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Spatial variation of disinfection by-product concentrations: Exposure assessment implications

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ABSTRACT

The use of public water system (PWS) average trihalomethane (THM) and haloacetic acid (HAA) concentrations as surrogates of "personal" exposures in epidemiological studies of disinfection by-products (DBPs) may result in exposure misclassification bias from various sources of measurement error including intra-system variation of DBPs. Using 2000–2004 data from 107 PWSs in Massachusetts, we assessed two approaches for characterizing DBP spatial variability by identifying PWSs with low spatial variability (LSV) and examining differences in LSV across DBP groups and by type of source water and primary disinfectant. We also used spatial differences to examine the association between THM concentrations and indices of social disadvantage; however, we found no correlations or statistically significant differences based on the available data. We observed similar patterns for the percentage of quarterly sampling dates with LSV across different types of source water for all DBPs but not across disinfectants. We found there was little overlap between sites classified as having LSV across different DBP groups. In the main analysis, we found moderate correlations between both approaches ($\varphi_{THM4} = 0.55$; $\varphi_{BrTHM} = 0.64$; $\varphi_{HAA5} = 0.67$); although Method 1 (based on concentration differences between samples) may be better suited for identifying PWSs for inclusion in epidemiological studies because it is more easily adapted to study-specific exposure gradients than Method 2 (based on categorical exposure percentiles). These data reinforce the need to consider different exposure assessment approaches when examining the spatial variation of multiple DBP surrogates as they can represent different DBP mixtures.

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1. Introduction

Disinfection by-products (DBPs) are formed when disinfectants combine with DBP precursors, such as natural organic matter. The formation of DBPs is dependent on many interrelated factors including disinfectant treatment processes (e.g., type, amount, and timing of disinfection dose), distribution system characteristics (e.g., size of system and residence time), and water characteristics (e.g., amount of organic and inorganic precursors, water temperature, and pH) (Hua and Reckhow, 2008; Rodriguez et al., 2004; Rodriguez and Sérodes, 2001). Trihalomethanes (THMs) and haloacetic acids (HAAs) are the most abundant classes of DBPs found in drinking water (Krasner et al., 1989). THM4 (sum of chloroform, dibromochloromethane, bromodichloromethane, and bromoform) and HAA5 (sum of monochloroacetic, dichloroacetic, trichloroacetic, monobromoacetic, and dibromoacetic acids) are measures of DBP mixtures currently regulated by the U.S. Environmental Protection Agency (EPA) and other regulatory agencies (Health Canada, 2004; U.S. EPA, 2006; WHO, 2004).

DBPs have been associated with adverse health outcomes in some epidemiological studies and meta-analyses of bladder cancer (Villanueva et al., 2004, 2007a), colorectal cancer (Rahman et al., 2010), and adverse reproductive outcomes such as fetal growth retardation, spontaneous abortions, stillbirths, and birth defects (Bove et al., 1995; Grazuleviciene et al., 2013; Grellier et al., 2010; Hinckley et al., 2005b; Hoffman et al., 2008; Hwang et al., 2008; Levallois et al., 2012; Lewis et al., 2006; Righi et al., 2012; Savitz et al., 1995; Summerhayes et al., 2012; Toledano et al., 2005; Villanueva et al., 2011; Waller et al., 1998; Wright et al., 2003, 2004). Exposure assessment has been identified as one of the major limitations in epidemiological studies of DBPs and may partially explain inconsistent study results observed for specific health outcomes (Grellier et al., 2010; Nieuwenhuijsen et al., 2000). Sources of exposure measurement error have important ramifications for epidemiological studies since they can lead to misclassification bias (Villanueva et al., 2007b; Waller et al., 2001; Whitaker et al., 2003; Wright and Bateson, 2005).

Distribution system-averages are calculated from spatially and temporally limited routinely-collected monitoring data and used as surrogates of personal exposure to DBP mixtures. In addition to uncaptured temporal and spatial variability in DBP formation, another key source of measurement error associated with aggregate exposure measures includes intra- and inter-individual differences in water intake and water-use activities (Wright et al., 2006). Some studies have also shown water use and water quality to be associated with socio-demographic characteristics (Castaño-Vinyals et al., 2011; Forssén et al., 2009; Hales et al., 2003; Smith et al., 2009; Williams et al., 2001), which is a potential environmental justice concern. Spatial variability data can be used to identify areas with higher exposure contrasts to facilitate the evaluation of the association between socioeconomic status and DBP concentrations.

Temporal variation in DBP concentrations, such as seasonality, has been observed with differing patterns of formation and degradation noted across DBP classes and individual species (Chen and Weisel, 1998; Krasner et al., 1989; Parvez et al., 2011; Rodriguez et al., 2004; Rodriguez and Sérodes, 2001; Stevens et al., 1989; Summerhayes et al., 2011). DBP concentrations can also vary within drinking water distribution systems (Keegan et al., 2001; Lynberg et al., 2001; Williams et al., 1998). This intra-system spatial variation can also differ across DBP groups. For example, THMs may increase in a distribution system, whereas HAAs may decrease over space and time (Chen and Weisel, 1998; Rodriguez et al., 2004; Williams et al., 1998). Therefore, using one DBP group as a surrogate for other groups may lead to additional exposure measurement error in epidemiological studies. This has increasing implications as brominated DBPs have been shown to be more potent toxicants than chlorinated DBPs (Colman et al., 2011; Plewa et al., 2008).

There have been few attempts to quantify intra-system DBP variation for exposure assessment application in epidemiological studies. Waller et al. (2001) examined the effectiveness of two approaches for decreasing potential exposure misclassification associated with using system-average THM concentrations; including (1) restricting their epidemiological study population to a subset of residents from public water systems (PWSs) based on a low spatial variability (LSV) approach (i.e., all samples were within 20 μg/L of each other) and (2) an estimated variance-weighted approach [$\text{weight}_{\text{system-average}} = 1 - (\text{standard deviation}_{\text{system-average}}/$ mean DBP concentration across the sampling database)]. Using the LSV restricted data set; these authors detected an increase in odds ratios that may be attributed to minimizing exposure misclassification. Since these authors noted reduced precision resulting from a decrease in effective sample size, it is important to examine the possible implications associated with the application of any LSV restriction approach. Hinckley et al. (2005a) used data from the Information Collection Rule (U.S. EPA, 2011) to compare two approaches for identifying systems with LSV of THM concentrations – one approach was based on categorical THM concentrations (requiring all samples in the same PWS to fall within the same THM exposure category) and the second approach was based on two-way analysis of variance that accounted for LSV that was dependent on season. The authors found that although the two approaches selected comparable numbers of sites as having LSV, they showed little overlap in the actual sites that were selected.

To date, available approaches for characterizing spatial variability of DBPs (1) have focused on THMs (Hinckley et al., 2005a; Waller et al., 2001), (2) have shown disparate results across approaches (Hinckley et al., 2005a), or (3) have required data that are not always available from regulatory monitoring data in the U.S. (Legay et al., 2010). The main objective of the current analysis was to examine approaches that aim to minimize potential exposure misclassification of routinely-collected THM4, brominated THMs (BrTHM; [sum of dibromochloromethane, bromodichloromethane, and bromoform]), and HAA5 data due to intra-system spatial variability from using aggregated DBP concentration averages. We compared two approaches for defining spatial variability based on THM4, BrTHM, and HAA5 concentrations within