Validation of drinking water disinfection by-product exposure assessment for rural areas in the National Children’s Study

Teresa L. Binkley, Natalie W. Thiex and Bonny L. Specker

The objective of this study was to provide evidence to evaluate the proposed National Children’s Study (NCS) protocol for household water sampling in rural study areas. Day-to-day variability in total trihalomethane (TTHM) concentrations in community water supplies (CWS) in rural areas was determined, and the correlation between TTHM concentrations from household taps and CWS monitoring reports was evaluated. Daily water samples were collected from 7 households serviced by 7 different CWS for 15 days. Coefficients of variation for TTHM concentration over 15 days ranged from 8% to 20% depending on the household. Correlations were tested between TTHM household concentrations and the closest date- and location-matched CWS monitoring reports for the 15-day mean ($R = 0.85$, $P < 0.01$). To simulate the NCS-proposed protocol, correlations were tested for 30 additional NCS household samples (polynomial fit: $R = 0.74$, $P = 0.04$). CWS reported TTHM concentrations $> 50 \mu g/l$ corresponded to measured NCS household concentrations ranging from 2 to 60 $\mu g/l$. TTHM concentrations were higher in CWS than NCS samples (11.2 $\pm$ 3.2 $\mu g/l$, mean difference $\pm$ SE, $P < 0.01$). These results show that in rural areas there is high variability within households and poor correlation at higher concentrations, suggesting that TTHM concentrations from CWS monitoring reports are not an accurate measure of exposure in the household.

INTRODUCTION

Tap water samples from community water supplies (CWS) are routinely tested for trihalomethanes (THMs) to ensure regulatory compliance. THMs are water disinfection by-products (DBPs) resulting from the reaction of chlorine and/or bromine with organic matter present in the water. Total trihalomethanes (TTHMs) include chloroform, bromoform, bromodichloromethane, and dibromochloromethane. Largely because of their availability, TTHMs are the most common surrogate of DBP exposure used by epidemiologists. In the United States, the Comprehensive Disinfectants and Disinfection Byproducts Rules Stage 2 established by the Environmental Protection Agency (EPA) require CWS to comply with established monitoring schedules for testing and standards for concentrations of TTHMs. The EPA limits TTHMs to 80 parts per billion (80 $\mu g/l$) in treated water.

Numerous studies have used CWS TTHM compliance monitoring data as an indirect indicator of exposure to test for associations of TTHM concentrations and birth outcomes, whereas only a few studies have sampled participant home taps or designated sites within the water supply area during the study period. Results of these studies have been mixed, ranging from not supporting an association to strong associations between TTHMs and adverse birth outcomes. Three recent reviews have indicated little or no evidence for associations between TTHM concentrations and adverse birth outcomes with the exception of an increased risk for delivering a small-for-gestational age infant in women with high TTHMs levels in their drinking water.

The National Children’s Study (NCS) plans to examine the effects of genetic and environmental factors on the growth, development, and health of children across the United States. The goal of the NCS is to improve the health and well-being of children and contribute to understanding the role the environment has on health and disease. Environment has been defined to include broad factors such as diet, sound, family dynamics, community, and cultural influences and also more traditional environmental samples from air and water exposures.

The NCS Vanguard Study protocol called for a home visit with the pregnant mother as early as possible during the first trimester of pregnancy. This visit included collection of tap water from a random sample of 10% of study participant’s households served by CWS to test for TTHMs. Existing data from CWS monitoring programs have been suggested to complement household tap water sampling as an indicator of exposure. Requirements established by EPA for rural CWS can vary depending on the source of the water supply (surface or groundwater) and the population of the community served. For example, in South Dakota, CWS monitoring can range from eight samples each quarter to two samples every 3 years depending on population size of the community served and historical contaminant concentrations in the water supply. The known seasonal variability in TTHM levels and broad range in EPA sampling requirements may further complicate the use of CWS monitoring data to assess exposure to DBPs in rural areas. For example, the existing data from CWS monitoring programs could be from a sample that was taken...
more than 1 year before or after the NCS home visit. Therefore, the protocol proposed by the NCS, in particular for rural areas, may be insufficient for capturing exposure levels during critical windows of fetal and child growth and development, and for understanding key confounding variables.

The objective of this study was to provide evidence to indicate whether the NCS-proposed water collection protocol is appropriate for use in rural areas such as South Dakota. The goal of the study was twofold. The first goal was to determine day-to-day variability of TTHM concentrations within a rural household served by a CWS. Variability was calculated over a 15-day period for seven households serviced by seven different CWS. These CWS served populations that varied in size and therefore had different monitoring requirements ranging from weekly to two samples every 3 years. The 15-day mean for each household was then linked with its respective CWS monitoring report matching for the closest date and location to test for correlations. Our second goal was to assess the correlation between TTHM concentration levels of 30 NCS samples collected over a 1-year pilot study period and CWS monitoring data matched for the closest sample date and location. These NCS samples were collected from households serviced by CWS in rural areas. The NCS-proposed protocol assumes that the home sample and the CWS data will be highly correlated and that CWS monitoring data will indicate exposure to TTHMs in lieu of testing all home taps. Our hypothesis was that low day-to-day variability and high correlations between household tap samples and CWS TTHM monitoring levels will support the use of CWS-submitted data as an indicator of exposure in rural areas for large epidemiological studies such as the NCS.

MATERIALS AND METHODS

Two related studies were conducted to achieve the goal of this project.

Study 1: Day-to-Day TTHM Variability and Relationships Between Household Measurements and CWS Monitoring Program Data

In order to calculate day-to-day variability, daily water samples were collected each morning from 7 households over a 15-day period from 24 September to 8 October 2010 to test for TTHM concentrations (Table 1). Water was tested from six eastern South Dakota and one western Minnesota community water suppliers. All of the eastern South Dakota suppliers obtained water from the Big Sioux Aquifer, with shallow wells located less than 50 feet below the surface, whereas the Minnesota community water supply obtained water from a well with a depth of ~250 feet. The water suppliers that we contacted stated that water leaving the plant has a concentration of chlorine that remains constant and there are no “treatment cycles”. In general, when water is in the distribution system for longer or when homes are further from the CWS release location there will be less chlorine and more DBPs present at the home tap. The 15-day study period included 2 weeks of water usage and is a suitable time period to reflect the variability in the TTHM levels in a particular household because of these factors.

The household samples were collected by individuals who were NCS trained and certified for tap water sample collection. Samples were collected according to NCS tap water collection protocol. In brief, the cold water kitchen tap was allowed to run for at least 3 min as described for collecting according to NCS tap water collection protocol. In brief, the cold water kitchen tap was allowed to run for at least 3 min as described for training and certified for tap water sample collection. Samples were collected from households serviced by CWS in rural areas. The NCS-proposed protocol assumes that the home sample and the CWS data will be highly correlated and that CWS monitoring data will indicate exposure to TTHMs in lieu of testing all home taps. Our hypothesis was that low day-to-day variability and high correlations between household tap samples and CWS TTHM monitoring levels will support the use of CWS-submitted data as an indicator of exposure in rural areas for large epidemiological studies such as the NCS.

Table 1. Sampling schema for each study.

<table>
<thead>
<tr>
<th>Study</th>
<th>Sample dates</th>
<th>Samples/ household</th>
<th>No. of households</th>
<th>Total samples</th>
<th>No. of CWS</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>24/9/2010–8/10/2010</td>
<td>15</td>
<td>7</td>
<td>105</td>
<td>7</td>
</tr>
</tbody>
</table>

RESULTS

Study 1: Day-to-Day Variability and Relationships Between Household and CWS Monitoring Program Concentrations

The daily TTHM concentrations by collection site are shown in Figure 1. Spikes in TTHM concentrations can be observed over the 15-day period. It should be noted that significant flooding occurred in the eastern South Dakota sampling areas on 23 September 2010 (day 0). Dates to collect water for this period were established well before this event rather than in conjunction with this event. TTHM concentrations that were above the EPA-recommended level (80 µg/l) were reported for one of the CWS (location 6) during this time period.

Day-to-day variability and mean TTHM concentrations for each location are shown in Table 2. The day-to-day coefficients of
variation (CVs) over the 15-day period ranged from 8% to 20% depending on the household (14.8 ± 5.4%, mean ± SD). There were significant differences among mean TTHM concentrations sampled from different locations. Location 6 had higher TTHM concentrations than any of the other sites. Location 5, with water supplied from a deep well source in western Minnesota, had the lowest TTHM concentrations. Household mean TTHM concentrations correlated with TTHM concentrations reported by the water companies (Figure 2; \( R = 0.85, P < 0.01 \)) and matched pairs analysis determined that the CWS TTHM concentration was not statistically different than the household 15-day mean TTHM concentration (−0.68 ± 13.0 \( \mu g/L \), mean difference ± SD, \( P = 0.22 \)). Even though these samples were matched for closest date, the range of days between the CWS and household sample was from 3 to 487 days, with 3 of the CWS report samples taken more than 1 year from the household sample dates.

Study 2: Relationship Between NCS Household TTHM Concentrations and CWS Monitoring Program Concentrations

Water samples from 30 households served by 8 different CWS were collected to test TTHM concentrations during NCS study visits that occurred between August 2009 and August 2010 (Table 1). Three of the water supply companies were located in eastern South Dakota (\( N = 14 \) samples) and five were located in southwestern Minnesota (\( N = 16 \) samples). Of the samples from South Dakota, nine were from the same CWS, four were from a second, and one was from a third CWS. Of the samples from Minnesota, 12 were from 1 CWS, whereas the other 4 samples were from 4 different suppliers. NCS sample TTHM concentrations correlated with concentrations reported by the water companies as a second-order polynomial (Figure 3; \( R = 0.74, P = 0.04 \)). There was a wide range of household TTHM concentrations for a given CWS concentration. In particular, for CWS TTHM concentrations > 50 \( \mu g/L \), the measured household TTHM concentrations ranged from ~2 to 60 \( \mu g/L \). At CWS TTHM levels < 5 \( \mu g/L \), there was no association between the measurement in the home and the CWS report (\( R = 0.05, P = 0.82 \)). At CWS levels > 5 \( \mu g/L \) TTHM levels, the relationship remained a second-order polynomial (\( R = 0.69, P = 0.003 \)), indicating a non-linear relationship. Separating the fit for lower or higher levels did not change the original results of a non-linear fit. Results from water samples tested by the NCS showed that >50% of the NCS concentrations were lower than what the CWS reported, indicated by points falling below the unity line (Figure 3). Matched pairs analysis confirmed that the CWS TTHM concentration was higher than the NCS TTHM concentration (11.2 ± 3.2 \( \mu g/L \), mean difference ± SE, \( P < 0.01 \)).

Table 2. Coefficient of variation (%) and mean TTHM (\( \mu g/L \)) over the 15-day testing period.

<table>
<thead>
<tr>
<th>CWS</th>
<th>Day-to-day variability CV (%)</th>
<th>Mean TTHM (( \mu g/L ))*</th>
</tr>
</thead>
<tbody>
<tr>
<td>6</td>
<td>18</td>
<td>87.5 ± 17.6( ^a )</td>
</tr>
<tr>
<td>2</td>
<td>12</td>
<td>41.3 ± 6.5( ^b )</td>
</tr>
<tr>
<td>1</td>
<td>16</td>
<td>41.0 ± 4.9( ^a )</td>
</tr>
<tr>
<td>4</td>
<td>20</td>
<td>31.2 ± 6.2( ^a )</td>
</tr>
<tr>
<td>3</td>
<td>9</td>
<td>24.9 ± 2.2( ^b )</td>
</tr>
<tr>
<td>7</td>
<td>20</td>
<td>14.6 ± 2.9( ^b )</td>
</tr>
<tr>
<td>5</td>
<td>8</td>
<td>0.7 ± 0.1( ^a )</td>
</tr>
</tbody>
</table>

*Data are mean ± SD. Means with similar letters are not different by Tukey’s HSD post hoc analysis of means (\( P < 0.05 \)).

Figure 1. TTHM concentrations from 7 CWS over 15 days in Eastern South Dakota and Southwestern Minnesota. Significant flooding occurred on day 0 that was not planned as part of the study. The EPA regulatory limit is marked by the dashed line.

Figure 2. Closest date- and location-matched CWS vs mean TTHM concentration from the 15-day period (linear fit: \( R = 0.85, P < 0.01 \)). Dashed line is the line of unity. Open circles are sample pairs that were ≥1 year apart.

Figure 3. CWS TTHM concentration vs NCS samples from 30 household tap water samples (second-order polynomial fit: \( R = 0.74, P < 0.001 \) for linear, \( P = 0.04 \) for quadratic). Dashed line is the line of equality. Note that ≥50% of the samples are below the line of unity, indicating that NCS concentrations were lower than the CWS-reported concentration.
Analysis was completed to test for an association between the number of days between the NCS and CWS results and the difference in the two results. The mean number of days between the NCS sample collection date and the closest date of the CWS monitoring sample was 107 days (range of 686 days before and 166 days after NCS sample date). There was no correlation between the number of elapsed days between the NCS and CWS sampling dates and the difference between the respective TTHM concentrations ($P = 0.55$, data not shown). For samples collected within ~1 month of each other, the difference in concentration ranged from $-7$ to 51.6 $\mu$g/l. For samples with a difference in results of $\pm 5 \mu$g/l, the number of days ranged from 337 days before to 101 days after the two samples were collected.

We analyzed associations considering the seasons the CWS and NCS samples were obtained. Twelve of the CWS-NCS sample pairs matched as both being collected in the summer season, but one of these pairs was off by 1 full year and one was off by 2 years, leaving only 33% that matched within the same season and year. Eleven were off by one season and 4 of these were from different years (9 months apart), whereas 7 were $\leq$ 3 months apart. Seven pairs were off by two seasons. Limiting the data to sample pairs that were collected in the same season and same year (33% of the data) or within 3 months of each other (57% of the data), we found a linear association between the CWS and NCS TTHM concentrations ($R = 0.78$, $P < 0.01$ and $R = 0.72$, $P < 0.01$, respectively). There was no association when the samples were off by two seasons ($R = 0.13$, $P = 0.8$).

DISCUSSION

In study 1, we found high day-to-day variability within 7 different CWS, with CVs ranging from 8% to 20%. We noted spikes in TTHM concentrations over a 15-day period that would not be identified if CWS TTHM monitoring concentrations were used to estimate participant exposure in epidemiological studies. The spikes seen may be related to the flooding event because of increased residence time of the water within the distribution system. One CWS reported that after a flood event much less water is used for watering lawns and gardens, resulting in increased residence time for water within the distribution system and increased formation of TTHMs. We do not think these spikes are because of treatment cycles as the companies we contacted do not treat or release water in cycles. Spikes were noted over the 15-day sample period and in a study such as the NCS, these spikes could coincide with specific windows of fetal development and would not be noted if CWS data were used as an indicator of exposure to TTHMs.

Variability in TTHM levels reported in previous studies is attributed to both spatial and seasonal factors.\(^1,15,17\) In study 1, six of the seven CWS in this region obtained water from the same type of water source, namely shallow wells, yet we found significant differences among mean TTHM concentrations sampled over the 15 days. Similarly, Legay et al.\(^15\) also reported significant differences in mean THM levels among different CWS obtaining water from the same type of water source within a region.

In study 2, we found that CWS and NCS household samples matched for the closest date and location had a polynomial association. Only 33% of the samples had CWS data that matched within the same year and season. CWS are required to sample during times when TTHMs are expected to be highest, namely summer. We investigated the particular sample pairs that had high CWS TTHM concentrations and low NCS household concentrations to test whether season, number of days between the two samples, or a particular CWS would explain the discrepancies, but they did not. We do not have data on the distance or time within the distribution system that water travels to reach each household. It is possible that spatial inconsistencies between the CWS sampling site and the household tap could explain the difference. Clearly, in rural areas it is difficult to match existing CWS sampling dates to the participant’s first trimester of pregnancy because of infrequent CWS sampling. Spatial factors may compound this challenge.

Our study demonstrates the limitations in using CWS data as an indicator of exposure to DBPs in rural areas. We found high day-to-day variability that could play an important role in possible misclassification of DBP exposure levels. The CWS monitoring data available for comparison to water samples from the NCS first trimester visit ranged from almost 2 years before to 0.5 years after study sample collection. Furthermore, NCS household tap and CWS monitoring concentrations of TTHMs were not linearly associated. In conclusion, TTHM concentrations from CWS monitoring data do not appear to be suitable to capture TTHM concentrations in the home. Future studies should be aware of these limitations and consider alternative water sampling strategies for participants in rural areas.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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