Practical Paper

Spatial and seasonal evolution of trihalomethanes in water distribution systems in Lebanon

Lucy Semerjian, John Dennis and George Ayoub

ABSTRACT

Spatial and seasonal changes in trihalomethane concentrations were investigated in various water distribution systems in Lebanon supplied by different water sources. A seasonal sampling program was initiated during which trihalomethanes and various contributing water quality and operational parameters were monitored at points along the distribution system. The majority of samples collected from points within the distribution network exhibited higher trihalomethane concentrations (1–13 folds) when compared to total trihalomethane levels detected in samples collected directly after chlorination. Recorded trihalomethane concentrations in Lebanon were highest in the spring, followed by the summer and winter. Moreover, total trihalomethane concentrations were higher when the abstraction sources were surface waters compared to ground water sources. Also, higher mean trihalomethane values were recorded in chlorinated waters originating from sources subjected to semi-conventional water treatment before the application of chlorine compared to mean trihalomethane values established in chlorinated waters originating from sources subjected to direct chlorination.

Key words | chlorination, distribution networks, drinking water quality, Lebanon, trihalomethanes

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ACRONYMS

AUB	American University of Beirut
DAC	Directly after chlorination
EERC	Environmental Engineering Research Center
NOM	Natural organic matter
THM(s)	Trihalomethane(s)
TOC	Total organic carbon
TTHM(s)	Total trihalomethane(s)
WTP(s)	Water treatment plant(s)

INTRODUCTION

Traditionally, water chlorination has been a preferred disinfection strategy because of its proven efficiency and cost effectiveness in improving microbiological water quality until Rook (1974) and Bellar *et al.* (1974) revealed that chlorine reacts with naturally occurring organic matter in water, particularly humic and fulvic acids, to generate potentially harmful by-products such as trihalomethanes (THMs). The primary THMs of concern in drinking water are chloroform, bromodichloroform, dibromochloroform, and bromoform. Concerns about health risks associated with THMs have prompted environmental agencies in most countries to regulate THMs in drinking water to safeguard consumers from adverse health impacts.

Past investigations have observed that the occurrence of THMs in chlorinated water may vary significantly according to season and geographical location due to changes in raw and treated water quality (fulvic and humic constituents, ammonia), as well as in operational parameters related to chlorination such as chlorine dose, water temperature, pH and travel time (Denne *et al.* 1984; Garcia-Villanova *et al.* 1997; Arora *et al.* 1997; Premazzi *et al.* 1997*a*; Rodriguez *et al.* 2000).

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In Lebanon, chlorination is the sole disinfection strategy adopted by public water treatment utilities. Until the initiation of this study, there was a lack of information concerning THM occurrence in drinking water distribution systems in Lebanon, as well as about utility capacity for compliance with proposed THM standards. The aim of this paper is to report the spatial and temporal evolution of THMs in chlorinated waters from various water utilities in Lebanon, as well as to investigate the impact of water source type and treatment schemes on THM formation.

MATERIALS AND METHODS

Water sources and sampling locations

Water samples were collected randomly from 42 water sources. Three sampling points (raw water, directly after chlorination (DAC), and/or distribution network) were selected for each sampling location to track the water from its source to the distribution network. General characteristics of surveyed water sources, sampling locations, and sampling points are summarized in Table 1.

Sampling strategy and laboratory analysis

A seasonal sampling program was initiated whereby duplicate samples were properly collected and preserved, then carried to the Environmental Engineering Research Center (EERC) at the American University of Beirut (AUB) for further analysis. At the laboratory, THM concentrations were determined in accordance with USEPA method 551.1 (USEPA 1995).

RESULTS AND DISCUSSION

Spatial variations in TTHM levels

Varying concentrations of individual and total trihalomethanes (TTHMs) were detected in the surveyed sampling locations. It was of significance to study THM speciation since the presence of brominated THMs in drinking water pose more adverse health impacts because of their higher potential for mutagenicity as revealed in previous research (Nokes *et al.* 1999; Nobukawa & Sanukida 2001; Stuart *et al.* 2001). Recorded average TTHM concentrations ranged between 0–9.95 μg/L for raw water, 0–18.39 μg/L for samples collected DAC, and 0.35–28.48 μg/L for samples collected from within or at the end of the water distribution networks. As for the percentage occurrence of individual THM species, mean values amounted to 14%TCM, 17%BDCM, 25% CDBM, and 44% TBM during spring, 19%TCM, 9%BDCM, 25% CDBM, and 47% TBM during summer, and 9%TCM, 16%BDCM, 27% CDBM, and 48% TBM during winter. It was noted that the majority of the samples collected from points within the distribution network, and thus at a distance from the chlorination point, exhibited higher TTHM concentrations when compared to TTHM levels detected in samples collected DAC. This phenomenon is in agreement with the findings of various researchers (Premazzi *et al.* 1997*b*; LeBel *et al.* 1997; Williams *et al.* 1998).

Recorded incremental increase of mean TTHM levels were calculated to range between 4–27 folds during spring, 2–15 folds during summer, and 2–13 folds during winter when comparing samples collected DAC to raw water samples. As for the incremental increases of mean TTHM levels when comparing samples collected from the distribution network to samples collected DAC, the computed ranges were 1–13 folds, 1–6 folds, and 1–11 folds, for the three consecutive seasons, respectively. Recorded percent incremental increases for each of the sampled locations in the specified sampling season are summarized in Table 2. The evolution patterns of TTHM concentrations from raw water to the distribution networks are graphically presented in Figures 1 to 12.

Seasonal variations in TTHM levels

Varying seasonal trends in TTHM levels were revealed from the investigations. When comparing mean TTHM concentrations recorded in the summer for samples collected DAC to the values recorded in spring for the same samples, their ratios ranged between 0.19–2.02 (mean = 0.88). For the same category of samples, computed ratios for summer to winter ranged between 0.53–4.03 (mean = 1.22), and between 0.58–5.71 (mean = 1.91) for spring to winter. As for samples collected from the distribution network, summer to spring TTHM ratios ranged between 0.04–2.52 (mean = 0.73), 0.07–1.97 (mean = 0.88) for summer to winter, and 0.64–4.41 (mean = 1.57) for spring to winter.

Table 1 | General characteristics of surveyed sampling locations

		Surveyed sampling point		
Sampling location	**Adopted treatment scheme**	**Raw**	**DAC**	**Network**
Dbayye WTP	S + C + F + Cl	✓	✓	✓
Naameh tank	Cl	✓	✓	✓
Jamhour station	Cl	× /✓	✓	✓
Hazmieh-Brazilia WTP	Sd + F + Cl	✓	✓	✓
Zheyma	Cl	✓	✓	✓
Al Rayyes, Shoueifat	Cl	✓	✓	✓
Al-Rishani, Shoueifat	Cl	✓	✓	✓
Kanaan, Kfarshima	Cl	✓	✓	✓
Assaily, Kfarshima	Cl	×	✓	✓
Dekwaneh	Cl	✓	✓	✓
Sabtiyeh	Cl	×	✓	✓
Jisr El-Basha	Cl	✓	×	✓
Bonjus, Fanar	Cl	✓	✓	✓
Nahr El-Mot	Cl	×	✓	✓
Der Tamish	Cl	×	✓	✓
Ayntoura	Cl	×	✓	✓
Jeita	Cl	✓	✓	✓
Chananiir	Cl	×	✓	✓
Jbeil WTP	Sd + F + Cl	✓	✓	✓
Kfar Helda WTP	Sd + C + F + Cl	✓	✓	✓
Kousba WTP	Sd + C + F + Cl	✓	✓	✓
Chekka	Cl	×	✓	✓
Anfeh	Cl	× /✓	✓	✓
Fih	Cl	×	✓	✓
Tripoli WTP	Sd + C + F + Cl	✓	✓	✓
Abu Samra, Tripoli	Cl	×	✓	✓
Al-Qebbeh, Tripoli	Cl	×	✓	✓
Al-Jisr, Tripoli	Cl	✓	×	✓

S: screening, Sd: sedimentation; C: coagulation (only in winter in instances of turbid influent water), F: filtration, Cl: chlorination.

When comparing the overall mean ratios of TTHM levels among various seasons and for all chlorinated samples combined, computed ratios amounted to 0.82, 1.05, and 1.74 when comparing summer to spring, summer to winter, and spring to winter, respectively. Therefore, recorded TTHM concentrations are highest during spring, followed by summer during which TTHM values were only slightly higher than those encountered during winter. The varying seasonal trend in TTHM levels exhibited in this research may be attributed to the following environmental or operational factors:

- In Lebanon, no significant seasonal water temperature variation exists between spring (20.0°C) and summer (24.7°C), especially in coastal or low-elevation regions. As for the winter sampling season, the mean water temperature recorded on-site in the distribution network was 16.1°C which may explain the slight decrease in TTHM levels for the samples collected in the winter season.
- Mean water temperatures recorded on-site in the distribution networks in spring and summer were higher than the critical temperature (≈19°C) specified by Garcia-Villanova *et al.* (1997) above which THM levels reduce drastically due to its increased volatility.
- The impact of temperature on THM formation is reported to be more significant at longer contact times (WHO 2000). In Lebanon, water distribution systems related to a particular water source are relatively short, except for urban areas that rely on water treatment plants. Consequently, contact times are relatively short and do not accentuate the impact of temperature on TTHM formation.
- Generally in Lebanon, chlorination is practiced erratically, specifically at wells serving confined areas. Such a practice causes hourly, daily, as well as seasonal variations in the concentrations of residual chlorine in the distribution networks which renders correlations difficult.
- A raw water characteristic that may explain the higher TTHM levels in spring samples is the higher mean natural organic matter (NOM) level during this season (TOC = 5.92 mg C/L) compared to summer (TOC = 3.94 mg C/L) and winter (TOC = 3.89 mg C/L).

Impact of water source type on THM formation

To probe the effect of investigated water source types on THM formation, TTHM levels recorded in surface water sources were compared to the concentrations encountered in ground water sources. As noted from Figure 13, sampling locations relying on surface waters as abstraction sources revealed higher mean TTHM concentrations during spring and summer compared to groundwater sources when sampled DAC. Winter mean TTHM values for the same samples were almost comparable for the two water source types. Regarding mean TTHM levels in the distribution networks, also surface waters exhibited higher levels for the three sampling seasons (Figure 14). Moreover, it can be concluded from the overall mean TTHM levels for the three sampling seasons that TTHM concentrations were higher when the abstraction sources were surface waters compared to ground water sources. For samples collected directly after chlorination, mean TTHM levels were 2.2 folds higher for surface water sources compared to ground water sources; while for samples collected from the distribution networks, mean TTHM levels were 1.6 folds higher for surface water sources compared to ground water sources. The higher mean TTHM values in chlorinated waters originating from surface water sources may be attributed to the higher TOC levels in the raw filtered surface waters since ground waters are naturally protected from runoff NOM, their higher temperatures, and the longer residence time in the distribution networks.

Impact of adopted treatment schemes on THM formation

To investigate the impact of adopted treatment schemes on THM formation, TTHM levels recorded at locations subjected to semi-conventional (screening, sedimentation, and filtration) water treatment, namely water treatment plants (WTPs) abstracting water from surface water sources, were compared to the concentrations encountered at locations subjected to direct chlorination, namely on-site wells. It can be noted from Figures 15 and 16 that water sources subjected to semi-conventional water treatment schemes revealed higher mean TTHM concentrations during spring and summer compared to water sources where direct chlorination was applied. Winter mean TTHM values for the same samples were almost comparable for the two treatment strategies. Moreover, it can be concluded from the overall mean TTHM levels for the three sampling

Table 2 | Recorded percent incremental increases in TTHM levels for the surveyed distribution networks

	Spring	Summer	Winter		Spring	Summer	Winter
Sampling location	**Network to DAC**	**Network to DAC**	**Network to DAC**	**Sampling location**	**Network to DAC**	**Network to DAC**	**Network to DAC**
Dbayye WTP	118.2	162.1	ND[a]	Jbeil WTP	133.1	279.5	ND
Naameh chlorination tank	324.2	162.2	ND	Kfarhelda WTP	173.0	101.0	456.0
Jamhour station	197.8	102.1	175.7		210.0	ND	346.0
Hazmieh-Brazilia WTP	264.0	154.8	ND		152.0	ND	108.0
	160.0	ND	ND		273.0	ND	ND
Zheyma	207.6	199.9	582.9	Kousba WTP	173.4	ND	148.9
Al Rayyes, Shoueifat	ND	347.7	ND	Chekka	473.6	160.1	ND
Al-Rishani, Shoueifat	182.6	160.4	ND	Anfeh	ND	ND	432.8
Kanaan, Kfarshima	ND	112.8	116.5	Fih	357.2	ND	364.8
Assaily, Kfarshima	ND	ND	241.6	Tripoli WTP	132.4	148.4	ND
Dekwaneh	121.6	106.2	116.4	Abu Samra, Tripoli	552.9	445.8	ND
Sabtiyeh	219.4	358.3	ND	Al-Qebbeh, Tripoli	421.0	549.9	ND
Jisr El-Basha	ND	129.2	1067.0		261.0	ND	ND
Bonjus, Fanar	200.6	333.8	148.7		1304.0	ND	ND
Nahr El-Mot	110.8	216.4	118.2	Al-Jisr, Tripoli	ND	ND	ND
Der Tamish	177.7	309.9	ND	Min[b]	110.8	101.0	108.0
Ayntoura	320.7	ND	ND	Max[b]	1304.0	549.9	1067.0
Jeita	197.1	ND	ND	Mean[b]	304.0	227.0	316.0
Chananiir	1093.8	ND	ND	Median[b]	204.1	162.1	208.6

[a]ND: Not determined, either due to inaccessibility to raw water sampling point or not sampling that particular point during the specified sampling season.
[b]Ratios <1.0 were not considered in these calculations.

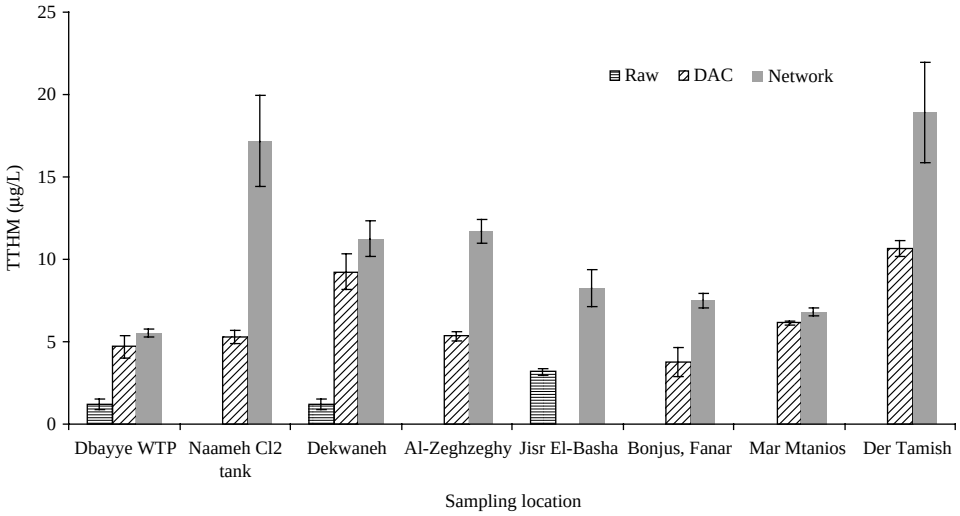

Figure 1 | Evolution of TTHM levels for sampling locations affiliated to Beirut Water Board (Spring 03).

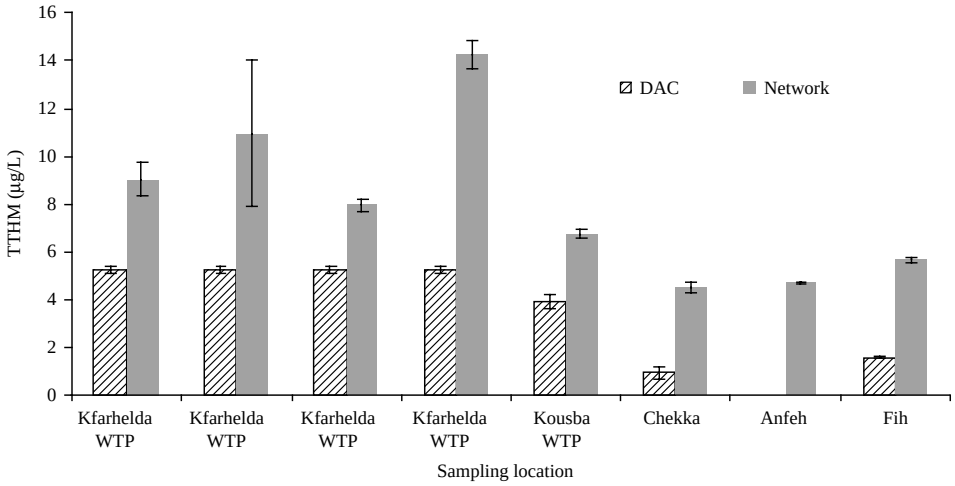

Figure 2 | Evolution of TTHM levels for sampling locations in Batroun and Koura Cazas (Spring 03).

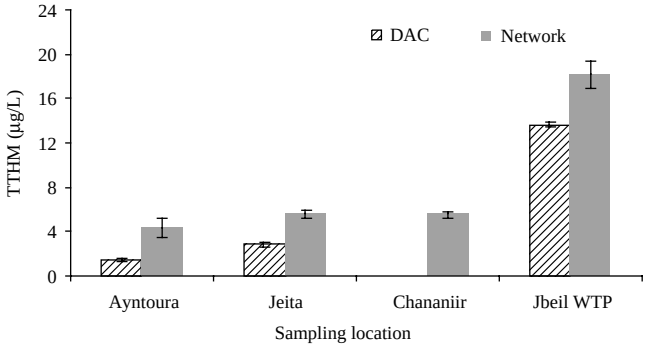

Figure 3 | Evolution of TTHM levels for sampling locations in Keserwan and Jbeil Cazas (Spring 03).

seasons that TTHM concentrations were higher when the raw water was subjected to a semi-conventional treatment scheme compared to locations where direct chlorination was employed. For samples collected DAC, mean TTHM levels were 2.3 folds higher for semi-conventionally treated waters compared to solely chlorinated waters; while for samples collected from the distribution networks, mean TTHM levels were 1.6 folds higher for semi-conventionally treated waters compared to solely chlorinated waters.

Typically and as reported in previous literature, the extent of THM formation is reduced in source waters subjected to conventional water treatment prior to chlori-

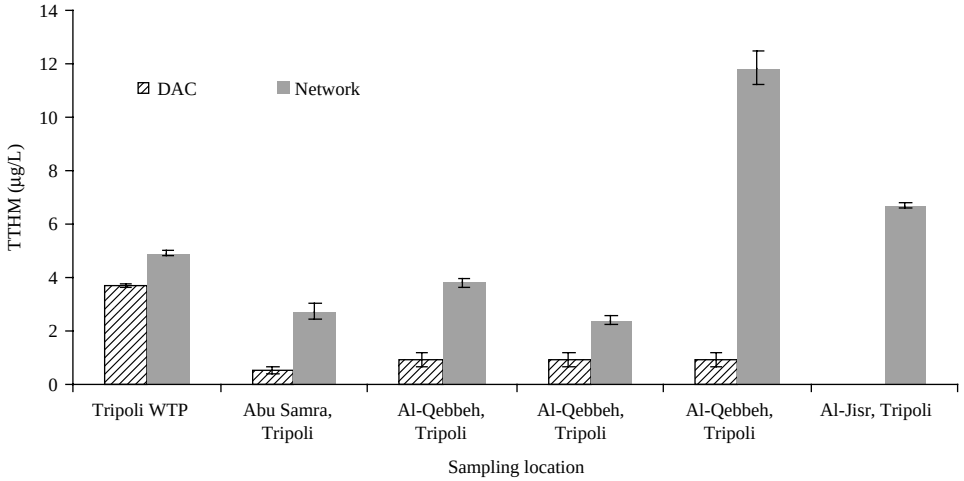

Figure 4 | Evolution of TTHM levels for sampling locations in Tripoli Caza (Spring 03).

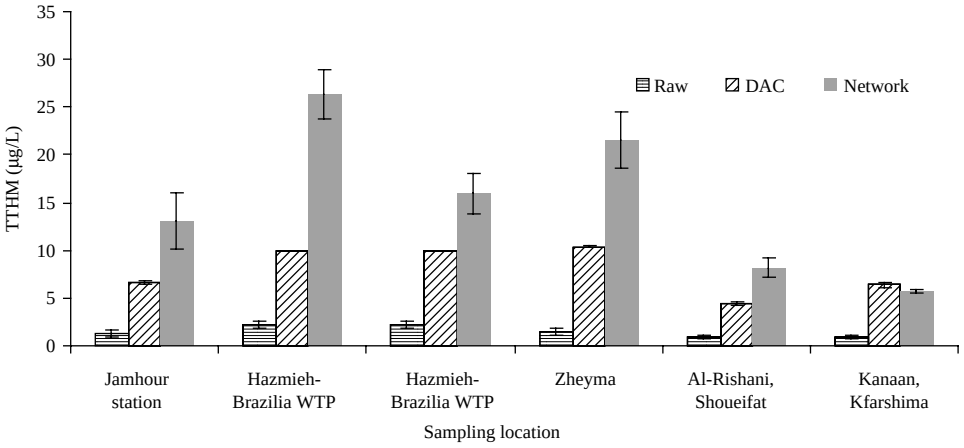

Figure 5 | Evolution of TTHM levels for sampling locations affiliated to Ain El Delbe Water Board (Spring 03).

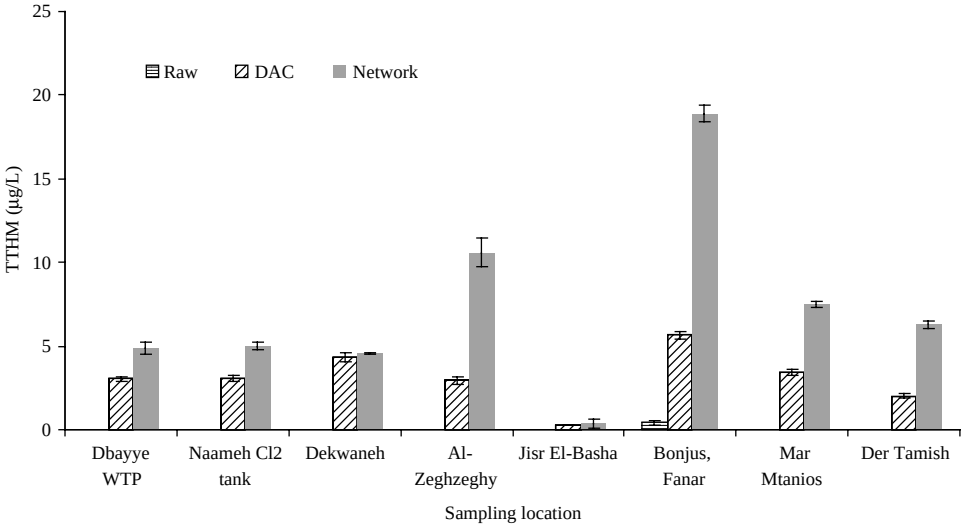

Figure 6 | Evolution of TTHM levels for sampling locations affiliated to Beirut Water Board (Summer 03).

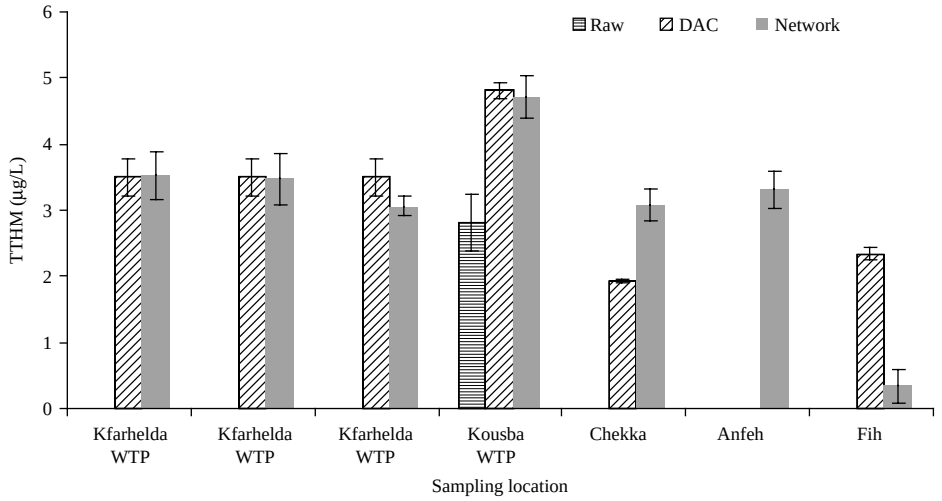

Figure 7 | Evolution of TTHM levels for sampling locations in Batroun and Koura Cazas (Summer 03).

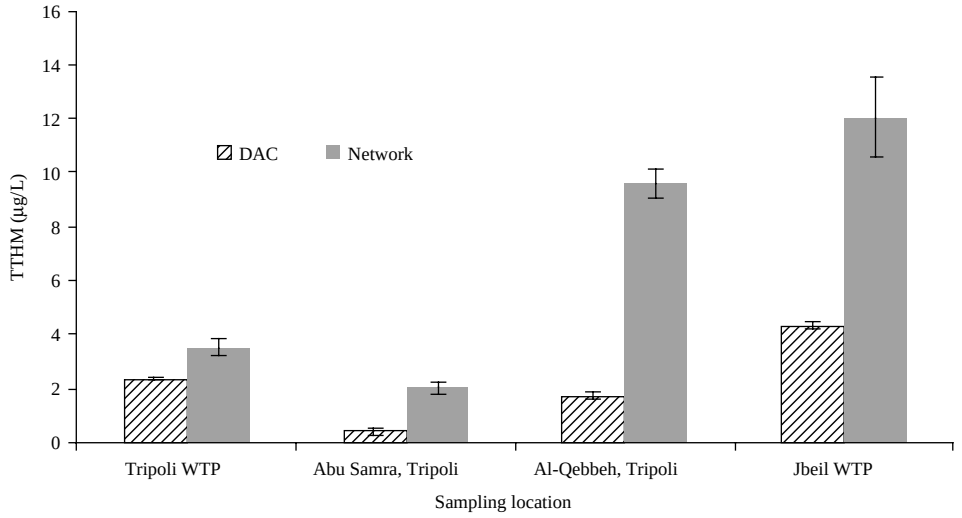

Figure 8 | Evolution of TTHM levels for sampling locations in Tripoli and Jbeil Cazas (Summer 03).

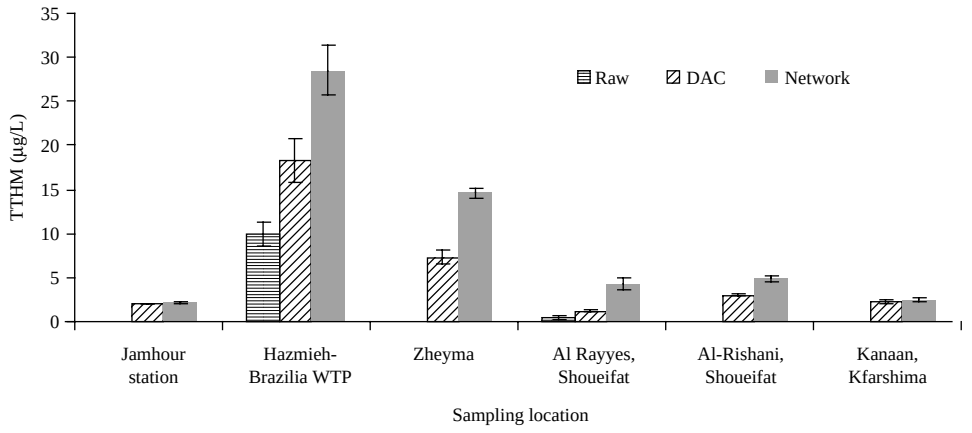

Figure 9 | Evolution of TTHM levels for sampling locations affiliated to Ain El Delbe Water Board (Summer 03).

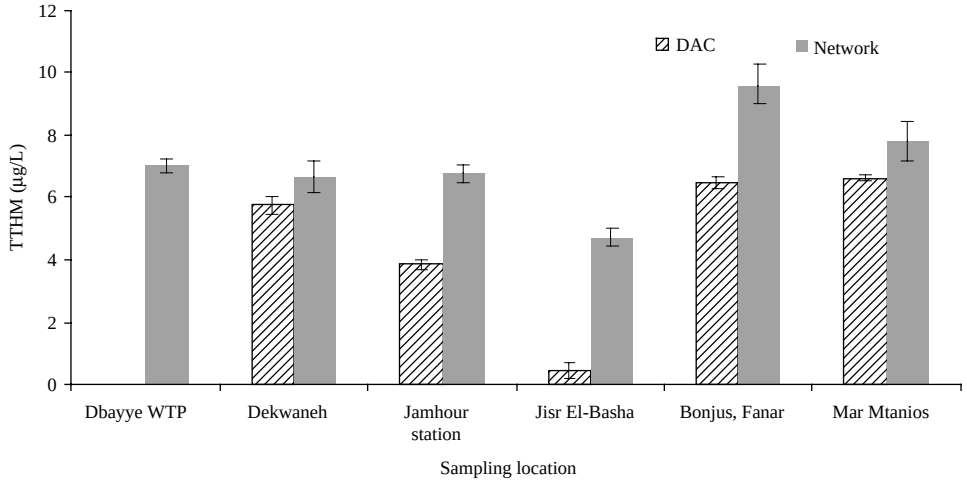

Figure 10 | Evolution of TTHM levels for sampling locations affiliated to Beirut Water Board (Winter 04).

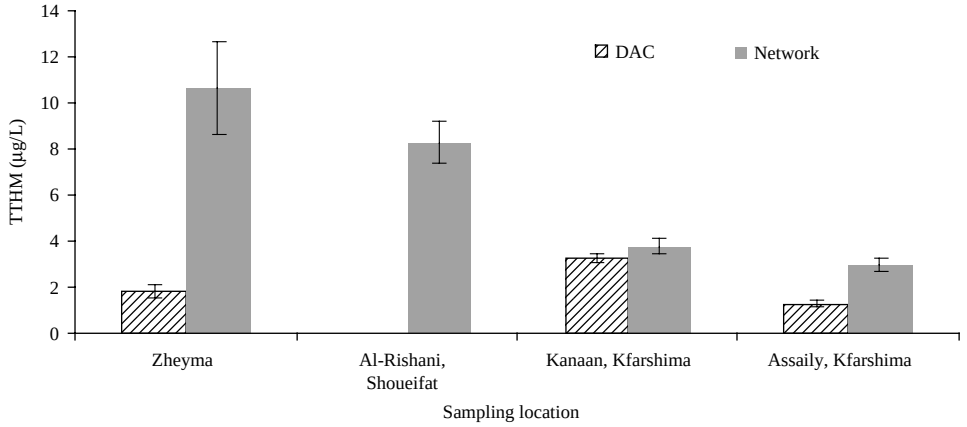

Figure 11 | Evolution of TTHM levels for sampling locations affiliated to Ain ElDelbe Water Board (Winter 04).

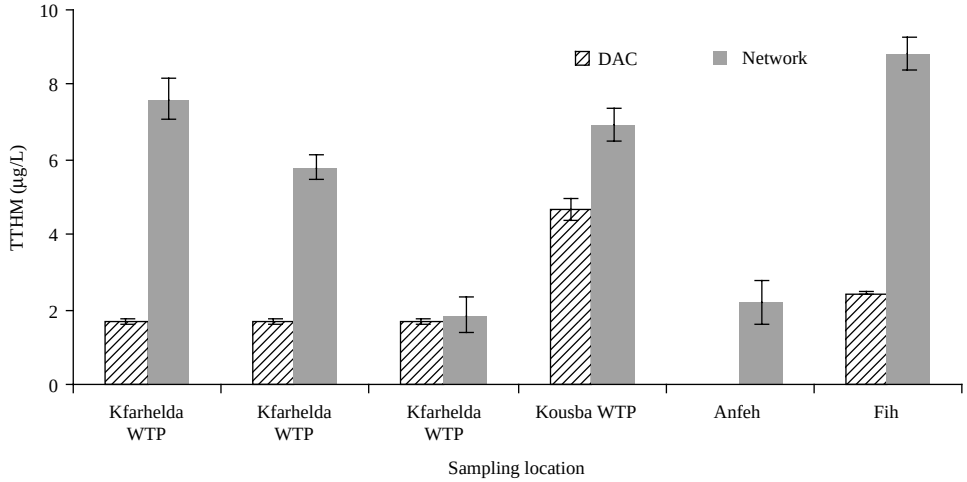

Figure 12 | Evolution of TTHM levels for sampling locations in Batroun and Koura Cazas (Winter 04).

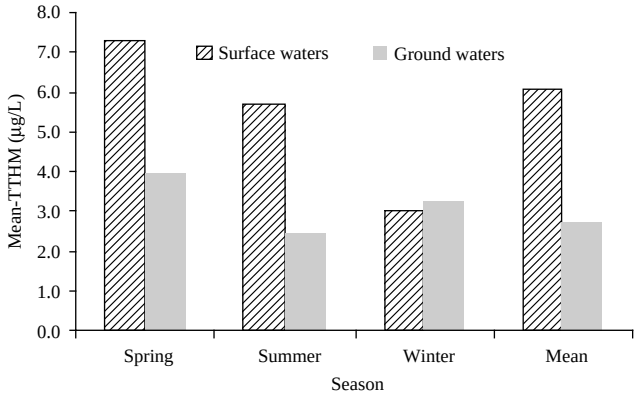

Figure 13 | Mean TTHM concentrations for samples collected directly after chlorination by source water type.

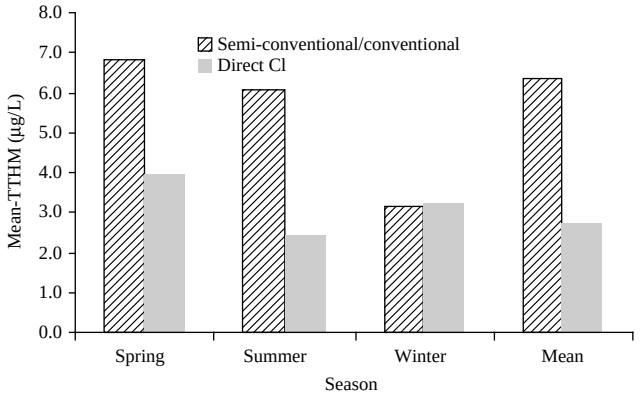

Figure 15 | Mean TTHM concentrations for samples collected directly after chlorination by type of adopted treatment scheme.

nation, i.e. screening, sedimentation, coagulation, and filtration, since enhanced coagulation is a key THM precursor removal technique (USEPA 1999; Bolto *et al.* 2002). On the other hand, direct chlorination of waters without any prior precursor removal attempts enhances THM formation. However, for the investigated sources and treatment techniques outcomes revealed a contrary situation. This may be attributed to the absence of coagulation from the adopted treatment process at WTPs, except occasionally when very high turbidity levels were encountered in the source water, typically during winter. This justification can be further confirmed by the slightly lower mean TTHM levels recorded for conventionally treated source waters in the winter season during which coagulation is practiced at WTPs. In fact, enhanced coagulation is the key NOM removal technique in water treatment, and thus its absence renders treatment ineffective in terms of THM precursor removal. Other contributing observations may be the higher TOC levels in the raw filtered source waters, the higher temperatures, and longer residence times in the distribution networks.

CONCLUSIONS

In conformity with findings of previous researchers, the majority of samples collected from points within the distribution network, and thus at a distance from the chlorination point, exhibited higher THM concentrations (1–13 folds) when compared to TTHM levels detected in samples collected directly after chlorination. Overall mean ratios of THM levels among various seasons revealed that TTHM concentrations in Lebanon are highest in the spring, followed by the summer during which THM values are only

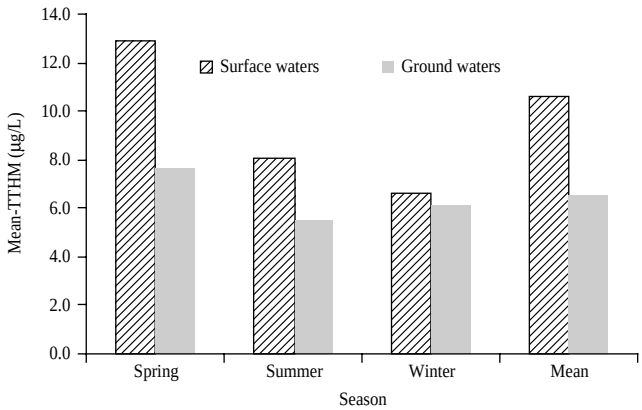

Figure 14 | Mean TTHM concentrations for samples collected from networks by source water type.

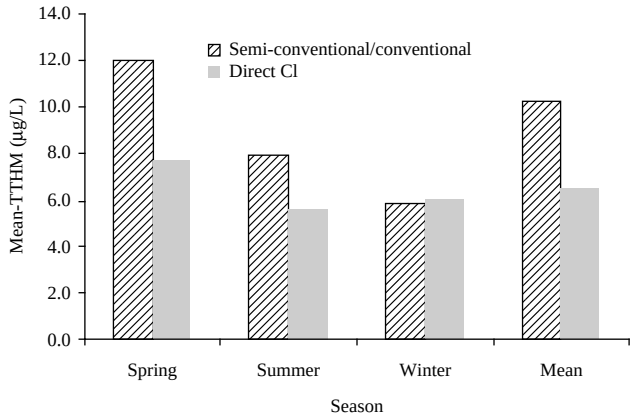

Figure 16 | Mean TTHM concentrations for samples collected from the distribution network by type of adopted treatment scheme.

slightly higher than those encountered in the winter season. Furthermore, overall mean TTHM concentrations in various seasons were higher when the abstraction sources were surface waters compared to ground water sources. Also, higher mean THM values were recorded in chlorinated waters originating from sources subjected to semi-conventional water treatment before the application of chlorine compared to mean THM values established in chlorinated waters originating from sources subjected to direct chlorination. This was attributed to the absence of coagulation from the adopted semi-conventional treatment scheme at WTPs, and possibly outcomes might have been different in the presence of coagulation.

On the basis of outcomes, WTP operators may pinpoint the water sources and networks at a relatively higher risk for THM formation and thus may propose the proper preventive (source water pollution prevention, removal of THM precursors, maintenance of water distribution networks) and curative approaches for THM control.

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