Variability of water residence time and free chlorine and disinfection by-product concentrations within a residential neighborhood

Simon Rochette, Geneviève Pelletier, Christian Bouchard and Manuel Rodriguez

ABSTRACT

Sampling campaigns at several points in a sector of a distribution network were conducted during several months to study the spatial and temporal variation of the free chlorine concentration (FCC) and disinfection by-product concentrations (DBPC) at the scale of a residential neighborhood. The water residence times (RT) at these sampling points and for the same period were also simulated using EPANET. Within the neighborhood, the results reveal significant differences in measured FCCs and DBPCs and low intra-day variability. Catching most of the FCC variability would require several sampling points to cover the spatial variability, whereas for DBPC, it is more relevant to obtain a higher sampling frequency. Moreover, the sampling point choices for DBPC should be based on previous FCC monitoring campaigns. At the studied scale, the RT alone is not the best indicator for the selection of the monitoring points for FCCs and DBPCs.

Key words | disinfection by-product, distribution network, drinking water, free chlorine, spatial and temporal variability, water residence time

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INTRODUCTION

Drinking water distribution network (DN) managers look to reduce risks linked to consumption of distributed water. Acute risks linked to transmission of waterborne diseases through regrowth or intrusion of pathogenic microorganisms in DNs (LeChevallier *et al.* 1996; Zhang & DiGiano 2002) are reduced by ensuring a minimum free chlorine concentration (FCC) in a DN and by proper maintenance procedures. On the other hand, disinfection by-products (DBPs), generated by chlorine's reaction with natural organic matter (NOM), pose potential chronic risks. DBPs found in greater concentrations include trihalomethanes (THMs) and haloacetic acids (HAAs) (Singer 1994).

Water quality is affected by the water treatment plant (WTP) and water residence time (RT) in DNs (Al-Jasser 2007; Machell & Boxall 2014). Free chlorine degrades as water travels in pipes, and it has been shown that areas with a higher RT are specifically linked to higher THM concentrations (Simard *et al.* 2011). However, certain HAA species are biodegraded by biofilms present on pipe walls, which can lead to a reduction in concentrations in areas with a higher RT (Pluchon *et al.* 2013). Problem areas in terms of low FCCs or high DBP concentrations can therefore be identified using relationships established between these parameters and the RT (Farren 2003; Rodriguez *et al.* 2007; Simard *et al.* 2011).

Previous studies also show that the RT has a greater impact in areas serviced by smaller pipes because there is more contact between the water and pipe walls (e.g. Vasconcelos *et al.* 1997). Presence of biofilm and wall corrosion lead to faster chlorine degradation over time (Lu *et al.* 1999; Dietz *et al.* 2007).

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Many studies have shown how the RT, FCC and DBP concentrations vary in a DN. RTs were considered directly (tracer tests or hydraulic modeling) or indirectly (via distance from the WTP or FCC) (Rodriguez *et al.* 2003; Toroz & Uyak 2005). In these studies, samples were collected weekly (Rodriguez *et al.* 2003; Toroz & Uyak 2005), bimonthly (Rodriguez *et al.* 2007), monthly (Summerhayes *et al.* 2011) or seasonally (Uyak *et al.* 2014). Significant spatial and temporal variability in FCC, THM and HAA concentrations have been observed throughout the DN. However, studies have not been conducted on a spatial scale small enough to evaluate local variations of water quality. Yet, this is important in terms of protection against risks of infection and for the selection of sampling points for operation purposes and verification of regulatory compliance.

The main purpose of this study is therefore to evaluate the spatial and temporal variability of RTs and water quality (FCC, THMs and HAAs) within a residential neighborhood to develop better water quality monitoring strategies supported by simple hydraulic modeling of RTs. The two specific purposes are to: (1) evaluate the spatial and temporal variability of the simulated RT, and FCC and DBP concentrations; and (2) identify the extent of the dominant variability, spatial or temporal, for FCC and DBP concentrations.

METHODOLOGY

To our knowledge, this is the first study to quantify the spatial and temporal variability of measured FCC and DBPs, at the scale at which a water manager must often select just one sampling point for regulatory monitoring. The methodological steps to generate the data presented in this paper can be generalized to comparable sites. Similar trends in variability of FCC and DBPs are expected within neighborhoods serviced by a DN with a large range of diameter of pipes, where free chlorine degradation and DBP formation happen mainly at the pipe wall (Al-Jasser 2007).

Case study

A well-defined area of Quebec's DN is studied and modeled to characterize the variations of water quality and RTs. This DN, located in a Nordic region, is characterized by major seasonal temperature and raw water quality variations. These conditions lead to frequent water treatment adjustments affecting the kinetics of free chlorine degradation and DBP formation. The residential neighborhood has nearly 6,000 inhabitants and covers an area of almost 2 km^2. The territory is mostly residential. Figure 1 presents the study area, the neighborhood's connection to the main pipes, valve chambers (VCs), location of the sampling points and population distribution. The neighborhood is supplied by two VCs (VC-A and VC-B) equipped with flow meters. Flows measured at VC-A are about 9% of VC-B flows, on average, during the study period. The neighborhood is serviced by 50 km of 30-year old (or more) cast iron pipes with diameters varying from 75 to 600 mm. The case study was selected to represent a typical neighborhood built in the 1960–1970 period, which was a period of rapid urban development not only in Quebec City but across the Northern East coast of North America. The few industries, businesses and institutions (IBI) have flow meters for their respective consumption.

Hydraulic modeling

Simple modeling of the RTs of a DN can be done using limited information (Khanal & Speight 2008). Hydraulic models are now widely used, often free access and the input information needed to run simulations is now much more accessible. Indeed, an uncalibrated hydraulic model can be built using information that is normally accessible to network managers such as the original features and age of the pipes, total quantity of water consumed and population distribution. In our case, RTs were modeled (not obtained from a tracer study) using EPANET 2.0 v.2.00.12 (Rossman 2000) with the *Age* analysis tool. Calculations of RTs in EPANET, established on the principles of mass and energy conservation, use a time-based Lagrangian approach to track the movement of each water element through the pipes, including mixing at nodes.

A thorough analysis of flow rates at the two VCs over each of the 22 campaign days led us to conduct steady-state simulations given the small flow variations during the 9 a.m. to 5 p.m. period, while significant increases were observed in the morning and at suppertime and decreases

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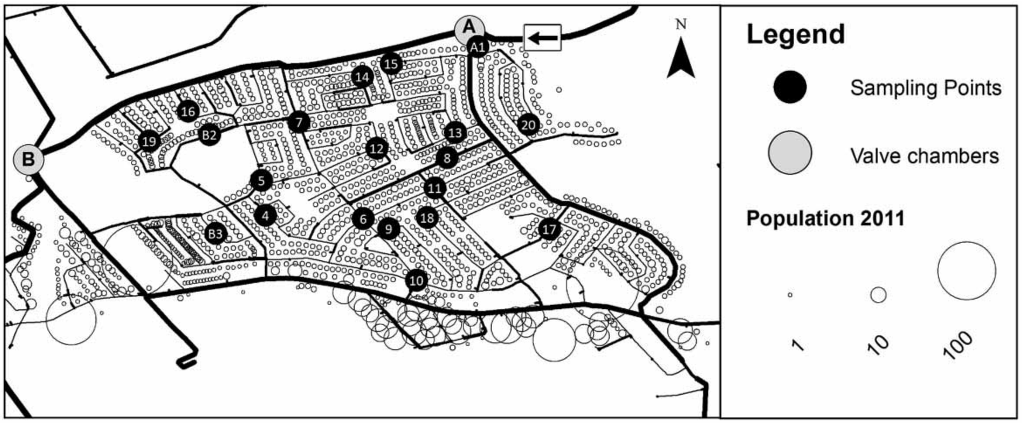

Figure 1 | Sampling points, VC locations and population distribution in the study area.

from daytime to nighttime. Entry flows measured at the VCs varied from 4,600 to 5,900 m^3/day. During a tracer test, the RT between the WTP and VC-A was estimated to be 6 hours (Delisle *et al.* 2015).

The features of the neighborhood allow for RTs to be modeled with limited uncertainty as entry flows are known and allow for flow to be distributed according to the population: the relatively homogenous occupation of the neighborhood limits errors linked to flow distribution. In fact, distribution of water demands within the network has the largest impact on RTs because it governs the hydraulic paths through the network but, in this case, the land use is residential with no large water consumer (e.g. industries) that can strongly affect hydraulic paths in a different way than residential use for which the patterns (peaks and lows in water consumptions) are better understood. Given the location in the city's DN, a mixture of water from different sources is unlikely. There is no tank nearby. However, the actual roughness of pipe walls, the actual diameter of the corroded pipes and the leakage flow distribution are unknown.

Evaluation of water quality

A sampling program was carried out to evaluate water quality between May and September 2012 and to observe the variability linked to the Spring and Summer seasons (during which water temperature goes from cold to warm and back down again in September) and to the hydraulic conditions in the DN. Fall and Winter seasons were excluded because DBP concentrations are known to be quite low when the water temperature is low. There were 22 sampling campaigns spaced 7 days apart except for the first two campaigns, which were spaced 5 days apart. The end of the sampling period corresponded to the shutdown of outdoor residential taps from which the samples were collected, with the winter frost approaching.

Twenty water sampling points were selected and monitored. For each campaign and for each point, three samples were collected at different times of the day, including in the morning (between 8 a.m. and 10 a.m.), at midday (between noon and 2 p.m.) and in the afternoon (between 4 p.m. and 6 p.m.) to also evaluate the intra-day variability. Location of sampling points were selected based on neighborhood coverage in terms of local and secondary pipes and are presented in Figure 1. Sample collection and field and laboratory measurement procedures are presented in Section 1 of the Supplementary Material (available with the online version of this paper).

There was no direct access to the VCs given their location in the middle of the road and the impact on traffic. In terms of RT, the closest residence to VC-A is sampling point A1 and sampling points B2 and B3 for VC-B. Counterintuitively, the neighborhood is mainly supplied by VC-B: RT increases and FCCs decrease starting from VC-B, while VC-A supplies a limited area associated to the secondary pipe to which it is directly connected. This limited area also undergoes a mixing of older water coming from VC-B.

HAA analysis was performed via the technique of liquid-liquid extraction by gas chromatography with

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electron capture detection. The average recovery for MCAA (monochloroacetic acid), MBAA (monobromoacetic acid), DCAA (dichloroacetic acid), TCAA (trichloroacetic acid), BCAA (bromochloroacetic acid) and DBAA (dibromoacetic acid) are 105%, 111%, 106%, 102%, 93% and 93%, respectively. THM analysis was performed via the extraction technique of SPME fiber by gas chromatography coupled to a mass spectrometer (ion trap). The average recovery for TCM (trichloromethane), BDCM (bromodichloromethane), DBCM (dibromomethane) and TBM (tribromomethane) are 98%, 96%, 97% and 95%, respectively.

Statistical analysis

A variability analysis was conducted using the SAS software to determine if there was a significantly dominant dimension between the spatial and temporal dimension in the variability of FCCs and DBPs. This analysis was conducted with generalized linear mixed models to compare the spatial variability associated with the location and the temporal variability linked to the day during which the samples were collected. This is meant to determine if there is a significant statistical difference ($p < 0.05$) between these variability components using SAS's PROC GLIMMI procedure.

RESULTS AND DISCUSSION

Mean values of water quality parameters (refer to Section 2 of Supplementary Material, available with the online version of this paper) confirmed orders of magnitude comparable to observations made during previous studies conducted on other parts of the City's DN (Rodriguez *et al.* 2003; Sadiq & Rodriguez 2004). There were major variations in terms of the FCC, and THM and HAA concentrations (differences between the minimum and maximum values) during the monitoring period, despite the relatively small size of the DN (refer to Section 2 of Supplementary Material for details). FCCs at the exit of the WTP and in the neighborhood increased from July with the increase in temperature which is followed by an increase in chlorine dosage to prevent a quick free chlorine deterioration in the DN.

THM and HAA levels remained below the regulatory standard values of 80 and of 60 μg/L (MDDELCC 2012) for the annual average, respectively, for all sampling campaigns. For THMs, the species present in quantifiable concentrations were TCM and BDCM, while for HAAs, only MCAA, DCAA and TCAA were present in quantifiable concentrations given the low bromide concentrations found in the water (refer to Section 3 of Supplementary Material for details, available with the online version of this paper).

Spatial and temporal variabilities of the simulated RT

Figure 2(a) and 2(b) present the simulated RTs that varied from 8 to 30 hours, including the 6 hours between the WTP and VC-A. The RT between VC-A and VC-B was estimated to be approximately 2 hours. Flows measured at VC-A and VC-B varied from 123 to 798 m^3/day and from 4,260 to 5,232 m^3/day, respectively, leading to a ratio of VC-A over VC-B of 3% to 15% depending on the campaign (Figure 2(b)).

The temporal variability of RTs was very low for most points (Figure 2(a)). The difference between the maximum and minimum RTs for the 22 campaigns was less than 2 hours for 18 of the 20 points, while points 18 and 20 showed a difference of 9 and 14 hours, respectively, which can be explained by their location. When water consumption peaks, water in the pipe supplying point 18 flows southbound with a larger flow resulting in a low RT. However, when consumption is lower, the supply comes from different water paths, resulting in a significant increase of the RT. The variability of the results for point 20 is linked to the low flow associated with the isolated loop on which this point is located.

RTs and flows simulated at both VCs for each campaign are presented in Figure 2(b). Entry flows at the VCs correspond to a consumption of 450 to 600 L/person/day. This order of magnitude is expected given leakage and IBI flows. RT distribution undergoes little change between the campaigns; however, the campaigns of July 17th and September 25th present higher maximal values. These values are linked again to points 18 and 20 for campaigns presenting lower flows at VC-A (123 and 129 m^3/day compared to an average of 424 m^3/day). High RT values are linked to the flow ratio between the two VCs and the water paths to supply these points. Higher RTs are associated with a change in the supply direction in the loops within which these points are located. Distribution of entry flows has a major impact on

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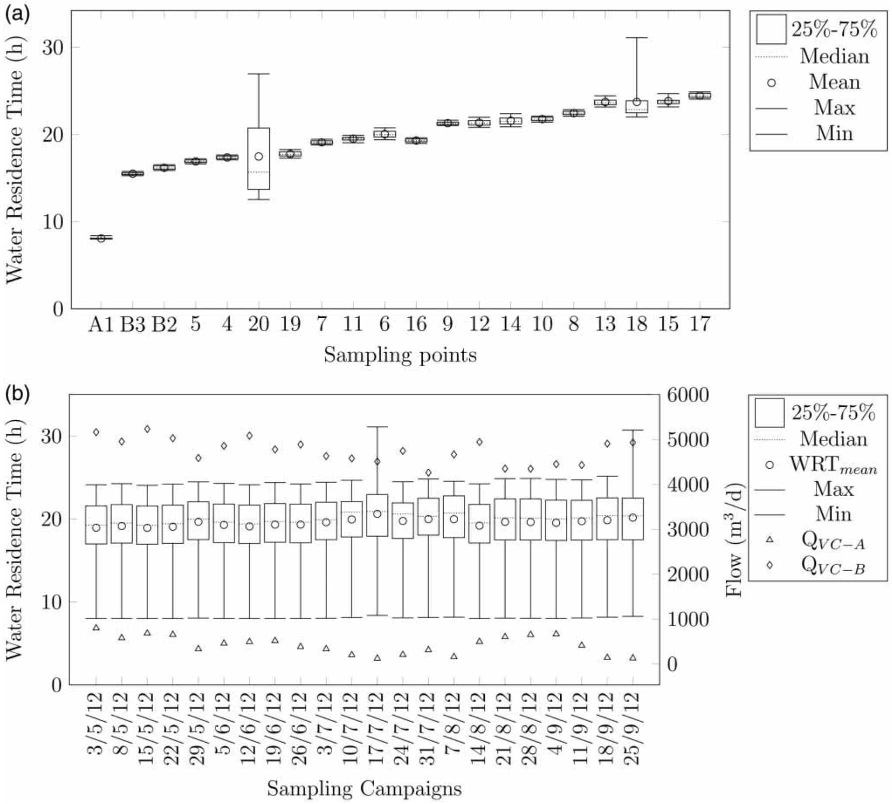

Figure 2 | Simulated RT from the WTP of: (a) all the samping points for all sampling campaigns; (b) all sampling campaigns and VC average flows.

RTs: the area located close to VC-B has lower RT values given the significant flow transiting via the main pipes.

Spatial variability of water quality

Figure 3(a) presents the spatial variability of FCCs. Points 19 and 20 present the lowest ranges of FCCs. These two points are located in an area of small-diameter (150 mm) pipes forming isolated loops, two conditions that are conducive to a significant FCC reduction. Degradation of chlorine takes place in the bulk flow and at the pipe wall and is accelerated at higher temperatures. At the neighborhood scale, the RT does not vary so much when compared to the variation that can be found in an entire DN. Small diameters, pipe age and metallic materials favour free chlorine degradation at the pipe wall. That is why, for such cases, time alone is not sufficient to explain spatially distributed FCC.

However, the spatial variability of FCCs is partially related to RTs. Indeed, certain points linked to the lower FCCs are linked to higher RTs (points 17 and 18), which is expected. However, the opposite is also true for points A1, B2 and B3. The lowest mean FCCs (points 19 and 20) do not correspond to the highest mean simulated RTs. At a given point and from a chemical perspective, FCC depends on the RT from the last point of chlorination, temperature and presence of NOM (essentially the same for all points), but also depends on chlorine degradation with the walls themselves according to the size of the pipes, their material and their level of corrosion.

The coefficient of variation (CV) was also calculated for each point. These results are shown in Section 4 of the Supplementary Material (available with the online version of this paper). The highest CV values are linked to the points with the lowest FCCs: 0.48 for point 18, 0.58 for point 19 and 0.90 for point 20. High CV values can be attributed to seasonal variations, which illustrates the challenge of maintaining relatively constant FCC levels throughout the year. Point 10 also presents a higher CV (0.43) and the greatest gap between the 25th and 75th percentile (0.5 mg/L),

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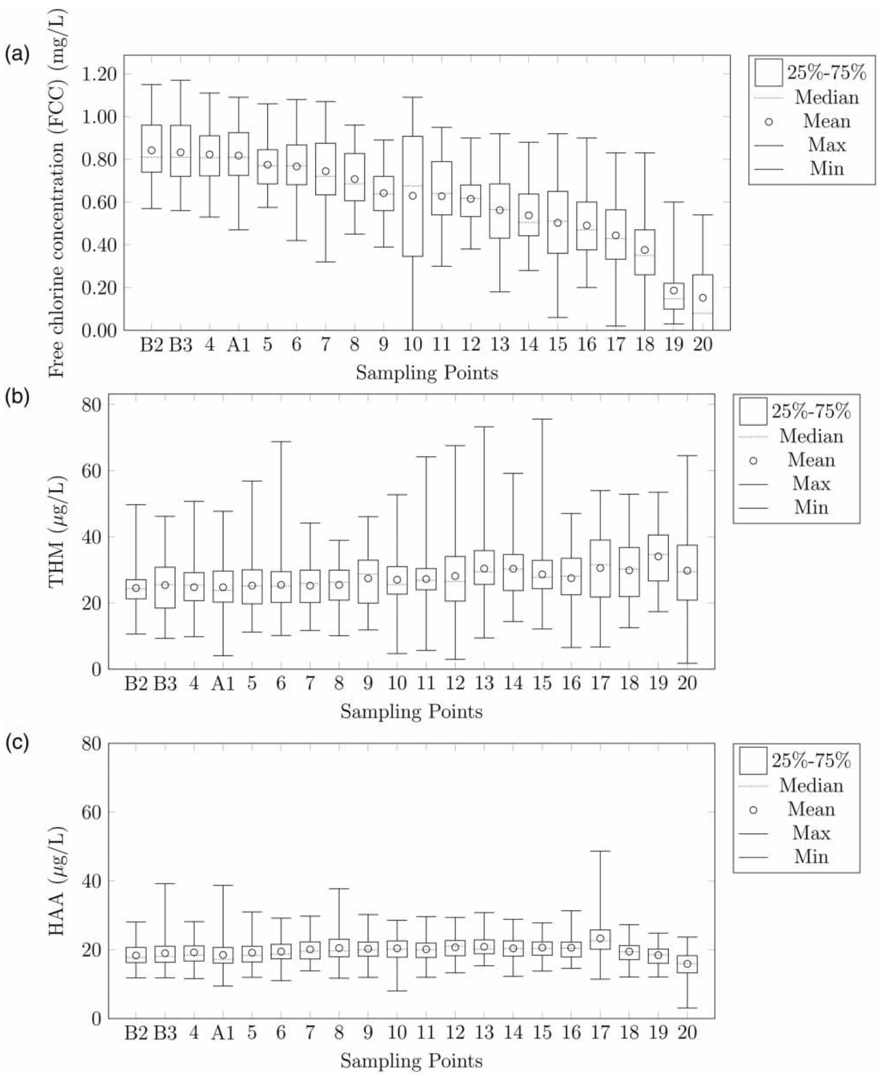

Figure 3 | Spatial variabilities of: (a) FCC; (b) THM; (c) HAA.

while this gap is less than 0.2 mg/L for the other points. This point is located close to a larger secondary pipe in which transits a larger flow: water flowing in the secondary pipe is characterized by a lower RT and less contact with the pipe walls.

Figure 3(b) illustrates the distribution of THM concentrations for each point. The average concentrations vary between 24 and 34 μg/L and the CV between 0.24 and 0.35 (refer to Section 4 of Supplementary Material for details). This translates into large variations at each monitoring point. Although the maximum THM concentrations can be found at the extremities of the DN, areas presenting a high RT also include isolated loops with small water demands, as was observed in numerous studies conducted within a DN (Rodriguez *et al.* 2004; Simard *et al.* 2011).

Figure 3(c) illustrates the range of HAA concentrations for each point. The average concentrations vary between 15 and 23 μg/L and the CV between 0.13 and 0.28 (refer to Section 4 of Supplementary Material). Points 17 and 20 present extreme concentrations, the highest and lowest, respectively. The CV of these two points is also greater than that of other points. Higher concentrations are found at point 17 because it is supplied by a path composed of pipes with a larger diameter, but presenting a lower flow rate: the RT is high enough for the free chlorine to deteriorate, but the relatively high diameter does not foster the contact of water with pipe

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walls and does not lead to the biodegradation of HAAs via the biofilm. Point 20 presents HAA concentrations that are slightly lower than other points (Figure 3(c)). Knowing that certain HAA species, namely DCAA, may undergo biodegradation via the biofilm present on the internal surface of pipes (Rodriguez *et al.* 2004; Pluchon *et al.* 2013) and that this point presents lower FCCs, this behavior is expected. The impact linked to pipe walls coupled with a potentially higher RT can explain this observation.

Risks considered for HAA and THM regulatory standards are chronic risks linked to average exposures. Sampling points used to monitor DBPs should thus be selected based on mean DBP concentrations. On the other hand, to avoid selecting a point that could lead to the overestimation or underestimation of risks, it would be logical to select monitoring points with the lowest variability. This is true because, from a regulatory perspective, a mean annual concentration is calculated using a few *ad hoc* measurements in time. For the neighborhood studied, and for the punctual and mean values per point, the relationships between DBP concentrations and estimated RTs are not very good. It would therefore not be recommended to use estimated RTs to select DBP sampling points.

However, relationships between mean DBP concentrations and mean FCCs are more significant as shown in Figure 4(a) for THMs and Figure 4(b) for HAAs, despite the relatively low variations in mean DBP concentrations. As shown in Figure 4(b), the mean HAA concentration goes through a maximum that corresponds to a mean FCC of approximately 0.5 mg/L. Information on mean FCCs can therefore be used to select DBP monitoring points within this neighborhood.

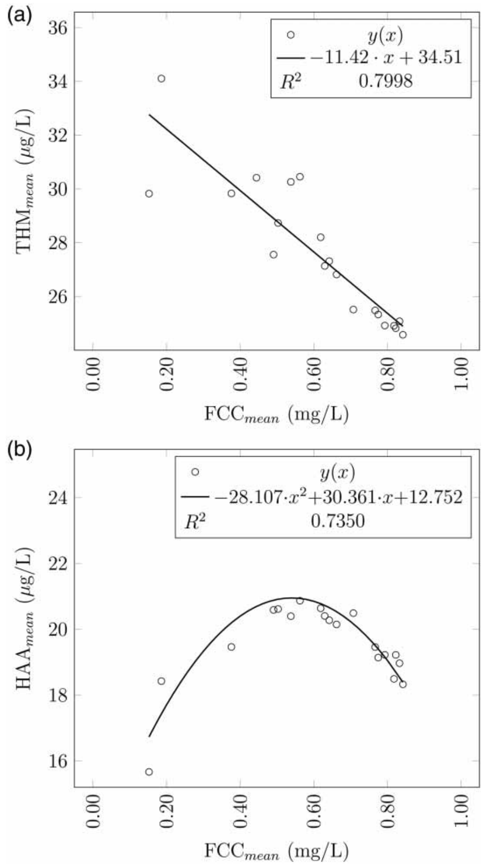

Figure 4 | Use of average FCC as an indicator for DBPs: (a) THM; (b) HAA.

Temporal variability of water quality

The average difference between two FCC measurements for the same point during the same day is 0.08 mg/L (median of 0.05 mg/L). FCC differences during the same day are thus small. Consequently, collecting one sample a day can be sufficient to obtain adequate information for this neighborhood. For DBPs, the average difference is 5.5 μg/L (median of 3.8 μg/L) and 2.0 μg/L (median of 1.3 μg/L) for THMs and HAAs respectively. The average for all THM and HAA measurements is 27.6 μg/L and 19.7 μg/L, respectively. Therefore, the intra-day variability is also very low. The time of day during which the water sample is collected does not have a major impact on the results, despite the local flow variations throughout the day.

Figure 5(a) presents the free chlorine demands between the WTP and the various points of the campaigns (FCCds). CVs were calculated for data relative to the FCCd, THM and HAA for each campaign and are presented in Section 5 of the Supplementary Material (available with the online version of this paper). As expected, campaigns during which the water temperature is lower present lower FCCds given the slowdown in chemical reactions. However, campaigns in May have both the lowest temperatures and the highest CVs. This higher variability can be explained by the water properties, given that the month of May has the highest average turbidity (0.23 UTN) and average UV absorptivity at 254 nm (0.026 cm^{-1}). The specific UV absorbance (SUVA) (0.018 mg/L.cm) is also the highest. Greater SUVA values

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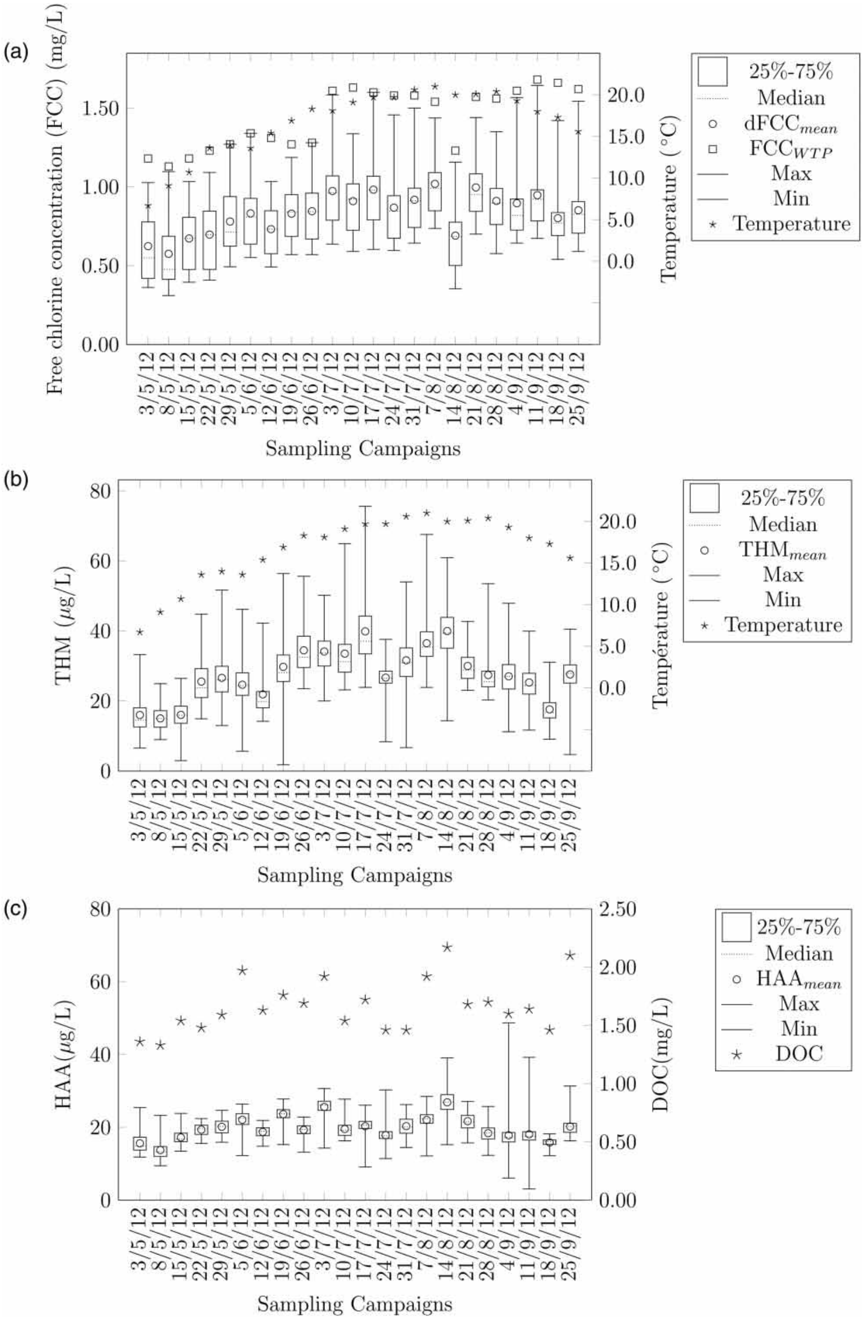

Figure 5 | Temporal variabilities of: (a) FCC; (b) THM; (c) HAA.

indicate a larger proportion of aromatic organic compounds in water, which can lead to higher FCCd (Abdullah *et al.* 2009). These factors can therefore explain the higher variability observed during the first campaigns in the month of May.

Figure 5(b) and 5(c) present the range of THM and HAA concentrations obtained for each campaign. For THMs, a value of 3 to 11 μg/L separates the 25th and 75th percentile during the campaigns, or 10 to 30% of the mean concentration. For HAAs, a value varying from 1 to 3 μg/L separates the 25th and 75th percentile for all campaigns, or approximately 10% of the mean concentration. The concentration variability during one campaign is therefore

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higher for THMs than HAAs. According to Figure 5(b) and 5(c), HAA concentration distribution remains more centered on the median value than THMs. The scale of the neighborhood and RTs therefore allow for additional THM formation leading to greater variability. In terms of HAAs, a certain balance between their formation and biodegradation can explain the lower variability.

Dominant variability dimension in water quality: temporal or spatial

A statistical analysis of the spatial and temporal variability of FCCs, THMs and HAAs was conducted to determine a dominant dimension. For the monitoring of water quality, such an analysis helps to determine if it is better to select more points or to increase the sampling frequency to better represent the variability. The number of measurements considered and the covariance parameters obtained and their respective error are presented in Section 6 of the Supplementary Material (available with the online version of this paper). Covariance parameters help to draw conclusions regarding the dominant dimension (point or campaign) and the χ^2 test helps to validate the statistical significance of this difference.

Spatial variability is much higher for FCC. Therefore, to obtain a portrait of FCC variability, the selection of different sampling points should be favored rather than taking repetitive measurements at the same points. Although the spatial dimension is the dominant aspect of FCC variability, treatment conditions at the WTP also influence the variability of concentrations at a given point of the DN.

DBP concentrations present a dominant temporal variability. Therefore, to obtain a representative portrait of THM and HAA, it is important to take into account the variability of conditions at the exit of the WTP, such as the chemical oxygen demand or temperature, rather than the location in the DN or within a residential neighborhood. Despite major FCC differences illustrating significant RT differences, the DBP levels found in the DN do not vary much from one point to another during a campaign. In order to characterize DBPs within the neighborhood, a higher sampling frequency, but for a limited number of points, should therefore be favored rather than numerous sample points at a reduced frequency.

Given the costs of laboratory analysis of DBPs, the number of samples analyzed by municipalities to ensure compliance with standards remains limited. It is important to take into account the fact that DBP monitoring is a way to verify that the limit allowed is not exceeded by calculating an annual concentration mean (Sadiq & Rodriguez 2004). This average is meant to represent the mean exposure level of citizens to these compounds. Obtaining a representative mean must take into account DBP concentration variability and, according to the results obtained within this neighborhood, this variability is mainly linked to the temporal dimension, or the variability of the characteristics of raw water at the exit of the WTP.

CONCLUSIONS

This study was conducted within a small residential neighborhood in which 22 campaigns with high-frequency sampling at several points of the DN were led. The results obtained show that, at this scale, there can be significant differences in simulated RTs, and measured FCCs and DBP concentrations. Results also show that water quality monitoring in the DN, performed by collecting a single sample during the day, does not limit the representativeness of the parameter variability given the low intra-day variability of FCC, THMs and HAAs. Within the neighborhood studied, the representativeness of the FCC variability resides in samplings at numerous points while the representativeness of THM and HAA variability is ensured by a higher sampling frequency. It is also shown that the campaigns used to monitor the free chlorine, a parameter that is inexpensive to measure, can help to select sampling points for THMs and HAAs. In summary, this study informs water managers that water RTs as calculated by EPANET, while easy to obtain, will not give as good an information as an intensive FCC campaign can, which one person can realize in one day with readily available equipment.

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