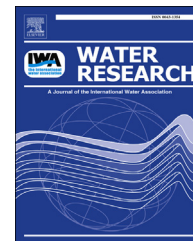


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Seasonal dynamics of water and air chemistry in an indoor chlorinated swimming pool

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ABSTRACT

Although swimming is known to be beneficial in terms of cardiovascular health, as well as for some forms of rehabilitation, swimming is also known to present risks to human health, largely in the form of exposure to microbial pathogens and disinfection byproducts (DBPs). Relatively little information is available in the literature to characterize the seasonal dynamics of air and water chemistry in indoor chlorinated swimming pools. To address this issue, water samples were collected five days per week from an indoor chlorinated swimming pool facility at a high school during the academic year and once per week during summer over a fourteen-month period. The samples were analyzed for free and combined chlorine, urea, volatile DBPs, pH, temperature and total alkalinity. Membrane Introduction Mass Spectrometry (MIMS) was used to identify and measure the concentrations of eleven aqueous-phase volatile DBPs. Variability in the concentrations of these DBPs was observed. Factors that influenced variability included bather loading and mixing by swimmers. These compounds have the ability to adversely affect water and air quality and human health. A large fraction of the existing literature regarding swimming pool air quality has focused on trichloramine (NCl_3). For this work, gas-phase NCl_3 was analyzed by an air sparging-DPD/KI method. The results showed that gas-phase NCl_3 concentration is influenced by bather loading and liquid-phase NCl_3 concentration. Urea is the dominant organic-N compound in human urine and sweat, and is known to be an important precursor for producing NCl_3 in swimming pools. Results of daily measurements of urea indicated a link between bather load and urea concentration in the pool.

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1. Introduction

Swimming in pools is a common recreational activity worldwide and is believed to provide health and fitness benefits. Chlorination is the predominant disinfection method applied in swimming pools due to its cost-effectiveness, ease of application, and ability to inactivate a wide variety of pathogenic microorganisms. Chlorination also promotes oxidation

of some dissolved organic compounds (AWWA, 1999). However, it is also known to produce disinfection by-products (DBPs), mostly via reactions between free chlorine (defined as the sum of the concentrations HOCl , OCl^- , and Cl_2) and natural organic matter in source water, as well as inputs from bathers through sweat, urine, hair, skin particles, and personal care products (Kristensen et al., 2009; Judd and Black, 2000). Both sweat and urine contain important precursors to the formation of volatile DBPs that are introduced to

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swimming pools by humans (Weaver et al., 2009; Li and Blatchley, 2007). Urine introduction to pools has been reported to be 25–77.5 mL of per bather (Gunkel and Jessen, 1986; Erdinger et al., 1997; Weng et al., 2011). The amount of sweat per bather is highly variable and depends on the individual level of physical activity and hygiene habits prior to entering the pool.

In the United States, the recommended free available chlorine concentration range in pools is 1–5 mg/L as Cl_2 , with an ideal range between 2 and 4 mg/L as Cl_2 (NSPF, 2010). The preferred pH range in swimming pools is 7.4–7.6 (NSPF, 2010). However, in European countries the recommended free chlorine range in the swimming pools is often lower. For example, in Germany the concentration of free chlorine must be kept in the range of 0.3–0.6 mg/L in pool water at a pH range between 6.5 and 7.6 (Zwiener et al., 2007).

Most studies regarding health effects of DBPs have focused on drinking water exposure; however, in some respects swimming may present a greater risk of exposure to DBPs as uptake may occur through three different exposure paths: ingestion, inhalation, and dermal absorption. Moreover, the concentrations of DBPs in pool water often exceed those of drinking water.

For some DBPs (e.g., CHCl_3 and halophenols), the dominant pathway for uptake during swimming is dermal absorption due to contact of large surface area of skin with water (Lindstrom et al., 1997; Weisel and Shepard, 1994; Xiao et al., 2012). For other compounds (e.g., NCl_3), inhalation has been identified as an important route of exposure (Zwiener et al., 2007). These pathways for human exposure are potentially problematic due to the toxic nature of some DBPs. Dermal uptake is particularly important because it provides direct access to the bloodstream, without going through the digestive system. Therefore, mutagenicity, genotoxicity, and cytotoxicity responses to swimming pool water can be greater than with tap water sources (Richardson et al., 2012).

More than 600 DBPs have been identified in chlorinated waters, and many of them are mutagenic or carcinogenic (Richardson et al., 2012). To date, the DBPs identified in swimming pools include trihalomethanes (THMs), iodo-THMs, haloacetates, haloacetic acids, haloacids, halodiacids, haloaldehydes, halonitriles, halo ketones, halopyrroles, halo-methanes, halonitromethanes, bromate, haloamides, haloalcohols, nitrosamines, combined chlorine, halofuranones (Richardson et al., 2012), halophenols (Xiao et al., 2012) and halobenzoquinones (HBQs) (Wang et al., 2013). In indoor chlorinated swimming pool facilities, air and water qualities are both relevant issues with respect to human health. Li and Blatchley (2007) identified eleven volatile DBPs that are formed in chlorinated swimming pools including: three inorganic chloramines, four THMs, cyanogen chloride, cyanogen bromide, dichloroacetonitrile, and dichloromethylamine. Acute and chronic human health problems that have been associated with exposure to these DBPs in swimming pools include: promotion of asthma (Bernard et al., 2003, 2007); increased incidence of rhinitis and hay fever (Bougault et al., 2010; Bernard et al., 2009); as well as skin (contact dermatitis) and eye irritation (Fantuzzi et al., 2010; Safranek et al., 2007). DBP exposure in swimming pools has also been associated with an increased risk of bladder cancer (Villanueva et al., 2007).

Therefore, it is important to understand the chemistry and physics that govern the formation, transfer, and decay of DBPs in pools; however, their behavior is complicated by competing reactions and transport phenomena, as well as a general lack of research for some of these DBPs.

THM formation in pools is believed to originate from reactions between free chlorine with organic precursors such as humic substances, skin particles, hair, lotions, and cosmetics (Kim et al., 2002). Aggazzotti et al. (1990, 1993, 1995, 1998) performed a series of studies in Italy and found correlations between chloroform concentrations in air and water and the number of swimmers, chloroform concentration in water, free and combined residual chlorine and water pH, but these were generally only weak to moderate correlations. Reported values of the concentration of chloroform have ranged from 33 to 140 $\mu\text{g/L}$. Measurements of the concentration of other THMs in water and in air in swimming pools have also been reported (see SI-1).

In general, chloroform is the dominant THM (by mass) in swimming pools. Chloroform (CHCl_3) is a highly volatile compound that can be inhaled and also readily absorbed via dermal uptake (Lindstrom et al., 1997). The presence of chloroform in the atmosphere of indoor swimming pools has been reported by several authors (WHO, 2006; Faust, 1993; Aggazzotti et al., 1998; Matthiessen and Jentsch, 1999; Lahl et al., 1981; L'evesque et al., 2000) with reported gas-phase concentrations ranging from 1.7 to 1630 $\mu\text{g/m}^3$. Some of the factors that have been reported to influence the chloroform concentration in air in indoor swimming pool facilities include: ventilation rate, bather loading, and free chlorine concentration (Aggazzotti et al., 1998; Chu and Nieuwenhuijsen 2002).

According to the International Agency for Research on Cancer, chloroform has been classified as a 2B (possible carcinogen); therefore, human exposure related to THMs in indoor swimming pools is an issue of concern (Aggazzotti et al., 1998). With respect to chloroform exposure, Lindstrom et al. (1997) estimated that 80% of uptake for swimmers is via dermal absorption. However, Erdinger et al. (2004) performed a study to clarify the uptake pathway of chloroform into the human body. Their data indicated that inhalation was the most important pathway, and that uptake over the skin is significantly less. According to the Erdinger et al. (2004) study, chloroform concentration in blood is correlated to air concentration of chloroform and is not linked directly to chloroform concentrations in water. However, the rate of inhalation depends on the intensity of the exercise (Chu and Nieuwenhuijsen 2002; Aggazzotti et al., 1995).

Dichloroacetonitrile (DCAN) and other halonitriles can be produced from the chlorination of free amino acids, heterocyclic nitrogen in nucleic acids, proteinaceous materials, and combined amino acids bound to humic structures (Lee et al., 2007; Li and Blatchley, 2007). Previous research has indicated that DCAN will undergo hydrolysis in aqueous solution; the rate of DCAN decay is enhanced in the presence of free chlorine and at high pH (Bieber and Trehy, 1983; Reckhow et al., 2001). DCAN toxicity was investigated after McKinney et al. (1976) identified this compound as the unique ingredient in tap water suspected of causing reduced fertility and decreases in litter size in a colony of laboratory rodents (Smith et al.,

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