Trihalomethanes Issues Drinking Water After Chlorination Treatment

MARIA COHL[1], LILIANA LAZAR[2]*, IGOR CRETESCU[3]*, ION BALASANIAN[2]

[1]SC APAVITAL SA Iasi, 6 Mihai Costachescu Str., 700495 Iasi, Romania

[2]"Gheorghe Asachi" Technical University of Iasi, Faculty of Chemical Engineering and Environmental Protection, Chemical Engineering Department, 73 Dimitrie Mangeron Str., 700050, Iasi, Romania

[3]"Gheorghe Asachi" Technical University of Iasi, Faculty of Chemical Engineering and Environmental Protection, Environmental Engineering and Management Department, 73 Dimitrie Mangeron Str., 700050, Iasi, Romania

Water disinfection, using oxidants bases-chlorine in order to diminish or to eliminate in totality the microbial charge in drinking water, presents the disadvantage of the formation of trihalomethanes (THMs), which are toxic and carcinogens compounds. The studies of THMs in water during the disinfection process and the possibilities for diminishing their content in drinking water were carried out in the municipal water treatment plant of Iasi, using gaseous chlorine and chlorine dioxide, respectively. Comparative analysis of the obtained results after the water disinfection by chlorination using the above mentioned disinfection agents, indicated that chlorine dioxide leads to lower concentrations of trihalomethanes and provides better quality drinking water in terms of the main water quality indicators. These results, based on seven years (2006 – 2013) of monitoring for THMs and for the main water quality indicators, recommended the use of chlorine dioxide for water disinfection, when the water is provided from the surface source, such as the Prut River.

Key words: chlorine, chlorine dioxide, chlorination, chlorinated by-products (CBPs), disinfection, monitoring, water treatment plant of Iasi

The decrease or even the total elimination of the microbiological charge of water can be achieved by chemical disinfection in the presence of physical agents or strong chemical oxidants [1–4]. The most common disinfection method used in the water treatment technology for drinking water preparation is chlorination, applied both in individual and combined systems. The purpose of water chlorination is destroying the pathogenic microorganism using gaseous chlorine (Cl_2), monochloramine (NH_2Cl), or chlorine dioxide (ClO_2) [3–6], in order to comply with the legislation, in the case of water quality indicators [7–9]. The alternatives to water chlorination are the use of ozone [3, 10] or UV radiations [11–13], as disinfection/oxidant agents, or combined procedures such as: O_3/H_2O_2, UV/H_2O_2, O_3/UV [3, 14].

A drawback of water disinfection by chlorination is the generation of secondary compounds by chemical reactions between the common disinfection agents and the organic compounds from water during the disinfection treatment [3, 4]. The usual products resulting from chlorine reactions are: the trihalogenated methanes, the halogenated acetic acid, acetaldehydes, acetonitriles and ketones; their presence is a strong drawback of water chlorination [15–18]. Trihalomethanes (THMs) are formed by the aqueous chlorination of humic substances, of soluble compounds secreted from algae and of naturally occurring nitrogenous compounds [1, 3]. The extent of the formation of trihalomethanes is dependent on several water quality parameters, such as TOC concentration, UVA_{254}, bromide concentration and temperature. It is also dependent on chlorination conditions, such as chlorine dose, pH, ammonia concentration and contact time [2, 3].

THMs are carcinogenic, mutant and toxic compounds, with major negative effects to human health [3, 19, 20]. The chemicals representing risk factors are of major interest for the scientists, due to the current use of a constant increasing number of chemicals widely used in industry and agriculture [18–20]. The issues connected to acute and semi acute intoxication are approached with respect to the exposure time of the species used in low concentrations. The biological risk factors are generally controlled in the water disinfection stage and by maintaining a chlorine concentration of 0.25 mg Cl_2/L in the water distribution network. New risks concerning the infectious diseases can show up due to the presence of resistant and adapted viruses from water, which contaminate it with even more aggressive pathogens. In consequence, the water re-chlorination at the final points of the distribution networks was applied [3, 18].

The water research during the last decades has focused on finding disinfection systems able to generate small amounts of THMs, as in the drinking water treatment plants for Iasi city. The water supply for Iasi comes from the underground water springs from Timisesti and from Prut River and Chirita Lake (which has the role of a pre-decantation basin of the Prut river water, being an important part of the public water system). The surface water treatment is performed in the water treatment complex of Chirita from "*APAVITAL*" Company [21–23].

The aim of this paper is a comparative study of the trihalomethanes issued in the drinking water during the disinfection process using gaseous chlorine and respectively chlorine dioxide as chlorination agents, in order to reduce the formation of chlorinated intermediates as secondary compounds.

Experimental part

In the "*APAVITAL*" Autonomous State Management Company of Water under the administration of Iasi Municipality, there are two main water treatment plants for drinking water preparation: Sorogari and Chirita, where the raw water is provided from two sources: Chirita Lake (supplied by gravitation) and Prut River (supplied by pumping) [23].

During the period 2006–2013, both these water treatment plants from the "*APAVITAL*" Company were investigated

* *email: lillazar@tuiasi.ro; icre@tuiasi.ro*

No.	Water quality indicator	MCL in drinking water	Specific determination method
1	Temperature [°C]	-	SR EN 1622/2000
2	pH	≥ 6.5 ... ≤ 9.5	SR ISO 10523/2009 SR ISO 10523/2012
3	Free chlorine, [mg Cl_2/L] - at the input of distribution network - at the output of distribution network (water consumers)	 0.50 0.25	SR ISO 9297/2001
4	Oxidability index [mg O_2/L]	5.00	SR EN ISO 8467/2001
5	Total Trihalomethanes (TTHMs) [µg/L]	100	SR ISO 12997/1997

MCL = maximum concentration limit, according to Law 311/2004.
Sampling was carried out according to SR ISO 5667-1/2007and SR ISO 5667-3/2004

Table 1
WATER QUALITY INDICATORS, MCL AND CORRESPONDING SPECIFIC DETERMINATION METHOD [9]

with regards the issue of trihalomethanes in the drinking water, as a consequence of the disinfection process achieved with gaseous chlorine and chlorine dioxide as chlorination/oxidation agents. The aim of using such kinds of disinfection agents was to diminish the formation of intermediates as secondary compounds.

The most widely used disinfection method is that based on gaseous chlorine (Cl_2), which is injected directly in the raw water [1–4]. When gaseous chlorine is dispersed in water, a rapid hydrolysis to form hypochlorous acid (HOCl) occurs according to the following reaction [2]:

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \quad (1)$$

Hypochlorous acid is a weak acid that dissociates partially in water as follows [2]:

$$HOCl \leftrightarrow H^+ + OCl^- \quad (2)$$

Relative proportions of hypochlorous acid (HOCl) and hypochlorite ion (OCl^-) are dependent upon *p*H [2, 3]. Between a *p*H of 6.5 and 8,5 this dissociation is incomplete and both HOCl and OCl^- species are present to some extent. Below a *p*H of 6.5, no dissociation of HOCl occurs, while above a *p*H of 8.5, complete dissociation to OCl^- occurs. As the germicidal effects of HOCl is much higher than that of OCl^-, chlorination at a lower *p*H is preferred [3]. Because HOCl dominates at low *p*H, chlorination provides more effective disinfection at low *p*H [2, 3]. At high *p*H, OCl^- dominates, which causes a decrease in disinfection efficiency. Hypochlorous acid, the prime disinfecting agent, is therefore dominant at a *p*H below 7.5 and is a more effective disinfectant than hypochlorite ion, which dominates above *p*H 7.5 [2].

The pathogen surfaces carry a natural negative electrical charge, being more readily penetrated by the uncharged, electrically neutral hypochlorous acid than the negatively charged hypochlorite ion. In order to enhance this process, it is recommended to maintain the predominance of hypochlorous acid during the disinfection treatment. The disinfecting action of hypochlorous acid is based on its moving through slime coatings, cell walls and resistant shells of waterborne microorganisms, leading to the destruction of these pathogens, or to annihilate their reproduction capacity [4]. The gaseous chlorine dose ranges between 0.5 – 2 mg Cl_2/L according to the initial organic content of raw water, while the residual chlorine does not exceed the maximum admissible value of 0.5 mg Cl_2/L [1-3].

In order to perform the water disinfection using chlorine dioxide (ClO_2), the solution is prepared using a special device, namely Alldos Oxiperm, and the corresponding dosing system is namely Grundfor-Alldos; both of them are integrated in the water disinfection line from the Chirita Water Treatment Plant of Iasi City. The chlorine dioxide is generated by a chemical reaction achieved at ambient temperature, between sodium chlorite and hydrochloric acid, as follows [3, 4]:

$$5\ NaClO_2 + 4\ HCl \rightarrow 4\ ClO_2 + 5\ NaCl + 2\ H_2O \quad (3)$$

Chlorine dioxide is a powerful oxidizing agent that can decompose into chlorite; in the absence of oxidizable substances and in the presence of alkali, it dissolves in water, decomposing with the slow formation of chlorite (ClO_2^-) and chlorate (ClO_3^-) [3]:

$$2\ ClO_2 + H_2O \rightarrow 6\ ClO_2^- + 2\ H^+ \quad (4)$$

Chlorite is relatively stable in the presence of organic material but can be oxidized to chlorate by free chlorine if added as a secondary disinfectant [3]:

$$ClO_2^- + OCl^- \rightarrow ClO_3^- + Cl^- \quad (5)$$

Chlorate is therefore produced through the reaction of residual chlorite and free chlorine during secondary disinfection.

Chlorine dioxide is an extremely effective disinfectant and bactericide, equal or superior to chlorine on a mass dosage basis because it is readily soluble in water. In contrast to the hydrolysis of chlorine gas in water, chlorine dioxide in water does not hydrolyze to any appreciable extent but remains in a solution as a dissolved gas [1, 2]. Theoretically, chlorine dioxide disappears and 50 % turns into chlorite and chlorate, relative to the initial amount. In comparison with chlorine, smaller quantities are required in order to obtain an active residual disinfectant with high efficiency in cases where the organic substances quantities are large [5]. Chlorine dioxide rapidly inactivates most microorganisms over a wide *p*H range. It is more effective than chlorine (for pathogens other than viruses) and is not pH dependent between *p*H 5–10 [3].

During our investigation on the influence of chlorination agents on the secondary disinfection compounds, the following trihalomethanes were identified by gas chromatography method: trichlor-metan (chloroform $CHCl_3$), bromo-dichloromethane ($CHBrCl_2$), dibromo-chloromethane ($CHBr_2Cl$), bromoform ($CHBr_3$) and the total trihalomethanes (TTHMs) [1, 3]. At the same time, the monitoring of the main quality indicators according to the actual standard methods was performed (table 1).

In order to control the process of trihalomethane formation in water during the disinfection treatment, the additional water quality indicators were monitored; so, the correlation between their nature and concentration and

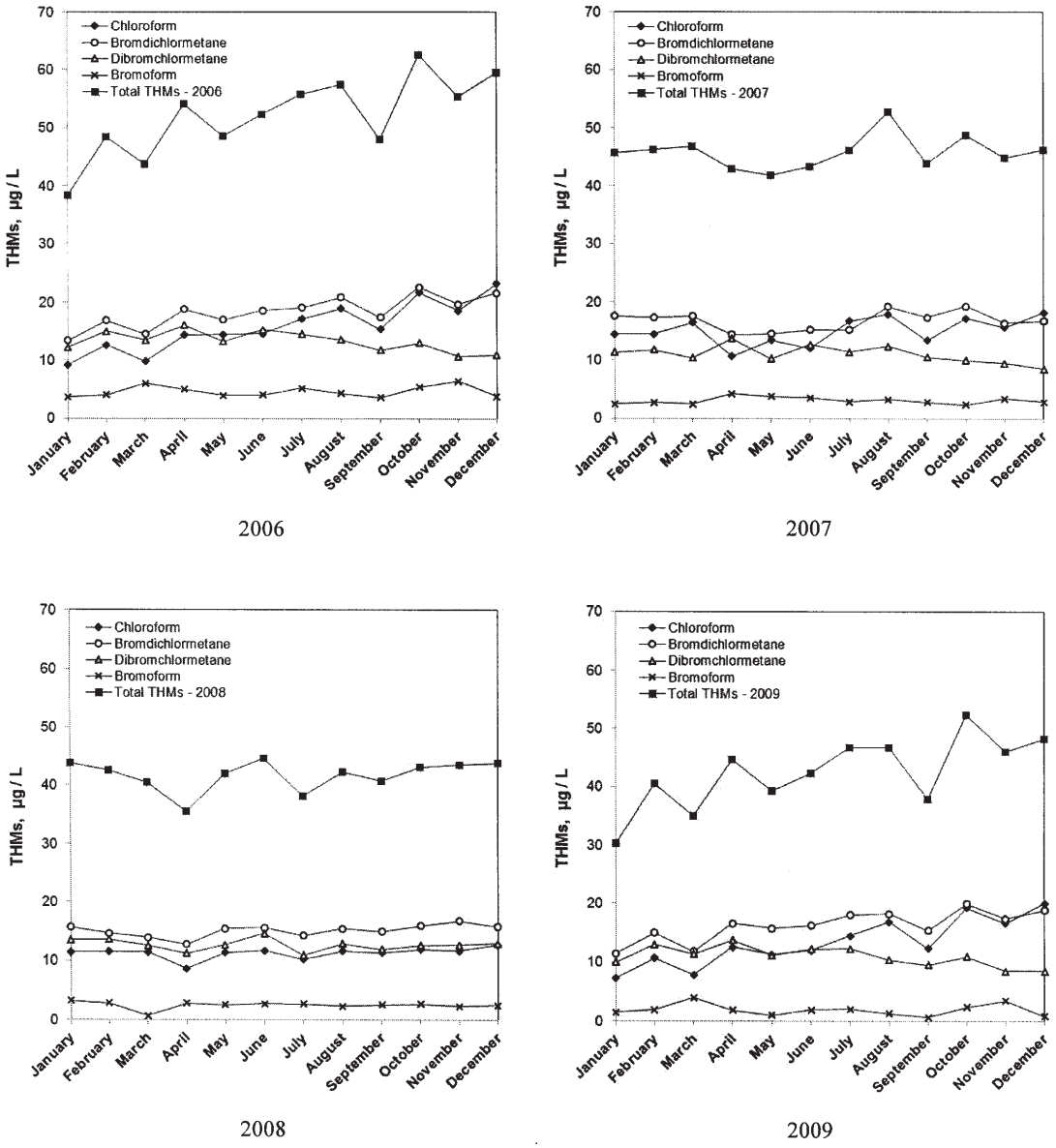

Fig.1. The influence of the gaseous chlorine (as disinfection agent) on the formation of trihalomethanes in water provided from the Prut River, in the period 2006–2009

the main water quality indicators was established. In this study, which focused on water disinfecting, the monitoring of the following water quality indicators are presented: pH, temperature, trihalomethanes concentration, free chlorine concentration, and the oxidability.

Oxidability is an indirect chemical index assessing the water pollution, expressed as the amount of oxygen required to oxidize some organic compounds of earthly origin or that are accidentally accumulated in the water [7–9].

The trihalomethanes in water were determined using a Shimadzu gas-chromatograph, applying the "static head space" procedure (see standard method in table 1). This method is based on the differences in the volatilities, molecular weights and polarities of the trihalomethanes involved in the analytical process [21, 22]. The determination of the oxidability index was performed by the oxidative method, according to the standard method mentioned in table 1. The free residual chlorine in the drinking water was determined by titration with methyl-orange in compliance with STAS 6334/1978. The residual chlorine dioxide in the treated water was determined using an on line electro-chemical analyzer ALLDOS type, which incorporates an EMIS electrode with high selectivity toward ClO_2 [21, 24].

Results and discussions

The trihalomethane formation in water during the disinfection process of raw water provided from the surface water (i.e. Prut River) was investigated during 2006–2009 at the Sorogari Water Treatment Plant, where gaseous chlorine was used as a disinfection agent, and in the period 2011–2013, at the Chirita Water Treatment Plant, where dioxide chlorine was used as a disinfection agent, respectively. Nowadays, the treated water at Chirita Treatment Plant reaches the storage tanks from Sorogari Treatment Plant, from where it enters the distribution network of the drinking water of the Iasi city by gravity.

The monitoring of water quality indicators of the raw water provided from Prut River, was carried out according to the analytical procedures, being the subject of a previous study [25]. According to the results presented in our previous studies in both Sorogari and Chirita treatment plants, during the period of 2009–2013, no significant changes of water quality indicators were reported. Based on these results, the comparative study presented in this article was achieved.

The influence of water chlorination agents on the monthly variations of trihalomethanes issued after water disinfection are graphically presented in Figure 1 (for gaseous chlorine) and in figure 2 (for chlorine dioxide). In

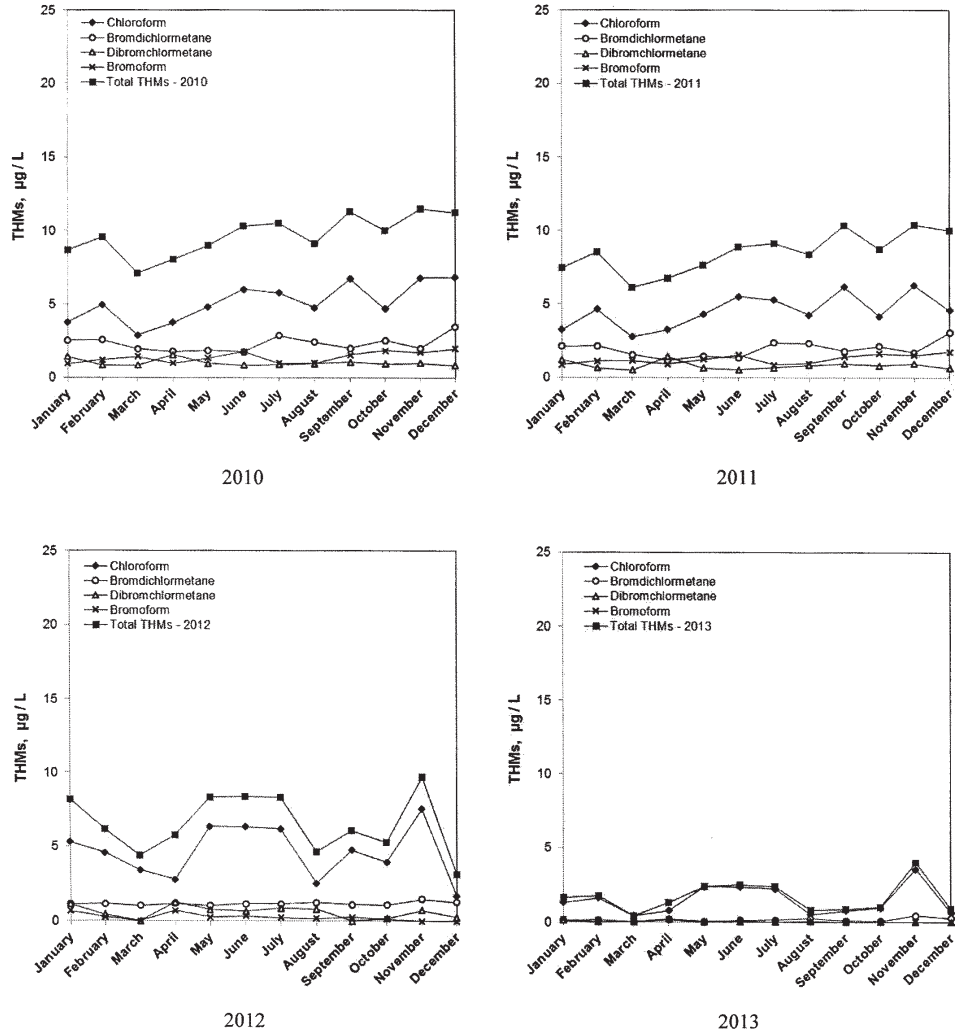

Fig. 2. The influence of the chlorine dioxide (as disinfection agent) on the formation of trihalomethanes in water provided from the Prut River, in the period 2010–2013

Table 2

THE AVERAGES OF TRIHALOMETHANES IN THE CASE OF WATER DISINFECTION USING GASEOUS CHLORINE AND CHLORINE DIOXIDE, FOR WATER PROVIDED FROM THE PRUT RIVER, IN THE PERIOD 2006–2013

Chlorination agents		Trihalomethanes concentration, µg/L															
		Chloroform				Bromo-dichloromethane				Dibromo-chloromethane				Bromoform			
Gaseous chlorine (Cl_2)	Annual season	*2006*	*2007*	*2008*	*2009*	*2006*	*2007*	*2008*	*2009*	*2006*	*2007*	*2008*	*2009*	*2006*	*2007*	*2008*	*2009*
	Spring	12.85	13.48	10.46	10.58	16.75	15.48	14.03	14.70	14.23	11.42	12.15	12.08	4.95	3.46	2.00	2.26
	Summer	16.84	15.55	11.19	14.45	19.47	16.54	15.09	17.46	14.40	12.14	12.81	11.60	4.48	3.19	2.56	1.73
	Autumn	18.52	15.40	11.69	16.07	19.86	17.66	15.89	17.56	11.79	9.96	12.41	9.66	5.12	2.82	2.50	2.17
	Winter	15.01	15.68	11.88	12.67	17.27	17.20	15.33	15.05	12.69	10.54	13.34	10.51	3.80	2.64	2.83	1.41
	Annual average	**15.81**	**15.03**	**11.31**	**13.44**	**18.34**	**16.72**	**15.09**	**16.19**	**13.28**	**11.02**	**12.68**	**10.96**	**4.59**	**3.03**	**2.47**	**1.89**
Chlorine dioxide (ClO_2)	Annual season	*2010*	*2011*	*2012*	*2013*	*2010*	*2011*	*2012*	*2013*	*2010*	*2011*	*2012*	*2013*	*2010*	*2011*	*2012*	*2013*
	Spring	3.81	3.44	4.16	1.16	1.85	1.38	1.05	0.05	1.10	0.86	0.65	0.08	1.24	1.15	0.30	0.06
	Summer	3.50	5.01	5.01	1.68	2.34	2.00	1.16	0.16	0.88	0.67	0.74	0.04	1.24	1.12	0.23	0.00
	Autumn	6.05	5.54	5.41	1.74	2.17	1.87	1.19	0.19	0.99	0.89	0.29	0.00	1.71	1.53	0.12	0.00
	Winter	4.56	4.16	3.84	1.17	2.85	2.44	1.16	0.16	1.03	0.82	0.59	0.03	1.37	1.24	0.30	0.05
	Annual average	**4.98**	**4.54**	**4.61**	**1.44**	**2.30**	**1.93**	**1.14**	**0.14**	**1.00**	**0.81**	**0.57**	**0.04**	**1.39**	**1.26**	**0.24**	**0.03**

Annual season: *Spring* (March – May); *Summer* (June – August); *Autum* (September – November); *Winter* (December – February)

table 2 are presented the seasonal and annual average s of four analysed trihalomethanes (chloroform, bromo-dichloromethane, dibromo-chloromethane, bromoform).

Monthly and annual averages of trihalomethanes formed after water disinfection with gaseous chlorine or chlorine dioxide were within the Prut River below the limit of 100 µg/L TTHMs imposed by law [9]. During 2006–2009, recorded annual limits for trihalomethanes analysed after water disinfection with gaseous chlorine were: 11.31 to 15.81 µg/L chloroform, 15.09 – 18.34 µg/L bromo-dichloromethane, 10.96 – 13.28 µg/L dibromochlormetane, 1.89 – 4.59 µg/L bromoform and 41.71 – 52.01 µg/L TTHMs.

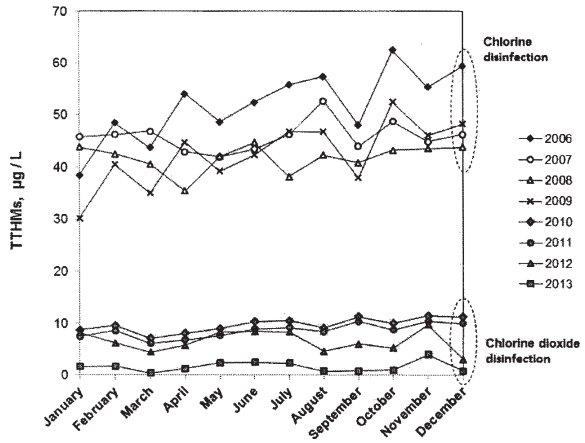

Fig. 3. The influence of water chlorination agent on the total trihalomethanes concentration

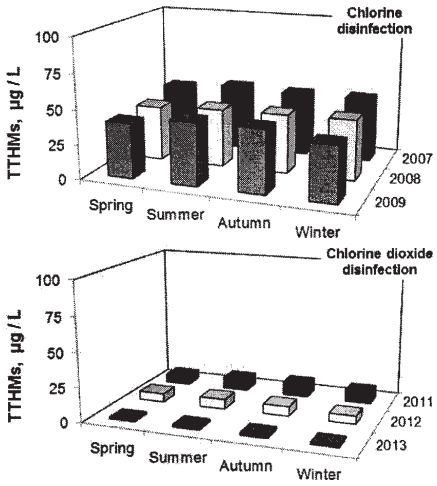

Fig. 4. Seasonal variation of total trihalomethanes concentration in the treated water, using gaseous chlorine (2007–2009) and chlorine dioxide (2011–2013)

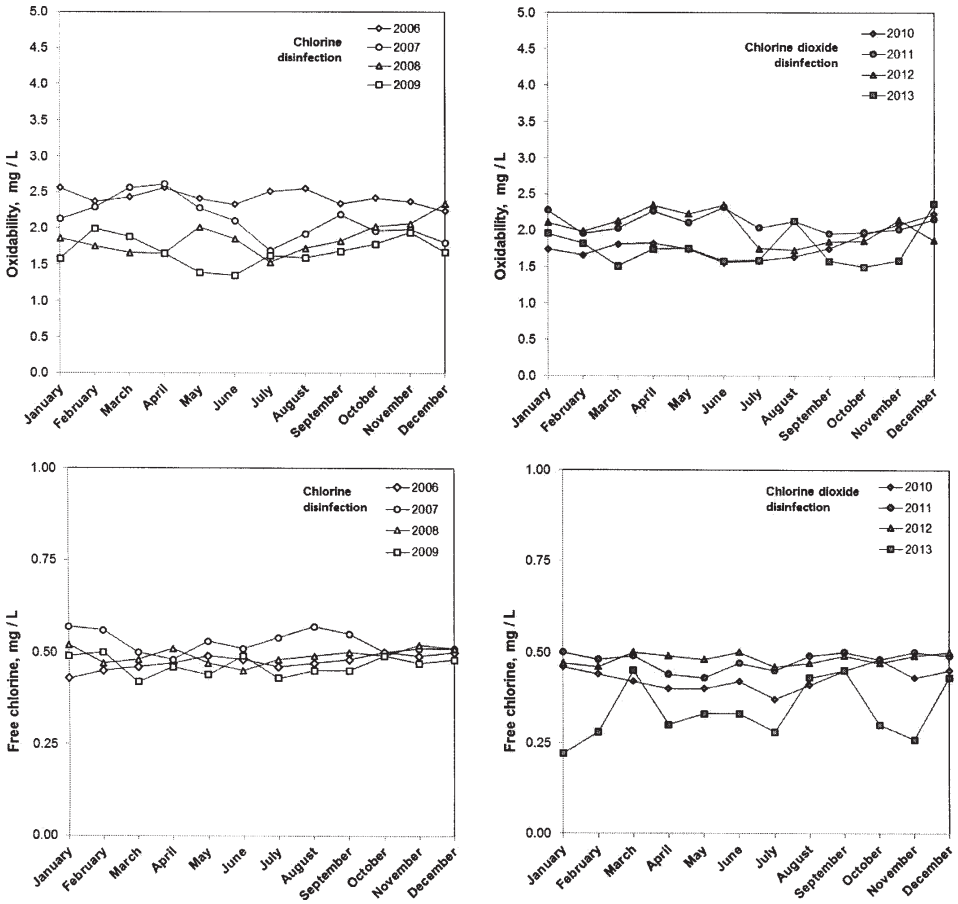

Fig. 5. The influence of water chlorination agents on the oxidability index of the chlorinated drinking water

Fig. 6. The influence of water chlorination agents on the free chlorine concentration of the chlorinated drinking water

In the period 2010–2013, the monitoring of the annual data limits recorded for trihalomethanes after water disinfection with chlorine dioxide are ranged as follows: 1 to 4.98µg/L chloroform, 0.81 – 4.54 µg/L bromo-dichloromethane, 0.24 – 4.61 µg/L dibromochlormetane, 0.03 – 1.44 µg/L bromoform and 4.78 – 6.26 µg/L TTHMs.

From the comparative analysis on the variation for the total trihalomethanes formed during the process of water disinfecting with chlorine dioxide it can be pointed out that the resultant drinking water has a higher quality than the drinking water obtained after disinfection with chlorine (fig. 3).

A comparative analysis of the seasonal variation for the average s of total trihalomethanes (fig. 4) and oxidability index of water after disinfection using gaseous chlorine and dioxide chlorine (fig. 5) demonstrated that the variation of total trihalomethanes concentrations is in concordance with the variation of oxidability index of disinfected water, which falls also under the maximum legal concentration of 5 mg O_2/L [9].

As it can be seen from figures 3 and 4, the chlorine dioxide significantly reduced the generation of disinfection by-products such as THMs. Thus, in the case of water disinfection with chlorine dioxide the annual average concentrations of total trihalomethanes during 2010–2013 were: 5.54 µg/L in 2010, 8.53 µg/L in 2011, 6.53 µg/L in 2012, 1.65 µg/L in 2013. These values are much lower than annual concentrations of total trihalomethanes, in the case of water disinfection with gaseous chlorine (41 – 52 µg/L).

These advantages allow the recommendation for using chlorine dioxide in the disinfection of drinking water, to the detriment of gaseous chlorine, taking into consideration the improvement of the water quality in terms of microbiological indicators (the concentration of pathogens drastically decreased, usually total disappeared if the disinfectant/oxidant dose is correctly applied).

The maximum admissible value for residual chlorine is the treated water 0.5 mg Cl_2/L at the output of the water

Table 3
THE AVERAGE VALUES OF THE MAIN WATER QUALITY INDICATORS DEPENDING ON THE TYPE OF DISINFECTION AGENT

Water quality indicator	UM	Disinfection using gaseous chlorine (2007 – 2009)				Disinfection using chlorine dioxide (2011 – 2013)			
		Spring	*Summer*	*Autum*	*Winter*	*Spring*	*Summer*	*Autum*	*Winter*
pH	pH units	7.63	7.54	7.46	7.70	7.80	7.56	7.56	7.61
Temperature	°C	13.63	24.36	16.15	6.78	13.79	23.86	16.71	6.66
Oxidability Index	mg O_2/L	2.01	1.96	1.94	2.06	1.97	1.71	1.83	1.93
Free Chlorine	mg Cl_2/L	0.48	0.49	0.50	0.51	0.43	0.43	0.44	0.41
Total Trihalomethanes	µg/L	40.93	44.77	44.59	42.10	4.78	5.94	6.26	5.60

Annual season: *Spring* (March – May); *Summer* (June – August); *Autum* (September – November); *Winter* (December – February)

treatment plant and the maximum of 0.1 – 0.25 mg Cl_2/L in the drinking water at consumers [9]. These monthly average concentrations of free chlorine during the monitoring period of 2006–2013 are presented in figure 6.

The average values of the main water quality indicators for the drinking water prepared in treatment plants from Iasi, using both above mentioned chlorinating agents, are presented in table 3. As can be observed, these values are below the limit imposed by the legislation concerning the quality of drinking water [9].

Conclusions

This paper shows the influence of the chlorination agent on the formation of trihalomethanes during the disinfection step performed in the water treatment plants of Iasi city. This study was started at Sorogari Treatment Plant, where gaseous chlorine was used as disinfection agent during 2006–2009. The study was continued at Chirita Treatment plant, where chlorine dioxide was used during 2011–2013.

The influence of chlorination agents on the secondary disinfection compounds was studied by monitoring the halogenated compounds such as: chloroform, bromo-dichloromethane, dibromo-chloromethane, bromoform and the sum thereof. The oxidability and residual chlorine were also measured. The analysis of trihalomethanes formed from the disinfection process has shown that for all chlorination agents used, the highest concentration was found for the chloroform and the lowest was noticed smallest in the case of bromoform.

The drinking water resulting from the disinfection process with chlorine dioxide has a better quality than that obtained in the case of disinfection with gaseous chlorine. Chlorine dioxide was proved to be more efficient, due to its higher disinfection efficiency and to the shorter contact.

Referecens

1.AGUS, E., VOUTCHKOV, N., DAVID L. SEDLAKA, D.L., Desalination, 237, nr.1-3, 2009, p.214

2.*** WHO EHC 216, Disinfectants and Disinfectant by-products, World Health Organization, Geneva, 2000

3.*** US EPA, Office of Water, Guidance Manual, Alternative Disinfectants and Oxidants, EPA-815-R-99-014, Washington DC 20460-0003, 1999

4.*** Drinking Water Chlorination - A Review of Disinfection Practices and Issues, Chlorine Chemistry Council, February 2003 (http://www.waterandhealth.org/)

5.NEGOITESCU, A., TOKAR, A., Rev. Chim. (Bucharest), **63**, no.10, 2012, p.1079

6.BOGATU, C., PODE, V., VLAICU, I., LEONTE, E., MARSAVINA, D., Rev. Chim. (Bucharest), **60**, no.7, 2009, p.735

7.*** EC DIRECTIVE 98/83/EC of 3 November 1998 on the quality of water intended for human consumption, Official Journal of the European Communities, L 330, 5.12.1998, p.32-54

8.*** Law no. 458/2002 regarding the drinking water quality, published in Romanian Official Monitor, part I, no. 552 from 29 July 2002

9.*** Law no. 311/2004 for the modification and completion of Law no. 458/2002 regarding the drinking water quality, published in Romanian Official Monitor, part I, no. 582 from 30 June 2004

10.POHONTU, C., CRETESCU, I., SECULA, M.S., CIOROI, M. MACOVEANU, M., Annal. "Dunarea de Jos" Univ. Galati, Section Mathematics, physics, chemistry, informatics, III(XXXII), Fasc. II, 2009, p.26

11.BRAUNSTEIN, H., CRETESCU, I., ANTOHI, C., DIACONU, M., MACOVEANU, M., Rev. Chim. (Bucharest), **59**, no.5, 2008, p.601

12.NEGREANU-PIRJOL, B., ZAGAN, S., GORUN, E., MEGHEA, A., ZAGAN, R., STANCIU, G., Rev. Chim. (Bucharest), **61**, no.12, 2010, p.1262

13.OGUMA, K., KITA, R., SAKAI, H., MURAKAMI, M., TAKIZAWA, S., Desalination, 328, 2013, p.24

14.CRETESCU, I., BRAUSTEIN, H., ANTOHI, C., SECULA, M.S., COJOCARU, C., MACOVEANU, M., Enhancement of phenolic wastewater treatment by advanced oxidation processes using Fenton-UV radiation in resonant elliptic cavity. Proceeding of "the First European Conference on Environmental Applications of Advanced Oxidation Processes", Chania, Greece, id 88, 2006, p.5

15.BARBEAU, B., DESJARDINS, R., MYSORE, C., PRÉVOST M., **Water** Res., 39, nr.10, 2005, p. 2024

16.GOPAL, K., TRIPATHY, S.S., BERSILLON, J.L., DUBEY, S.P., J. Hazard. Mater., 140, nr.1-2, 2007, p.1

17.LEGAY, C., RODRIGUEZ, M.J., SÉRODES, J.B., LEVALLOIS, P., Sci. Total Environ., 408, nr.3, 2010, p.456

18.SORLINI, S., GIALDINI, F., BIASIBETTI, M., COLLIVIGNARELLI, C., Water Res., 54, 2014, p.44

19.CHOWDHURY, S., CHAMPAGNE, P., Sci. Total Environ., 407, nr.5, 2009, p. 1570

20.RIGHI, E., BECHTOLD, P., TORTORICI, D., LAURIOLA, P., CALZOLARI, E., ASTOLFI, G., NIEUWENHUIJSEN, M.J., FANTUZZI, G., AGGAZZOTTI, G., Environ. Res., 116, 2012, p.66

21.COHL, M., TEODOSIU C., BALASANIAN, I., Bull. Instit. Polit. Iasi, Section Chimie si Inginerie Chimica, 57, nr.4, 2011, p.99

22.POPOVICI, D.G., ONISCU, C., Environ. Eng. Manag. J., 9, nr.3, 2010, p.435

23. "APAVITAL" Autonomous State Management Company, under the administration of Iasi Municipality, Romania, http://www.apavital.ro

24.DASCALESCU, I., COHL, M., TEODOSIU, C., Environ. Eng. Manag. J., 10, nr.11, 2011, p.1789

25.COHL, M., LAZAR, L., BALASANIAN, I., Studies on the quality of water supply sources in Iasi city, Environ. Eng. Manag. J., 2014 (in press)

Manuscript received: 27.03.2014