

FORMATION OF DBPs IN THE DRINKING WATER OF ATHENS, GREECE: A TEN-YEAR STUDY

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Received: 10/01/05
Accepted: 30/03/05

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ABSTRACT

The formation of Disinfection By-Products (DBPs) in drinking water results from the reaction of chlorine or other disinfectants added to the water with naturally occurring organic materials, and has raised concerns during the last decades because these compounds are harmful for human health. During the present work, the formation of different categories of DBPs was investigated in four water treatment plants (WTP) using chlorine as disinfectant, and in selected points of the distribution network of Athens, Greece, which is supplied from these four WTP, during a period of ten years. The concentrations of DBPs were generally low and the annual mean concentrations always well below the regulatory limit of the European Union (EU) for the total trihalomethanes (TTHMs). The haloacetic acids (HAAs) have not been regulated in the EU, but during this investigation they often occurred in significant levels, sometimes exceeding the levels of TTHMs, which highlights the importance of their monitoring in drinking water. Apart from THMs and HAAs, several other DBPs species were detected at much lower concentrations in the chlorinated waters: chloral hydrate, haloketones and, in a limited number of cases, haloacetoneitriles.

KEYWORDS: chlorination, trihalomethanes, haloacetic acids, chloral hydrate, haloketones

INTRODUCTION

The occurrence of Disinfection By-Products (DBPs) in drinking water has become an issue of great scientific concern during the last decades, due to the adverse effects of these compounds for human health. DBPs have been detected in drinking waters all over the world, and are formed during treatment with chlorine and other disinfectants, necessary for the good microbiological quality of the drinking water. Several hundreds of different species of DBPs have been identified up-to-date and continue being discovered (Richardson *et al.*, 2002), as more reliable and precise analytical techniques are becoming available, which allow their determination at trace levels.

The first and most widely studied group of DBPs was trihalomethanes (THMs) (Rook, 1974), followed by haloacetic acids (HAAs), however a large number of other compounds may also be formed in disinfected water, which have been less extensively studied, and with health effects which remain largely unknown. Other categories of DBPs frequently

detected in drinking water, apart from THMs and HAAs, include haloacetonitriles (HANs), haloketones (HKs), chloropicrin (CP) and chloral hydrate (CH) (Christman *et al.*, 1983; Miller and Uden, 1983; Oliver, 1983; Krasner *et al.*, 1989; Nieminski *et al.*, 1993; Summers *et al.*, 1996; LeBel *et al.*, 1997; Williams *et al.*, 1997; Golfopoulos *et al.*, 1998; Simpson and Hayes, 1998; Nikolaou *et al.*, 1999; Golfopoulos and Nikolaou, 2001; Nikolaou *et al.*, 2002), as well as MX, NDMA and related compounds (Andrzejewski and Nawrocki, 2005) which also have adverse health effects (Reckhow and Singer, 1985; Bull and Robinson, 1986; Pereira *et al.*, 1986; Bull and Kopfler, 1991; Richardson, 2005).

Taking into account the increasing evidence available regarding their adverse health effects, the maximum contaminant level (MCL) of $100 \mu\text{g l}^{-1}$ for the concentration of total THMs in drinking water set by USEPA (1979) was later lowered to $80 \mu\text{g l}^{-1}$ (EPA, 1998). The sum of concentrations of five HAAs (monochloro-, dichloro-, trichloro-, monobromo- and dibromoacetic acid) has also been regulated by USEPA to $60 \mu\text{g l}^{-1}$. The European Union has regulated the concentration of total THMs to $150 \mu\text{g l}^{-1}$ until 2008 and to $100 \mu\text{g l}^{-1}$ after 2008 (EEC, 1998), but has not yet established any regulatory limit for HAAs or other DBPs. Further data regarding both the concentration levels and adverse effects of the DBPs other than THMs in treated drinking waters is required in order to better define the health risks associated with their consumption and to safeguard their quality by optimization and/or modifications in the treatment process.

The objective of the present work was to obtain information on the occurrence of the most commonly encountered different categories of DBPs in the drinking water of Athens, Greece. THMs, HAAs, HANs, HKs, CP and CH were investigated in samples collected from four major treatment plants supplying drinking water to Athens, the capital of Greece, and in representative samples from the distribution network of the city. Sampling and analytical determinations were performed monthly, for a period of ten years. Apart from THMs, which were the most significant category of DBPs detected, along with HAAs, which in some cases predominated, the systematic occurrence of CH and the HKs 1,1-dichloropropanone (1,1-DCP) and 1,1,1-trichloropropanone (1,1,1-TCP) in almost all chlorinated samples, and the presence of some species of HANs and CP in a limited number of cases were observed. These findings highlight the need of more detailed investigation of the speciation of DBPs mixtures in drinking water for a more complete evaluation of its quality.

MATERIALS AND METHODS

Water treatment plants

The four Water Treatment Plants (WTP) of the Athens Water Supply and Sewerage Company (EYDAP SA), which supply drinking water to approximately 4,000,000 inhabitants in the area of Attiki, Greece, were investigated in this study for the formation of DBPs. These WTP (Galatsi, Menidi, Aspropyrgos and Polydendri) receive water from lakes Mornos, Marathon and Iliki. Their design is conventional and includes coagulation, flocculation, sedimentation and filtration. The disinfectant used is chlorine. Prechlorination is applied, with chlorine dose $1.2\text{--}2.5 \text{ mg l}^{-1}$, and postchlorination, with 6.5 mg l^{-1} . During prechlorination, chlorine is added to the raw water at the entrance of each WTP along with the coagulant (aluminium sulphate), i.e. before coagulation and sedimentation. The postchlorination in Galatsi WTP is applied after sedimentation and before filtration, while in the other WTP (Menidi, Aspropyrgos and Polydendri) it is applied after filtration.

The flowcharts of Galatsi and Menidi WTP are presented in Figures 1 and 2 respectively, including the chlorination, postchlorination and sampling points, from where samples have been analyzed for DBPs on a monthly basis during the last ten years. The water samples were collected from the raw water, the sedimentation tanks and the finished

water reservoirs of the WTP. Samples were also collected from representative points of the distribution network of Athens city, to evaluate the levels of DBPs in the treated water reaching the consumers' tap. The sampling network and the abbreviations of the sampling points are presented in Table 1.

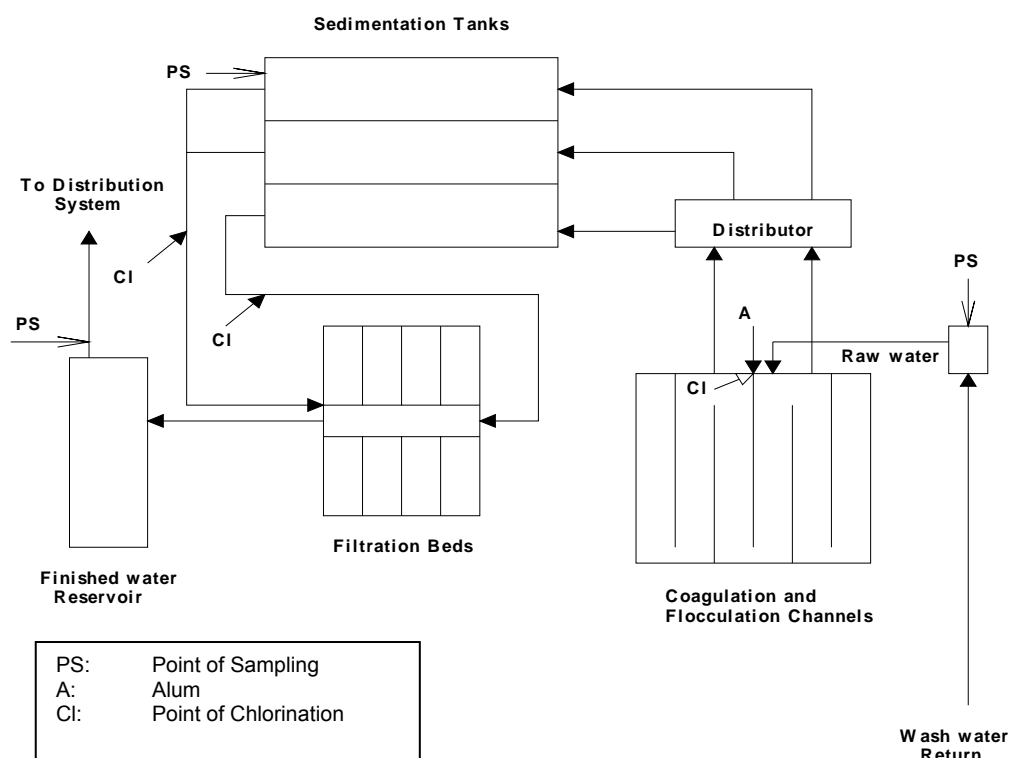


Figure 1. Flowchart of Galatsi WTP

Sampling

Samples were collected in 40-mL amber glass bottles with polypropylene screw caps and TFE-faced septa (Pierce 13075), which were carefully filled so that trapping of air bubbles inside was prevented. Quenching was performed by addition of sodium sulfite for THMs and other DBPs, and ammonium chloride for HAAs samples (100 mg per liter of sample). The samples were kept in the dark at 4 °C until analysis and were analyzed within one week.

Analytical procedure

Glassware. Before use, all glassware was washed with detergent, rinsed with tap water, ultrapure water (Millipore: Milli-Ro 5 plus and Milli Q plus 185), acetone (Mallinckrodt Chemical Works St. Louis) and oven-dried at 150 °C for 2 hours.

Reagents-standard solutions. Methanol purge and trap grade was purchased from Sigma-Aldrich, methyl-tert-butyl ether (MTBE) suprasolv grade, sodium sulfite, ammonium chloride, sodium sulfate, copper (II) sulfate pentahydrate and sulfuric acid concentrated ISO for analysis from Merck. Ultrapure water was from Milli-Q water purification system (Millipore: Milli-Ro 5 plus and Milli Q plus 185). Certified commercial mix solutions (purity>99%) of THMs were from Chemservice and of HAAs from Supelco. Stock solutions of other DBPs were prepared in methanol purge and trap grade by addition of appropriate amounts of monochloroacetonitrile, dichloroacetonitrile, trichloroacetonitrile, bromochloroacetonitrile, dibromoacetonitrile, chloral hydrate, 1,1-dichloropropanone, 1,3-dichloropropanone, 1,1,1-trichloropropanone and chloropicrin.

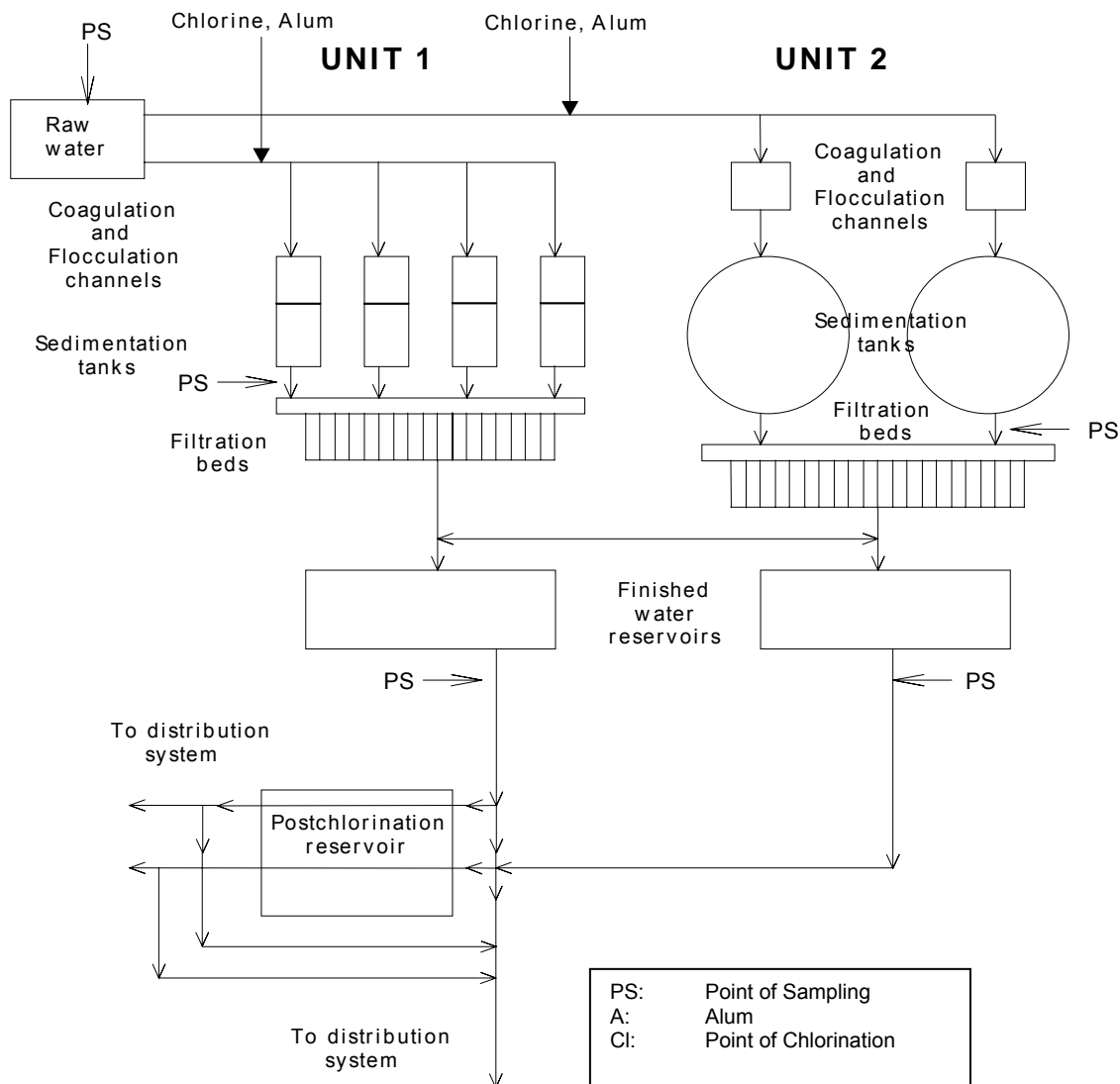


Figure 2. Flowchart of Menidi WTP

Sample preparation. For THMs and other volatile DBPs, a modification of EPA Method 551.1, which includes liquid - liquid extraction (LLE) with MTBE was performed (EPA, 1998b), as described in previous work (Nikolaou *et al.*, 2002b; Goulinopoulos and Nikolaou, 2001). For HAAs, LLE with MTBE followed by acidic methanol esterification (Cancho *et al.*, 1999) was applied, as previously described (Nikolaou *et al.*, 2002c).

Apparatus. A HP 5890 Series II Gas Chromatograph equipped with a ^{63}Ni Electron Capture Detector (ECD), with a fused silica capillary DB-1 column 30 m x 0.32 mm i.d. x 0.25 μm film thickness was used for the analysis of DBPs. Injections were made in splitless mode, with helium as carrier gas and nitrogen as makeup gas. The analytical conditions, as well as validation data of the analytical methods have been presented in previous work (Nikolaou *et al.*, 2002b,c). The detection limits of the studied DBPs and their abbreviations are shown in Table 2.

Table 1. The sampling network for the DBPs monitoring

Sampling point	Abbreviation	Sampling point	Abbreviation
Galatsi WTP		Polydendri WTP	
Raw	GR	Raw	PR
Coagulation channel	G1	Sedimentation tank	P1
Sedimentation tank	G2	Storage tank	P2
Sand filters	G3	Aspropyrgos WTP	
Storage tank	G4	Raw	AR
Menidi WTP		Sedimentation tank	A1
Raw	MR	Storage tank	A2
Coagulation channel – Old Unit	M1	Distribution network	
Sedimentation tank – Old Unit	M2	Attiki square	D1
Storage tank – Old Unit	M3	Pireas	D2
Sand filters – Old Unit	M4	Menidi	D3
Coagulation channel – New Unit	M5	Perama	D4
Sedimentation tank – New Unit	M6	Voula	D5
Storage tank – New Unit	M7	Sourmena	D6
Sand filters – New Unit	M8	Varibobi	D7
Postchlorination tank	M9	Fokea	D8
		Salamina	D9

Table 2. Abbreviations and detection limits of the DBPs studied

DBPs	Abbreviation	Detection limit ($\mu\text{g l}^{-1}$)
<i>THMs</i>		
Chloroform	CM	0.010
Dichlorobromomethane	DCBM	0.005
Dibromochloromethane	DBCM	0.007
Bromoform	BM	0.010
<i>Other DBPs</i>		
Monochloroacetonitrile	MCAN	0.040
Trichloroacetonitrile	TCAN	0.070
Dichloroacetonitrile	DCAN	0.007
Chloral hydrate	CH	0.007
1,1-Dichloropropanone	1,1-DCP	0.040
Monobromoacetonitrile	MBAN	0.040
1,1,1-Trichloropropanone	1,1,1-TCP	0.040
1,3-Dichloropropanone	1,3-DCP	0.070
Dibromoacetonitrile	DBAN	0.070
Chloropicrin	CP	0.040
Bromochloroacetonitrile	BCAN	0.040
<i>HAAs</i>		
Monochloroacetic acid	MCAA	0.20
Monobromoacetic acid	MBAA	0.05
Dichloroacetic acid	DCAA	0.02
Bromochloroacetic acid	BCAA	0.02
Trichloroacetic acid	TCAA	0.01
Dibromoacetic acid	DBAA	0.02
Bromodichloroacetic acid	BDCA	0.10
Dibromochloroacetic acid	DBCA	0.20
Tribromoacetic acid	TBAA	0.20

RESULTS AND DISCUSSION

None of the compounds studied was detected in the raw water samples, but all categories of the DBPs occurred in all chlorinated samples within the WTP and the distribution network during all sampling periods. This fact confirms that all species of DBPs detected during the present investigation have been formed only due to the chlorination process, although for some DBPs also other sources than disinfection have been reported, for instance agriculture or production by organisms (Gribble, 1998; Hoekstra *et al.*, 1998).

The most frequently detected compounds were CM, DCBM, DBCM, CH, 1,1-DCP, 1,1,1-TCP, MCAA, DCAA, TCAA, DBAA and BCAA. Chlorinated species predominated, due to the non-detectable or very low bromide concentration in the raw water. In a low number of samples, and in very low concentrations ($<0.5 \mu\text{g l}^{-1}$), BM, DCAN, TCAN, DBAN, CP, BCAN, 1,3-DCP, MBAA, BDCA, DBCA and TBAA were also detected. MCAN and MBAN were not detected in any sample.

The levels of the DBPs concentrations were generally low. The major categories were THMs and HAAs, in accordance with the literature (Lekkas, 2003). The HAAs have not been regulated in the EU, but during this investigation they often occurred in significant levels, sometimes exceeding the levels of TTHMs, which highlights the importance of their monitoring in drinking water. Also, the other DBPs species detected, such as CH, HKs and, in a limited number of cases, HANs, indicate the necessity of the need of a more complete evaluation of water quality through monitoring of more compounds.

During the ten-year period of the present investigation, a gradual increase of the levels of DBPs was observed, although these levels did not in any case exceed the regulatory limits of EU and EPA. The range of annual average concentrations of the total THMs was $12.6\text{-}33.9 \mu\text{g l}^{-1}$ during the period 1999-2001 and increased to $19.4\text{-}68.2 \mu\text{g l}^{-1}$ during the period 2002-2003. The range of annual average concentrations of the total HAAs was $10.1\text{-}28.4 \mu\text{g l}^{-1}$ during the period 1999-2001 and also increased to $17.8\text{-}72.8 \mu\text{g l}^{-1}$ during the period 2002-2003. The concentrations of total THMs and total HAAs detected during this study are presented in Figures 3 and 4 respectively.

CH, 1,1-DCP and 1,1,1-TCP were detected in much lower concentration levels, but also with long-term increasing trends. During the period 1999-2001, their annual average concentrations ranged from $0.3\text{-}0.8 \mu\text{g l}^{-1}$, $0.4\text{-}3.8 \mu\text{g l}^{-1}$ and $0.1\text{-}0.8 \mu\text{g l}^{-1}$, respectively. The corresponding annual average concentrations for the period 2002-2003 were significantly increased for CH ($2.5\text{-}10.7 \mu\text{g l}^{-1}$), and slightly increased for 1,1-DCP and 1,1,1-TCP ($0.6\text{-}1.6 \mu\text{g l}^{-1}$ and $0.2\text{-}1.3 \mu\text{g l}^{-1}$ respectively). The concentrations of these compounds in the different samples during the period of this monitoring study are presented in Figures 5, 6 and 7.

The THMs concentrations in the sedimentation tank effluents were always lower than those in the finished waters in all WTP, which was expectable due to the longer reaction time in the second case. For the same reason, their concentrations in the samples from distribution network were even higher (Krasner *et al.*, 1989; Stevens *et al.*, 1989; Pourmoghaddas and Stevens, 1995; Carlson and Hardy, 1998; Golfopoulos and Nikolaou, 2001). The behaviour of CH was similar to the THMs, whereas the compounds TCAN and 1,1-DCP were detected in slightly lower concentration levels in the finished water compared to the sedimentation tank effluent, probably due to decomposition (Singer, 1994). The levels of 1,1,1-TCP were similar in the sedimentation tank effluents and in the finished water. The HAAs DCAA, BCAA and TCAA occurred at higher concentrations in finished water compared to the sedimentation tank effluent, while in contrast DBAA, BDCA, DBCA and TBAA in many cases showed decreasing trends with time, suggesting their possible degradation (Heller-Grossman *et al.*, 1993).

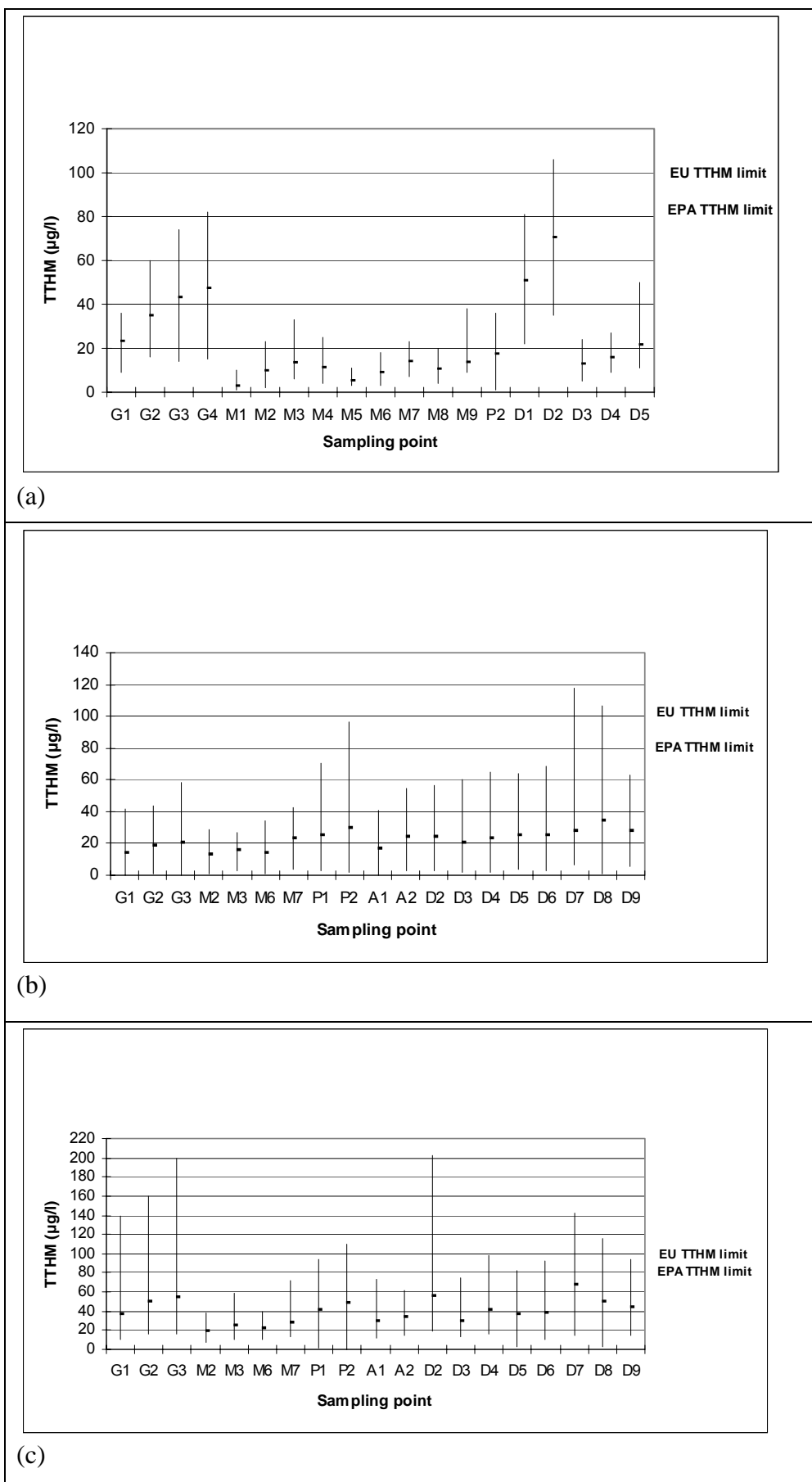
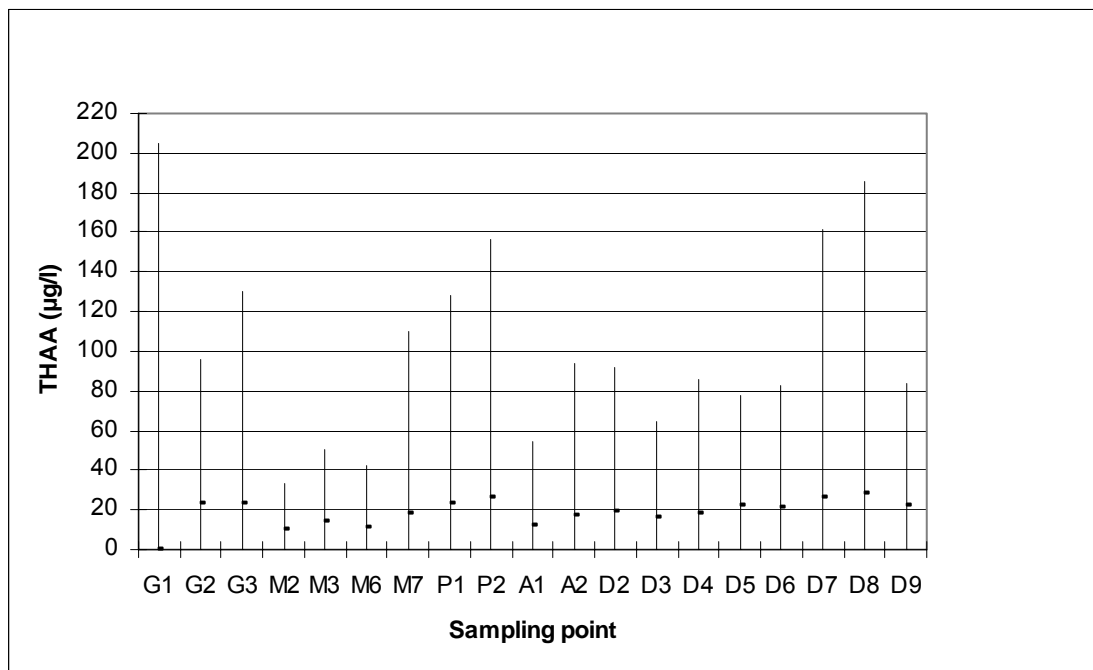
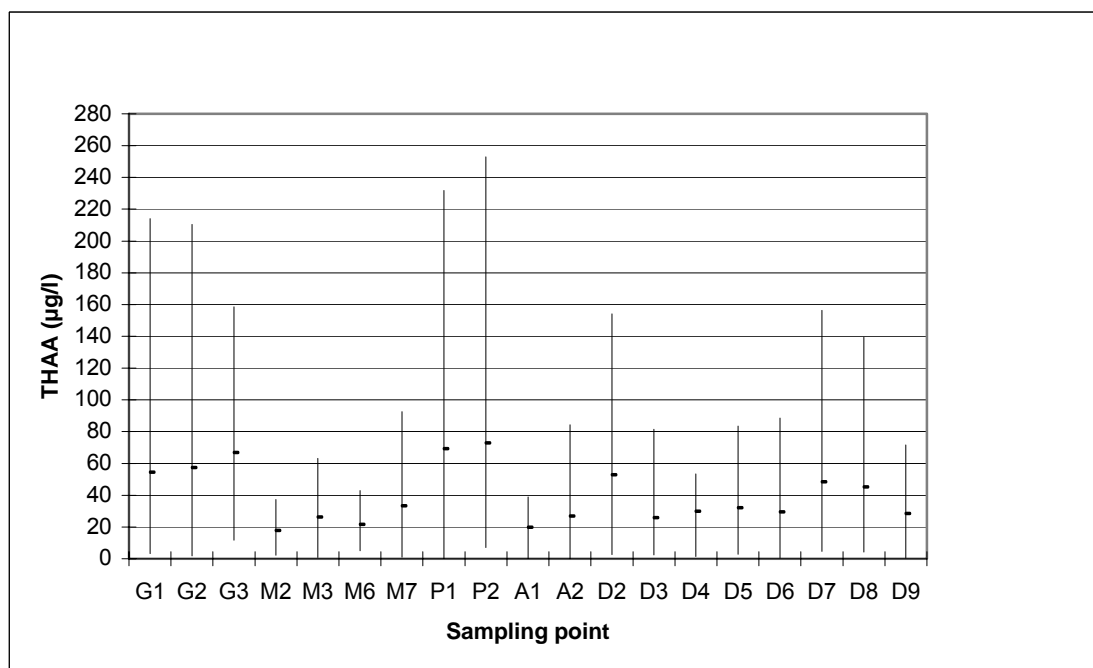


Figure 3. Minimum, mean and maximum TTHMs concentrations in the chlorinated samples during the years (a) 1993-1998, (b) 1999-2001, (c) 2002-2003 (For abbreviations of sampling points see Table 1)



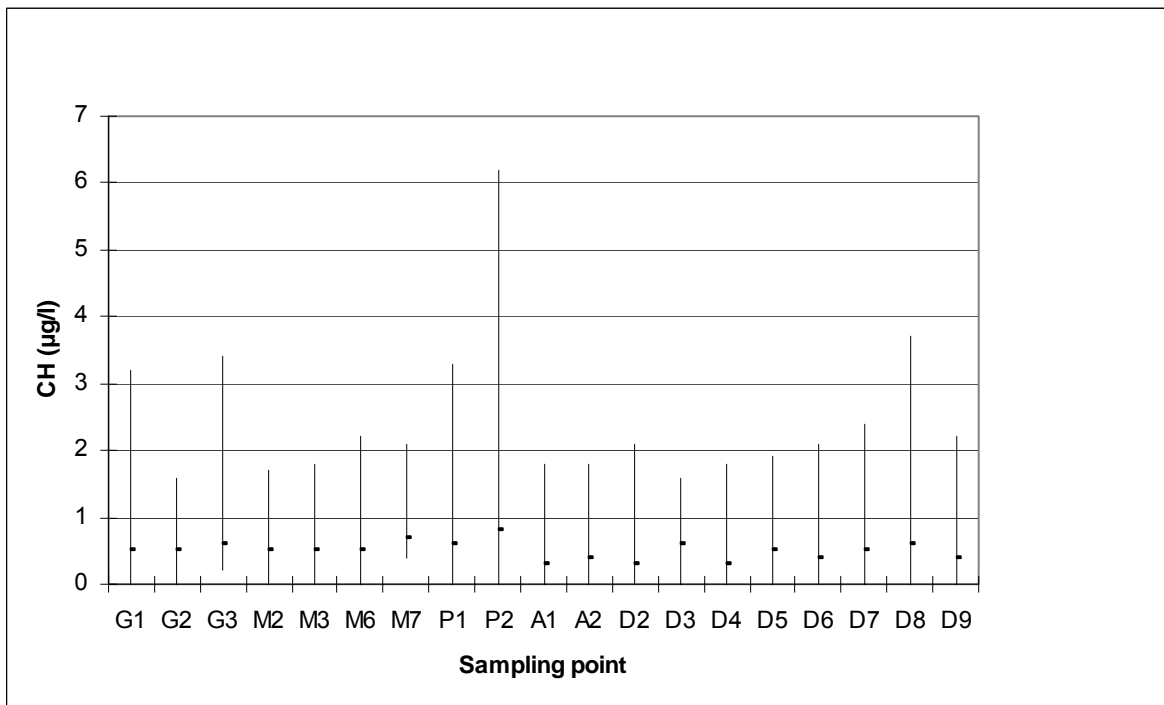
(a)



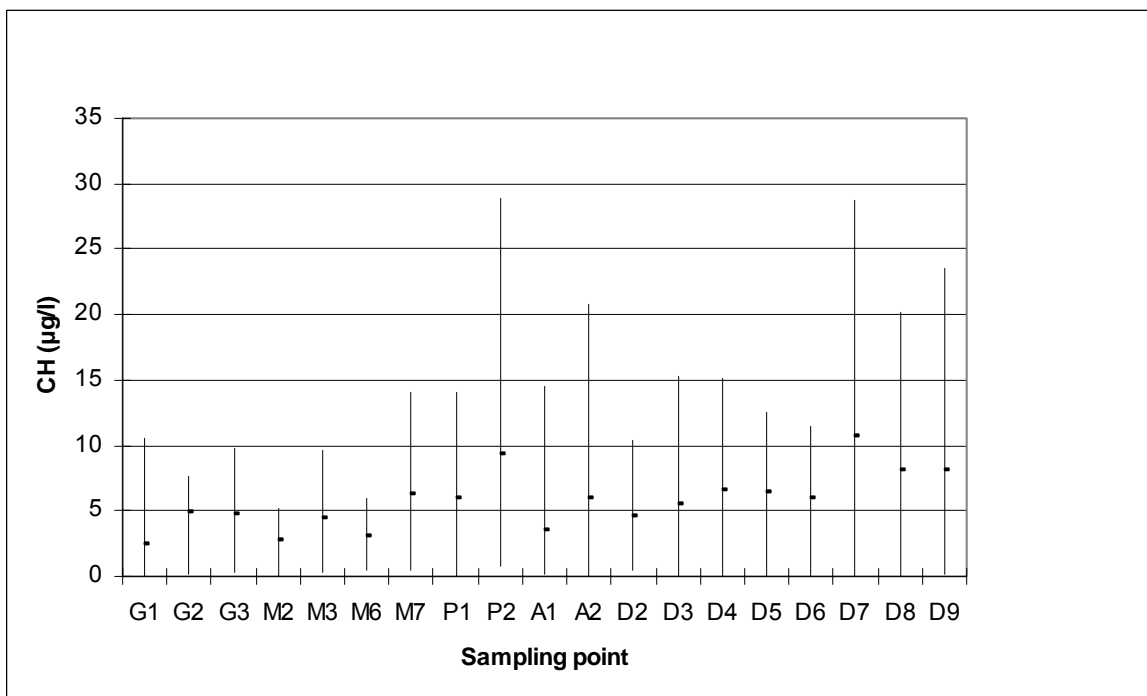
(b)

Figure 4. Minimum, mean and maximum HAAs concentrations in the chlorinated samples during the years (a) 1999-2001, (b) 2002-2003 (For abbreviations of sampling points see Table 1)

In the samples from the distribution network, the concentrations of THMs and HAAs generally ranged in similar or slightly higher levels than within the WTP, because their formation continues as water passes through the distribution network. On the contrary, the HKs and HANs in most cases showed decreasing trends due to decomposition with time.

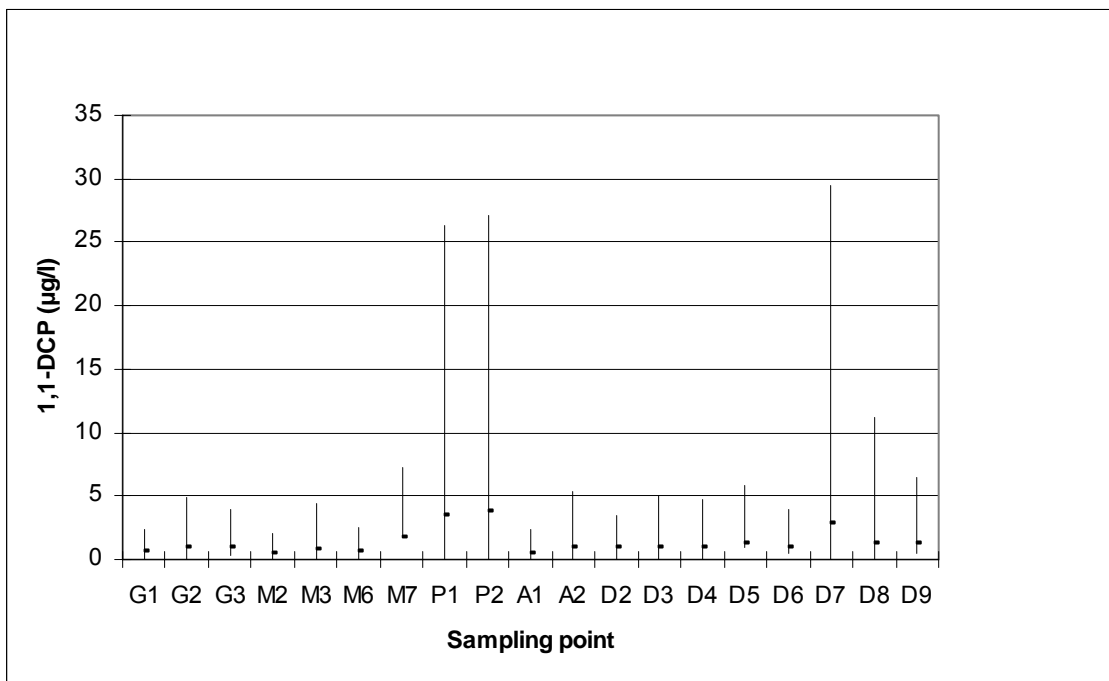


(a)

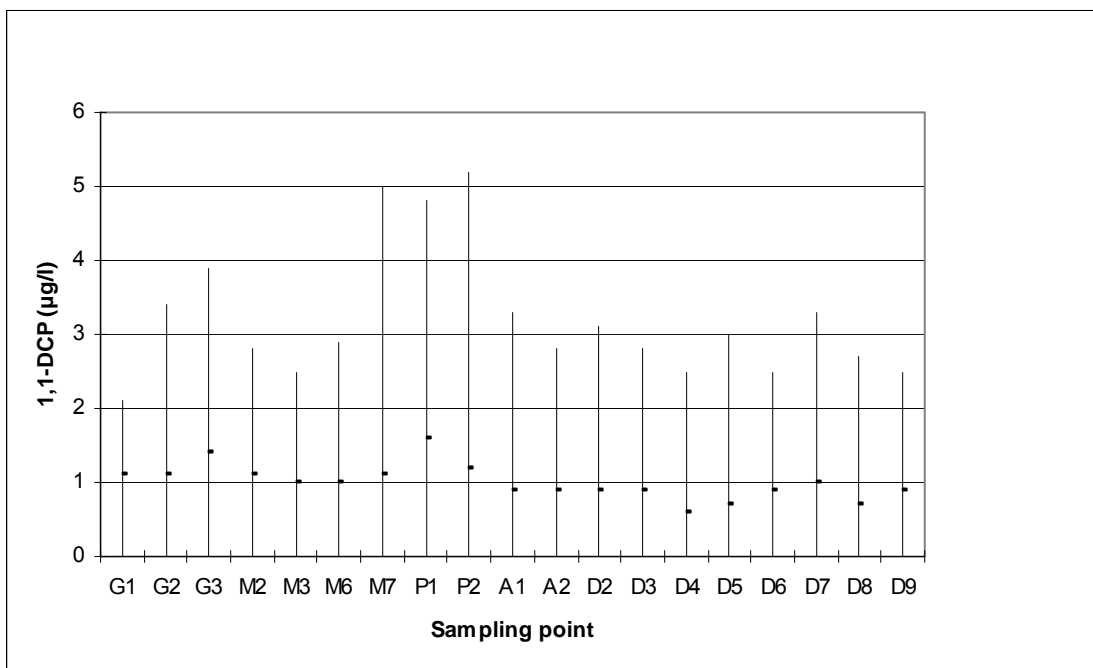


(b)

Figure 5. Minimum, mean and maximum concentrations of CH in the chlorinated samples during the years (a) 1999-2001, (b) 2002-2003 (For abbreviations of sampling points see Table 1)

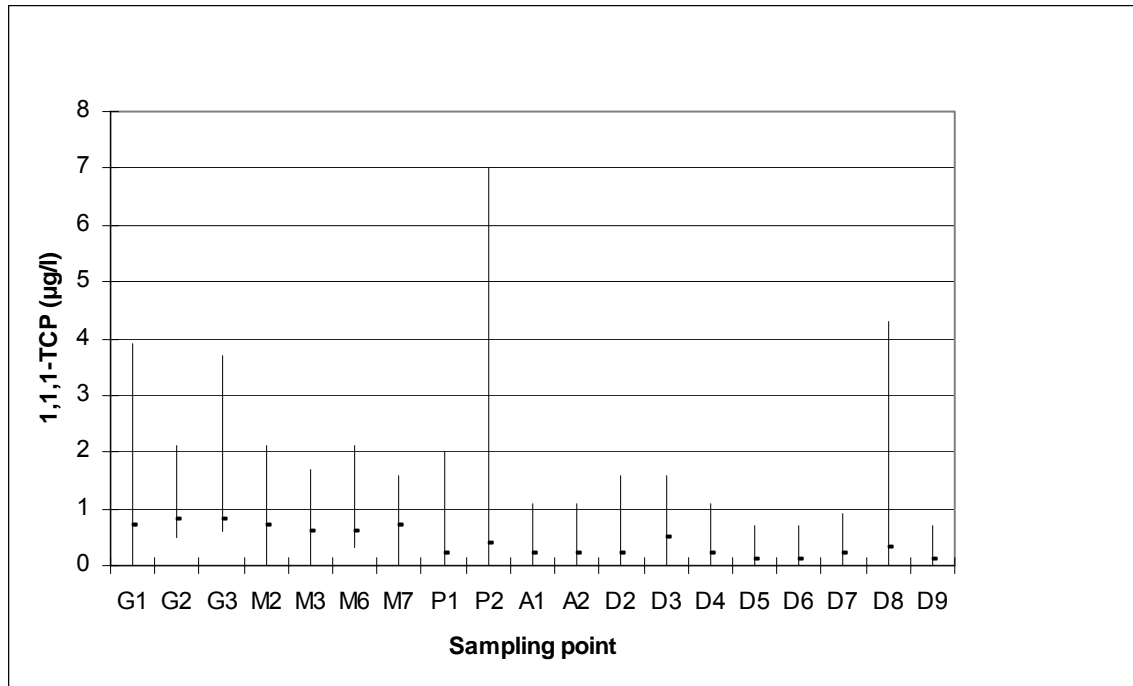


(a)

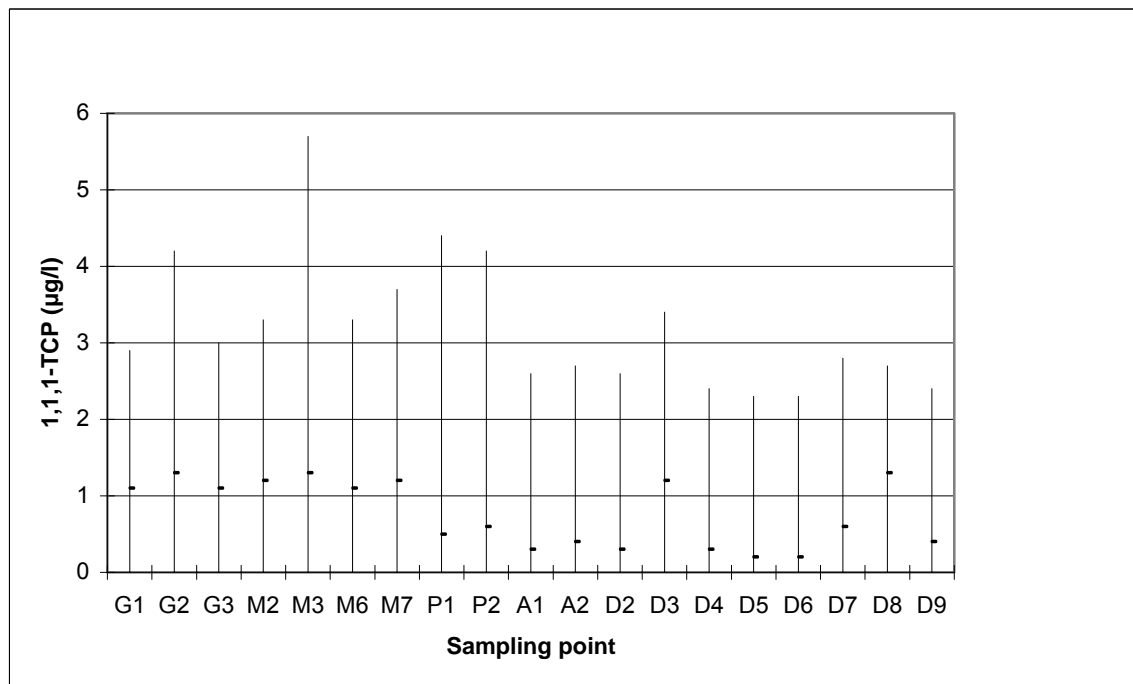


(b)

Figure 6. Minimum, mean and maximum concentrations of 1,1-DCP in the chlorinated samples during the years (a) 1999-2001, (b) 2002-2003 (For abbreviations of sampling points see Table 1)



(a)



(b)

Figure 7. Minimum, mean and maximum concentrations of 1,1,1-TCP in the chlorinated samples during the years (a) 1999-2001, (b) 2002-2003 (For abbreviations of sampling points see Table 1)

The quality of raw water, which changes during time, as well as chlorination conditions affect the formation of DBPs. The different categories of DBPs can be influenced in a different manner from changes in these parameters. High pH values enhance the formation of THMs, but limit the formation of HAAs and favor the decomposition of HKs.

Long reaction time leads to the formation of larger concentrations of THMs and some species HAAs, but also to decomposition of HANs, HKs and other species of HAAs (Singer, 1994; Nikolaou *et al.*, 2002; 2004). Therefore, more detailed studies are required regarding the behaviour of the different DBPs species under varying disinfection conditions, in order to be able to define the optimum treatment scheme to minimize the formation of all DBPs and not only the regulated/most studied ones. Moreover, the DBPs that may form in disinfected water are much more than those determined during the present study, and also more than those that can be determined with the currently established analytical methods worldwide. This is confirmed by the fact that 50% of the total organic halide (TOX) in chlorinated waters and 50% of the assimilable organic carbon (AOC) in ozonated waters (Richardson, 2005), is still not accounted for. The inclusion of more compounds in future studies, and the development of analytical methodologies for determination of new DBPs consist challenging aspects for the complete evaluation of the quality and safety of drinking water.

ACKNOWLEDGEMENTS

The authors would like to thank the Athens Water Supply and Sewerage Company (EYDAP SA) for the financial support of this study, and in particular Dr. Ph. Tzoumerkas and Leonidas Kousouris for their scientific and technical assistance.

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