBPs) are formed (World Health Organization, 2006). An important group of DBPs is the trihalomethanes (THM), represented by the four components chloroform (CHCl_3), bromoform (CHBr_3), bromodichloromethane (CHBrCl_2), and dibromochloromethane (CHBr_2Cl). The sum of the four is referred to as total THM (tTHM). The tTHM is volatile and can penetrate the skin easily, making both inhalation and dermal absorption important pathways of exposure (Erdinger et al., 2004; Chowdhury, 2015). Long-term health effects, such as adverse reproductive outcomes, cancer, and stillbirth, have been associated with exposure, but the evidence is somewhat inconsistent (World Health Organization, 2017; Rivera-Núñez et al., 2018). In some studies, in which multi-pathway exposures have been estimated, the cancer risk of exposure to THMs in indoor swimming pool facilities is found to be unacceptably high compared to the upper limit of the acceptable cancer risk proposed by the European Commission (10^{-5}) (European Commission, 2009; Lee et al., 2009; Chen et al., 2011).

Some European countries have established exposure limit values for tTHM in swimming pool water, ranging from $20 \mu\text{g/l}$ to $100 \mu\text{g/l}$ (ANSES, 2010; Ohlsson et al., 2014; Rijksinstituut voor Volksgezondheid en Milieu, 2014). In Norway, no such limit for pool water exists. Although inhalation is recognized as the most important exposure pathway (Erdinger et al., 2004; Aprea et al., 2010), only one suggested limit value, proposed by the German Federal Environmental Agency (in VDI 2089) for CHCl_3 in indoor pool facilities exists ($200 \mu\text{g/m}^3$) (Verein Deutscher Ingenieure, 2010). It is important to understand the cause and magnitude of the variabilities in the contaminants in order to comprehend the dose-response relationship used in designing a sampling strategy or evaluating compliance with limit values and control measures (Burdorf, 2005). In pool facilities, the users are exposed to a mixture of components in the air and water (Catto et al., 2012; Chowdhury, 2015; Tardif et al., 2016), and the exposure concentrations of tTHM are relatively low. Studying the effect of chronic exposure to low-exposure concentrations is complicated (Goldberg and Hemon, 1993), especially since researchers often have limited time and resources available to conduct sufficient sampling for exposure characterization.

Identifying the determinants of exposure is essential in the ultimate control of exposure but also for a valid and precise assignment of exposure levels (Rappaport and Kupper, 2008).

Although the occurrence and exposure to DBPs in swimming pools have been investigated in several previous studies, the determinants causing the concentration to vary within and between sampling locations have not received attention and no systematic sampling strategy to collect representative air samples has so far been proposed. The aim of this study is to determine the size and magnitude of the variability and to analyse which determinants affect the exposure within three pool facilities in order to be able to optimize a sampling strategy for DBPs.

Methods

The pool facilities

Three swimming pool facilities, each with several pools, located in the middle part of Norway were included in this study. The physical parameters and types of water and disinfectants used in these pools are shown in Table 1. In facilities 1 and 3, the swimming pools are located in the same room and ventilated using one ventilation system. In pool facility 2, the therapy pool and sports pool are located in two separate rooms, and these rooms have two different systems for both water circulation and ventilation.

Table 1: Physical parameters, disinfectants, freshwater/seawater ratio, and number of bathers for the three pool facilities

Facility	Pool	Size (m)	T _{water} (°C) ¹	Disinfectants	Bathers/year
1	Sports pool	25.0 x 12.5	27	Ca(OCl) ₂	120,000
	Therapy pool	12.5 x 6.5	33	Ca(OCl) ₂ + UV	
2	Sports pool	50.0 x 21.0	28	NaOCl+ UV	360,000
	Therapy pool	16.7 x 9.5	34	NaOCl+ UV	
3	Sports pool	25.0 x 12.5	28	Electrolysis ² + UV	100,000
	Therapy pool	12.5 x 9.0	34	Electrolysis ² + UV	

¹T_{water}= Water temperature, ²Sodium hypochlorite (NaOCl) produced on-site using electrolysis

In all, 120 stationary air samples were collected. In each pool facility, samples were collected above one therapy pool (location 1) and one sports pool (location 2), both 0.05 m and 0.60 m above the water's surface, during the morning and afternoon. Auxiliary data, such as bather load, water temperature, pH-value, and free and combined chlorine were also recorded on each day of sampling. In one of the pool facilities (facility 3), 61 air samples were collected on six days over a period of six weeks. In facilities 1 and 2, 29 and 30 samples were collected, respectively, on ten different days over a period of four weeks.

Sampling and analytical method

The sampling, quality assurance, and analytical methods are based on Method TO-17, published by the United States Environmental Protection Agency (US EPA) (United States Environmental Protection Agency, 1999). Samples were collected using the method of active air sampling, in which two low-flow pumps (Markes Int.) were calibrated to deliver between 40 ml/min and 50 ml/min for 20 minutes to collect ambient air onto automatic thermal desorption tubes containing 200 mg Tenax TA (Markes Int.). Analyses were performed using a Unity Thermal Desorber (Markes Int.) coupled with an Agilent Technologies 5975T Low Thermal Mass Gas Chromatography/Mass Selective Detector. The analysis setup is explained elsewhere (Nitter et al., 2017).

Statistical analysis

Statistical analyses were performed using the Statistical Package for the Social Sciences (IBM SPSS) 25. To verify that the three facilities and the sampling locations were statistically different from each other, the Bonferroni post hoc test and t-test were used. The linear mixed-effect model (LMM) was used to examine the relationship between the concentrations of tTHM and to identify the determinants of importance for the variability in measured concentrations. Variation in exposure levels can be large, and the probability distribution is often best described using a log transformed distribution. (Boleij et al., 1995) Before the analyses were carried out, the fit of the residuals of the log transformed exposure data was evaluated.

The LMM was fitted as follows:

$$B_{ij} = \beta_{0i} \times r_{0i} + \beta_{1i} \times F + \beta_{2i} \times h + \beta_{3i} \times P + \beta_{4i} \times \text{Day} + \beta_{5i} \times t + \beta_{6i} \times \text{Bathers} + e_{ij}$$

Where,

- B_{ij} = natural log-transformed value of the tTHM concentration i for observation j ;
- h = height above water surface in meters;
- P = type of swimming pool;
- Day = day of the week;
- t = time during the day;
- Bathers = number of bathers in the pool;
- β_{0i} is the fixed intercept; r_{0i} is the random intercept, and e_{ij} is the random error.

All parameters were estimated using the method of restricted maximum likelihood (REML) (Symanski et al., 2001; West et al., 2006).

Analysing the variability and determinants of exposure

The dataset consisted of categorical variables, such as pool facility, swimming pool, and height above the water's surface, time during the day, day of the week, and the number of bathers, and continuous variables, such as free and combined chlorine, air temperature, relative humidity, pH value, and the log-transformed tTHM. First, the within and between subject variability was estimated using REML and without including additional factors and covariates. For this analysis, each pool facility was analysed separately, and sampling location, day of the week and height above the water's surface were treated as the subject. The subject was included as random combination variable using the default variance components as a covariance structure. Models with different combinations of fixed effects were constructed by forward selection and the subsequent likelihood ratio test using maximum likelihood estimation. The final model was estimated by REML. Sampling location was the subject of the analysis and was included as random variable using the default variance components as a covariance structure.

Results

The characteristics for the three different pool facilities with sampling location, tTHM, bathers, free and combined chlorine, water temperature, pH-value, and number of samples are given in Table 2. All water quality parameters were in accordance with Norwegian regulations (Norwegian Ministry of Health, 1996).

Table 2: Mean descriptive statistics for chemical parameters, bathers, and number of samples from each sampling location

Facility	Sampling location	tTHM (SD) ($\mu\text{g}/\text{m}^3$)	Bathers	Cl _{Comb} (mg/l)	Cl _{Free} (mg/l)	T _{water} ($^{\circ}\text{C}$)	pH	N
1	1 ^A	132.6 (33.3)	8.2	0.21	1.22	33.1	7.5	17
	2 ^B	185.2 (66.4)	13.3	0.21	0.82	26.7	7.3	12
2	1	362.7 (118.9)	31.4	0.19	1.02	34.1	7.2	20
	2	549.2 (129.3)	16.1	0.04	0.49	28.6	7.3	10
3	1	179.9 (54.1))	7.9	0.24	1.14	33.6	7.4	37
	2	234.1 (80.5)	7.7	0.24	1.01	28.1	7.3	24

Abbreviations: ^ATherapy pool; ^BSports pool; SD= Standard deviation; N= Number of repetitions

As shown in Table 2, the highest level of tTHM was observed above the sports pools in each pool facility, and the highest concentrations were observed in pool facility 2. The THM levels for all pool facilities and sampling locations were statistically significantly different from each other ($p < 0.05$).

Variability within each facility

In Table 3, the estimates of the within and between variability for sampling locations, days, and heights for each of the three pool facilities are given.

Table 3: Within and between variability of the logarithm of the exposure concentration at different sampling locations, days, and heights for each pool facility

Pool Facility 1	N	μ (SD)	σ_b^2 (SD)	σ_w^2 (SD)
Sampling locations	29	4.99 (0.32)	0.04 (0.06)	0.09 (0.02)
Day			0.04 (0.04)	0.08 (0.02)
Height			0.03 (0.04)	0.09 (0.02)
Pool Facility 2	N	μ (SD)	σ_b^2 (SD)	σ_w^2 (SD)
Sampling locations	30	6.06 (0.22)	0.09 (0.14)	0.09 (0.03)
Day			0.03 (0.03)	0.11 (0.03)
Height			0.01 (0.02)	0.13 (0.03)
Pool Facility 3	N	μ (SD)	σ_b^2 (SD)	σ_w^2 (SD)
Sampling locations	61	5.25 (0.34)	0.03 (0.04)	0.11 (0.02)
Day			0.03 (0.02)	0.09 (0.02)
Height			0.03 (0.05)	0.10 (0.02)

N= number of samples; μ =mean concentration of the lognormal tTHM; SD= Standard deviation; σ_b^2 = between variability (sampling location, day, height); σ_w^2 = within variability (sampling location, day, height)

As shown in Table 3, the within sampling location variability was greater than or equal to the between sampling location variability. The variability between days was less than that within days, and the variability within heights was greater than that between heights, reflecting the importance of the time of the day for sampling.

Determinants of exposure

The determinants shown to affect significantly the exposure contamination level of the different sampling locations are shown in Table 4. The fixed factors height, pool, day, time, and bathers explain 42% of the total variability for each sampling location, of which these fixed factors reduced the within sampling location variability by 36% and the between sampling location variability by 98%.

Table 4: Significant determinants of exposure

Fixed Effect	β	SE	e^{β}
Df	13		
Intercept	4.84*	0.17	126.47
Facility			
1	-0.18*	0.09	0.84
2	0.91*	0.07	2.48
3	0	0	1
Height			
0.05 m	0.22*	0.05	1.25
0.60 m	0	0	1
Pool			
Therapy pool	-0.31*	0.06	0.73
Sports pool	0	0	1
Day			
Monday	0.24*	0.09	1.27
Wednesday	0.04	0.11	1.04
Friday	0	0	1
Time			
Morning	0.11*	0.05	1.12
Afternoon	0	0	1
Bathers			
0 - 6	0.25*	0.13	1.28
7 - 16	0.26*	0.13	1.30
17 - 34	0.35*	0.12	1.42
35 - 50	0	0	1
Random effects for sampling locations	Variance		
σ_w^2	0.062		
σ_b^2	0.001		
-2LL	39.5		

Df= degrees of freedom; β = regression coefficient of the different determinants; SE= Standard error; e^{β} = determinants for estimating the geometric mean (GM) of different exposure scenarios

*p < 0.10

The geometric mean (GM) (e^{β}) for different exposure scenarios can be estimated using the values obtained in Table 4. To determine the worst case (Monday morning) and best case (Friday afternoon) for exposure amongst a team of 15 swimmers training in the sports pool in the morning in pool facility 2, the following calculation can be done:

$$GM = \text{Intercept} * \text{Facility} * \text{Height} * \text{Pool} * \text{Day} * \text{Time} * \text{Bathers} \quad (2)$$

$$\text{Worst case}_{\text{Facility 2}} GM = (126.47 \times 2.48 \times 1.25 \times 1.0 \times 1.27 \times 1.12 \times 1.30) = 725.0 \mu\text{g}/\text{m}^3$$

$$\text{Best case}_{\text{Facility 2}} GM = (126.47 \times 2.48 \times 1.25 \times 1.0 \times 1.27 \times 1.12 \times 1.30) = 530.1 \mu\text{g}/\text{m}^3$$

Swimmers training Monday morning are, on average, exposed to 37% higher concentrations compared to swimmers training on Friday afternoon. Using the same scenario to calculate the worst case in facility 1, the mean exposure is estimated to $292.3 \mu\text{g}/\text{m}^3$, meaning that the swimmers in facility 2 are exposed to 2.48 times higher concentrations compared to the swimmers in facility 1.

Discussion

In our study, the within days and within heights variability was greater than the between days and between heights variability in all pool facilities. The within and between sampling location variabilities obtained from facility 2 were equal (0.09), while, in the other two facilities, the within swimming pool (sampling locations) variabilities were 2.3 (facility 1) and 3.7 (facility 3) times greater compared to the between sampling location variabilities. The equality in the variabilities within and between pools for facility 2 is probably explained by the swimming pools being located in two different rooms and being ventilated by two different ventilation systems. Greater variability within pools than between pools was also found in the water of eight swimming pools in London (Chu and Nieuwenhuijsen, 2002). In a cross-sectional study, in which air and water samples were collected once from 41 pool facilities, the results showed great variability between the different swimming pools (Tardif et al., 2016). However, in this study the variabilities between days and times of day were not considered.

For components such as THMs, the long-term average exposure is relevant, as the health effects are caused by long-term chronic exposure (Boleij et al., 1995). To understand the long-term average exposure, our findings highlight the importance of collecting repeated samples during different scenarios over time. Samples should also be collected from each room in the facility or from each zone in which different ventilation systems are used.

In facility 2, the greatest measured concentration during a 20 min sampling time within the same day ranged from $361.7 \mu\text{g}/\text{m}^3$ to $781.7 \mu\text{g}/\text{m}^3$, and the mean day-to-day concentration in this facility ranged from $341.7 \mu\text{g}/\text{m}^3$ to $590.9 \mu\text{g}/\text{m}^3$. This highlights that one single sample or one-day sample is not representative of the exposure of a user who spend only a few hours

in the pool. Using a cross-sectional study design to collect information about the air quality and exposure, which was done in several previous studies (Chen et al., 2011; Löfstedt et al., 2016; Tardif et al., 2016), is not considered suitable for collecting representative air samples in pool facilities. Rather, repeated measures over time are important in terms of understanding the total variability and the change in exposure over time.

Our study shows that the swimming facility, height above the water's surface, swimming pool, day of the week, time during the day, and number of bathers contribute to the mean exposure level. These fixed factors also reduced the magnitude of the between sampling location variability within the facilities and suggest that some of the heterogeneity observed in each facility was due to these determinants. From the literature, the level of THM is reported to increase rapidly within the first few hours after the precursors are introduced to the chlorinated water (Urano et al., 1983), and, in a previous study, it was suggested that an equilibrium period of 72 hours is the optimal time for THM formation in water (D Eichelsdörfer and Jandik, 1983). As shown in Table 4, the concentration of THM was consequently higher on Mondays compared to Wednesdays and Fridays. We also measured higher concentrations in the morning compared to in the afternoon. These findings are in accordance with a previous study from the USA, in which a weekly pattern of DBPs in the air was observed and the concentrations of DBPs were typically high on Mondays, then decreased gradually through the following Friday (Afifi and Blatchley Iii, 2015).

Although increased water temperatures are related to increased formation of THMs (Padhi et al., 2012), there are several reasons why higher concentrations were measured above the sports pools compared to above the therapy pools, despite the fact that the therapy pools had higher bather loads as well as higher water temperatures. In Norway, it is a requirement that 60 l of fresh water per bather be supplied in therapy pools, while, for pools with lower water temperatures, the requirement for fresh water is only 30 l/bather (Norwegian Ministry of Health, 1996; Norwegian Institute for Water Research, 2000). In addition, it might be reasonable to assume that the swimmers in the sports pool have a higher activity level and release more precursors into the water.

As shown in Tables 2 and 4, the choice of pool facility significantly effects the exposure level. In a recent study, we documented that chlorination method and type of water significantly effects the types and formation of THMs (Nitter et al., 2017). This is explained partly by contaminates of bromide in the brine solution used to produce NaOCl (World

Health Organization, 2017) since previous studies have documented that the presence of bromine has been found to increase the formation of THMs (Amy et al., 1987).

A swimming club training Monday morning in the sports pool will be exposed to higher concentrations compared to swimmers training Friday afternoon. However, if these swimmers are categorized in the same exposure category during an epidemiological investigation, doing so might bias the estimated risk of exposure towards association considering that the afternoon swimmers are less exposed compared to swimmers swimming in the morning.

Taking into account the determinants of exposure identified in Table 3, a more dynamic strategy for both the water exchange and the ventilation system is necessary. As of today, the fresh air and water supply are approximately constant regardless of the day of the week, time of day, or number of bathers in the pool. A balanced system, accounting for these determinants, may more appropriate considering the great within day, within height, and within sampling location variability. Considering that the average exposure level above the sports pool, which is 32% higher than that above the therapy pool, despite the lower water temperatures, it would make sense to increase the fresh water supply per bather for waters with lower temperatures.

7. Conclusions

The determinants of exposure represented in Table 4 indicate which factors are important to consider in effective sampling strategies, as these determinates explained most of the heterogeneity within each pool facility. Our findings highlight the need for a more dynamic water and air circulation system, one which is able to identify the variations within each sampling location in terms of times, days, and bather loads.

Considering that the exposure concentration is very time-dependent, varying with days of the week and times during the day, this time dependency should be considered before exposure categories amongst the different users of the pool facilitates in epidemiological investigations are created. Putting every swimmer in the same exposure category might prevent the investigator from understanding the dose-response relationship.

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