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Occurrence of regulated and non-regulated disinfection by-products in small drinking water systems

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HIGHLIGHTS

- The occurrence of 16 regulated and non-regulated DBPs were evaluated in 25 small systems.
- The gap between levels of regulated and non-regulated DBPs was comparable to that observed in large systems.
- All investigated DBPs followed a comparable seasonal evolution during the year.
- The seasonal fluctuations of DBPs were lower than those observed in large systems.
- Spatial variations of DBPs during distribution were high even if network sizes are small.

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ABSTRACT

The occurrence of regulated and non-regulated disinfection by-products (DBPs) was investigated in the drinking water of small systems in two provinces in Canada, Newfoundland and Labrador (NL) and Quebec (QC), through an intensive sampling program. Sixteen DBPs were studied: four trihalomethanes (THMs), five haloacetic acids (HAAs), four haloacetonitriles (HANs), one halonitromethane, chloropikrin (CPK) and two haloketones (HKs). Average measured concentrations of these compounds were much higher than those reported in the literature for medium and large systems. The measured average value for THMs was 75 $\mu g\,L^{-1}$ (Stdv = 69 $\mu g\,L^{-1}$); HAAs, 77 $\mu g\,L^{-1}$ (Stdv = 75 $\mu g\,L^{-1}$); HANs, 2.5 $\mu g\,L^{-1}$ (Stdv = 1.8 $\mu g\,L^{-1}$); CPK, 0.4 $\mu g\,L^{-1}$ (Stdv = 0.3 $\mu g\,L^{-1}$) and HKs, 6.0 $\mu g\,L^{-1}$ (Stdv = 4.5 $\mu g\,L^{-1}$). The gap (some 10 times difference) between the average levels of regulated DBPs (THMs, HAAs) and non-regulated DBPs (HANs, CPK and HKs) is comparable to that observed in large systems where the occurrence of the same compounds has been reported. Generally, investigated DBPs followed a comparable seasonal evolution during the year: they decreased between the fall and winter and then increased to eventually reach a maximum in late summer. This trend was less observable in NL than in QC. However, observed seasonal fluctuations of DBPs were less considerable than those observed in medium and large systems located in similar temperate environments reported in the literature. Spatial variations from the plant to the extremities were high and comparable to those observed in large systems, which is surprising, considering the smaller size of distribution networks supplying small communities. Generally speaking, the results support the premise that problems associated with implementing treatment that removes DBP precursors in water submitted to chlorination can increase population exposure to these contaminants in small systems.

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1. Introduction

Disinfection by-products (DBPs), generated by the reaction between a chemical disinfectant usually chlorine with organic matter, are an important concern for water supply, especially surface water supply, as they are generally rich in natural organic matter (Cedergren et al., 2002; Mouly et al., 2010).

DBPs constitute a large family of compounds presenting various levels of toxicological effects: more than 600 DBPs have been detected, but few have been identified (Richardson, 2011). Trihalomethanes (THMs) and haloacetic acids (HAAs) are the most prevalent DBPs in drinking water. Their formation is relatively well understood and their levels are regulated in various countries (in particular for THMs) (Singer, 2002; Richardson, 2011).

Recently, there has been an increased interest in investigating the presence of other DBPs, for example, haloacetonitriles (HANs), haloketones (HKs) and halonitromethanes (HNMs). In fact,

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nitrogen DBPs (like HANs and HNMs) may have greater toxicological effects than HAAs and THMs (Muellner et al., 2007; Richardson et al., 2007), that can become a public health problem with the increasing use of alternative disinfectants as a way to reduce concentrations of regulated DBPs such as chloramines (Adams et al., 2005).

Most studies on the occurrence of regulated and non-regulated DBPs have been conducted in large systems (Rodriguez et al., 2004; Krasner et al., 2006; Goslan et al., 2009; Ye et al., 2009; Mouly et al., 2010). However, small water systems (i.e., serving 5,000 or fewer people) using surface waters may be more vulnerable to DBPs because of financial constraints, a relatively low capacity to implement adequate treatment technologies to remove contaminants, in particular DBP precursors, and an inability to hire qualified operators (Charrois et al., 2004; Coulibaly and Rodriguez, 2004; Edwards et al., 2012). There is presently very little information on the spatio-temporal variability of DBPs in the water of small communities. Only a few studies on the occurrence of regulated DBPs are available (Charrois et al., 2004; Tung and Xie, 2009). In many countries, as is the case in Canada, available data are particularly inexistent for non-regulated DBPs.

The purpose of this study is to improve knowledge on the occurrence of regulated and non-regulated DBPs in the drinking water of small communities in Canada. Accordingly, spatial evolution (inter-regions, intra-regions and along the distribution system) and temporal evolution (seasonally) of DBPs in water were investigated. For the first time (according to our knowledge), the spatial and temporal presence of non-regulated DBPs was investigated in the drinking water of small communities based on intensive and structured sampling programs.

2. Methodology

2.1. Case studies

Twenty-five small municipal systems were selected and studied in two provinces of Canada: Newfoundland and Labrador (NL) and Quebec (QC). Sampling campaigns in the systems were conducted monthly for one year between September 2010 and October 2011 (from September 2010 to September 2011 in NL and from October 2010 to October 2011 in QC). All systems were supplied by surface water sources and used chlorine as their main disinfectant (for primary and secondary disinfection). Systems in NL served a population varying from 330 to 2120 inhabitants. In QC, systems served a population varying from 1000 to 6220 inhabitants. Systems in NL did not present any prior treatment to chlorination, whereas in QC, systems had implemented one or more treatment processes prior to disinfection.

2.2. Sampling and analysis

During this study 1500 samples were collected representing over 21 000 data for numerous parameters. Water was sampled at source (RW) and in the water treatment plant (WTP) just after filtration and before chlorination. Various points were identified along the distribution system (DS) in order to collect water samples at different residence times (Table 1). Water was sampled at the beginning (DS1), middle (DS2) and end of the DS (DS3). In NL, systems had no treatment prior to chlorination, chlorination being the main treatment process. Thus, in NL, RW and WTP were represented by the same point. Samples were collected by water operators (in NL) and by U. Laval personnel (in QC). Samplers were trained to follow equivalent sampling processes for both regions. Following field collection, samples were sent to the U. Laval laboratory for analysis.

Five families of DBPs were considered: THMs, HAAs and three families of non-regulated DBPs (HANs, HNMs, HKs). Four THMs (chloroform (TCM), bromodichloromethane (BDCM), dibromochloromethane (DBCM) and tribromomethane (TBM)), five HAAs (monochloroacetic acid (MCAA), monobromoacetic acid (MBAA), dichloroacetic acid (DCAA), trichloroacetic acid (TCAA) and dibromoacetic acid (DBAA)), four HANs (dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), bromochloroacetonitrile (BCAN) and dibromoacetonitrile (DBAN)), one HNM (chloropicrin (CPK)) and two HKs (1,1-dichloropropanone (DCP) and 1,1,1-trichloropropanone (TCP)) were analyzed during the study. The THM quantification limit was 3.6 $\mu g\ L^{-1}$ for TCM, 2.0 $\mu g\ L^{-1}$ for BDCM, 3.4 $\mu g\ L^{-1}$ for DBCM, 2.6 $\mu g\ L^{-1}$ for TBM. The HAA quantification limits were 1.0 $\mu g\ L^{-1}$ for all HAAs. The HAN, HNM and HK quantification limit were 0.01 $\mu g\ L^{-1}$ for all compounds. If the concentration of a DBP was lower than his LOQ, concentration was considered as null.

Many water characteristics were also measured. The sample strategy allowed us to obtain some 800 measures of each physical chemical parameter (turbidity, conductivity, UV absorbance at 254 nm (UV-254) and dissolved organic carbon (DOC)), some 300 for bromide, 490 for pH, about 470 for temperature and about 1000 for free chlorine and each THM, HAA and non-regulated DBP under study. Bromide was analyzed by the MA.303-3.1 method (Centre d'expertise en analyse environnemental du Québec, 2009). Physico-chemical parameters were not sampled in every location because we only wanted to gain a general overview of water characteristics in the DS. Table 1 summarizes parameters measured at each sampling point. Details about analytical methods used are provided elsewhere (Mercier-Shanks et al., 2013).

2.3. Data analysis

Data were collected in a detailed Excel database for which all descriptive analyses were carried out. SYSTAT 13 Software Version No.13.1 was used for statistical analyses for this paper. Statistical analyses included Student's *t*-test (for means comparison of water characteristics measurements between NL and QC) and ANOVA (for comparison of DBP mean levels between all systems in each region in order to detect a significant statistical difference in DBP levels between all systems belonging to a region), followed by a Games Howell test (for quantification of statistical differences between DBP levels in each system belonging to a region and identification of systems most or least correlated between each other).

3. Results and discussion

3.1. Portrait of DBP occurrence in small systems

Because there are regulations in QC that mandate water utilities supplied by surface waters to remove turbidity and NOM (mainly through filtration), the water submitted to chlorination (WTP) is of much higher quality in QC than in NL (data available in Supplementary section). The gap in levels of DBP precursor indicators (UV-254, DOC, SUVA and bromide) in water before disinfection explains why the DBP concentrations in small water systems (SWS) under study are significantly different between the two regions. In fact, the levels of these precursors have an impact on disinfectant demand and the potential for DBP formation. Table 2 presents the concentration distribution of all studied DBPs in SWS for both regions during the study period, as well as their 5% and 95% percentiles and coefficient of variation (CV).

It is important to note the gap between levels of regulated DBPs (THMs and HAAs) and non-regulated DBPs (HANs, HKs and CPK) in both regions. Both THM and HAA average concentrations were

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