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Variability of disinfection by-products at a full-scale treatment plant following rainfall events

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HIGHLIGHTS

- Slight increases in TOC and SUVA254 were measured in filtered waters after rainfall.
- Peaks in THM4 and HAA6 occurred in treated waters following rainfall.
- Rainfall leads to decreases in brominated THM and HAA proportion.

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ABSTRACT

The quality of drinking water sources can decrease when contaminants are transported by overland and subsurface flow and discharged into surface waters following rainfall events. Increases in organic contaminants such as road salts and organic matter may occur and potentially modify disinfection by-products (DBPs) concentration and speciation. This study investigated the effects of various spring rainfall events on the quality of treated waters at a large water treatment plant through the implementation of intensive water quality monitoring of raw, filtered and treated waters during different rainfall events. DBPs (four trihalomethanes and six haloacetic acids) and their explanatory variables (pH, turbidity, water temperature, specific ultraviolet absorbance, total and dissolved organic carbon, bromide and chlorine dose) were measured during four rainfall events. The results showed that water quality degrades during and following rainfall, leading to small increases in trihalomethanes (THM4) and haloacetic acids (HAA6) in treated waters. While THM4 and HAA6 levels remained low during the pre-rainfall period (<9 μg/L) for the four sampling campaigns, small increases in THM4 and HAA6 during and after spring rainfall events were observed. During the rainfall and post-rainfall periods, concentration peaks corresponding to 3-fold and 2-fold increases (respectively 27.5 μg/L for THM4 and 12.6 μg/L for HAA6) compared to pre-rainfall levels were also measured. A slight decrease in harmful brominated THM and HAA proportion was also observed following rainfall events.

1. Introduction

Climate can affect freshwater drinking water sources in several ways. The quality of source waters may decrease when contaminants are transported by overland and subsurface flow and discharged into surface and ground waters (McCullough et al., 2012; St-Hilaire et al., 2015). De-icing salts commonly applied on roads in winter in North America, may also produce saline waters that run off into the environment and contaminate surface waters during rainfall events (Dailey et al., 2014). According to Cooper et al. (1985), de-icing salts may also constitute a source of bromide in source waters. It has been shown that chloride salts are generally contaminated by bromides (Granato, 1996). Due to widespread suburban development, roadway extension and related de-icing salt use, a rise in salinity in urban and rural streams has been observed in northeastern America, impacting aquatic environments and drinking water supplies (Kaushal et al., 2005). Surface waters in eastern Canada (e.g., Québec, Ontario) are particularly sensitive to this type of contamination (Evans and Frick, 2001; Mayer et al., 1999).

Changing precipitation patterns caused by climate changes could have a deleterious effect on the chemical and microbiological quality of water intended for human consumption (Beaudeau et al., 2011; Delpla et al., 2009; Jiménez Cisneros et al., 2014; Roig et al.,

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2011). Indeed, with climate change, changes in rainfall intensity, frequency and duration could possibly lead to modifications in contaminant transport by run-off from the watershed to surface water sources (rivers, lakes). This in turn could result in elevated levels of suspended sediments, organic matter (OM), pathogens and other organic contaminants in surface waters (Delpla et al., 2009; Whitehead et al., 2009). Furthermore, with climate change, an increase in freeze-thaw cycles frequencies and winter rainfall are predicted in the Province of Quebec, Canada (Des Jarlais et al., 2010). This could potentially lead to increased surface waters vulnerability to water pollution events during the cold season.

Few studies have assessed the effects of climatic variability on the quality of drinking water supplied by municipalities. However, consequent to difficulties experienced by water treatment facilities to respond adequately and quickly to rapid variations in source water quality, one might expect a deterioration of drinking water quality (Li et al., 2014; Ritson et al., 2014). Some studies have focused on the impacts of a changing climate on the variability of OM quality and quantity in source water and on its treatability (Gough et al., 2015; Nguyen et al., 2013; Ritson et al., 2014). A rise in OM in raw waters could pose a series of treatment difficulties such as membrane fouling and blocking, and operational cost increases (higher chlorine demand and disinfectant dose, coagulant dose, pH adjustments). It could also impact odour, taste and colour, and increase binding and transport of pesticides and pharmaceuticals (Ritson et al., 2014). Furthermore, an increase in OM could also favour the formation of potentially harmful disinfection by-products (DBPs) during water disinfection. As an example, recent studies have noticed an increase in DBP formation potential in raw waters following rainfall events (Jung et al., 2014; Pifer et al., 2014). DBPs are an important issue in drinking water because of their potential health effects: cancers (e.g., bladder and colorectal) and reproductive outcomes (Costet et al., 2011; Levallois et al., 2012; Richardson et al., 2007). More than 600 DBPs have been identified in water (Richardson et al., 2007). Among them, trihalomethanes (THMs) and haloacetic acids (HAAs) have been studied more widely due to their relatively high concentrations and prevalence in drinking water. Increasing levels of bromide in raw water could also affect DBP speciation and shift DBP formation to more brominated species (Luong et al., 1982; Pan and Zhang, 2013; Pourmoghaddas et al., 1993; Roccaro et al., 2013) that are suspected to be more carcinogenic than their chlorinated analogs (Nobukawa and Sanukida, 2000; Pals et al., 2013; Richardson, 2003).

Some modelling attempts have been made to predict the DBP variations associated with climate. In a study conducted on 112 drinking water utilities in Québec, Cool et al. (2015, *submitted*) showed that changing temperature and precipitation patterns with climate change could eventually increase the probability of THM concentrations exceeding a threshold of 80 μg/L by the end of the century. Another study by Delpla et al. (2016) showed that a small increase in DBP levels in drinking water (THMs and HAAs) might be expected with climate changes, with the highest increases in winter and spring due to changes in precipitation and a reduction of the duration of the frozen period. Both studies note important differences according to season and the type of treatment implemented, highlighting an increasing resilience capacity associated with the complexity of the treatment process.

Despite an increasing number of DBP modelling studies, no field studies in full-scale conditions have been carried out on the impact of rainfall on DBP variation and speciation, even in large treatment plants. Consequently, drinking water managers and operators continue to lack knowledge to efficiently adapt their practices to the potential impacts of climatic variability on DBP formation. It is particularly important to understand variations of DBP formation during the spring when the ice cover disappears, water temperatures increase and surface waters are potentially more vulnerable to contamination by OM and salts. The aim of this study is to provide data on the impact of spring rainfall events on raw and drinking water quality (DBPs and their precursors) based on high-frequency field monitoring campaigns.

The specific objectives of this study are: i) to improve the understanding of the variation of DBP precursors levels following rainfall events during the spring season; ii) to assess the impacts of rainfall events on the variability of DBP concentration and speciation (in particular THMs and HAAs), and iii) to propose a model-based assessment of the impact of rainfall events on DBPs.

2. Materials and methods

2.1. Study site

The study was carried out in a catchment located in the region of Québec City, Canada. The Saint Charles river watershed is partially located within the city of Québec. The area is mainly covered by forest and urban land (Jobin et al., 2007). The watershed is known to be polluted by road salt application leading to high levels of chloride and high conductivity (APEL, 2010). This river is used as a water source by a large-size water treatment plant to produce drinking water. The Québec water treatment plant provides water to 306 000 inhabitants. Treatment steps include coagulation/flocculation with aluminium salts (alum), sedimentation, inter-ozonation, and filtration (primary treatment) followed by chlorination.

2.2. Sampling and analysis

Two 24-bottle refrigerated autosamplers (ISCO 6712FR) were installed at the Québec water treatment plant, one at the raw water intake and one after the filtration step, just before final chlorination. Since some monitored DBPs (namely THMs) are volatile compounds, manual sampling was performed after disinfection and just before the distribution system (treated waters) in order to minimize DBP losses. Four borosilicate vials for THM and HAA analysis were filled and all the necessary precautions taken to avoid loss of volatile compounds. Each vial contained a dechlorinating agent (NH_4Cl) to neutralize free chlorine residual and avoid further DBP formation during sample transportation to the laboratory. Additionally, we took account of the water transit time between different points in the treatment works (from the raw water uptake to final chlorination) for the sampling campaign implementation. Then, for a single campaign, a delay between raw water sampling and drinking water sampling was set to approximately 90 min to match the residence time of water within the plant at the time of monitoring.

High frequency sampling campaigns were carried out after at least three consecutive dry days (rainfalls<1 mm) and for forecasted rainfall height greater than 10 mm in 24 h. This rainfall threshold was used to define heavy rainfall (Aguilar et al., 2005; Vincent and Mekis, 2006). A total of 4 rainfall events were monitored during the 10-week study period (between April 9 and June 18, 2015) using these requirements. The sampling frequency was adapted to the duration of the rainfall event in order to capture the post-rainfall effect on water quality. The frequency was 1 sample/4 h for short-duration rainfall events and 1 sample/8 h for long-duration rainfall events. Then, the monitoring length for each campaign was extended from 3 to 7 days during our study, depending on the duration of the rainfall.

During each sampling campaign, samples were collected and transported to the laboratory every day, between 1 h and 24 h after collection. Preservation agents were added if needed at the