Review Article

Global Nest: the Int. J. Vol 1, No 3, pp 143-156, 1999

ORGANIC BY-PRODUCTS OF DRINKING WATER CHLORINATION

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Received: 01/07/98
Accepted: 29/10/99

ABSTRACT

Chlorination of drinking water leads to the formation of a variety of Disinfection By-Products (DBPs) that may have adverse health effects on humans. Research on this subject has been continued and new epidemiological and toxicological studies have been conducted. This review summarizes factors affecting DBP formation and predictive equations proposed for it, physical and chemical properties, environmental fate, actual measurements of these compounds and technologies for controlling them, as well as regulation and currently proposed changes of their Maximum Contaminant Levels (MCLs) after evaluation by EPA of new data available.

KEYWORDS: Disinfection by-products, Chlorination, Trihalomethanes, Haloacetic acids

INTRODUCTION

The reaction of chlorine with Natural Organic Materials (NOM) existing in surface water leads to the formation of halogenated Disinfection By-Products (DBPs) (Stevens et al., 1976; Babcock and Singer, 1979; Christman et al., 1983) including trihalomethanes, haloacetic acids, haloaldehydes, haloketones, haloacetonitriles, chloropicrin, chlorophenols.

Trihalomethanes (THMs) were the first category of DBPs to be detected in drinking water (Bellar et al., 1974; Rook, 1974), followed by haloacetic acids (Quimby et al., 1980; Christman et al., 1983; Miller and Uden, 1983; Reckow and Singer, 1984; Krasner et al., 1989) and haloacetonitriles, haloketones, chloropicrin at lower concentrations (Trehy and Bieber, 1980; Barry, 1983; Krasner et al., 1989; Williams et al., 1997). According to animal studies, dichloroacetic acid is believed to be a more potent carcinogen than THMs (Bull and Kopfler, 1991; Regli et al., 1992). Dichloroacetonitrile has been shown to be mutagenic in bacterial assays (Simmon et al., 1977).

These observations resulted in an increasing number of studies and regulations, in an attempt to reduce the health risk deriving from exposure to DBPs and simultaneously keep the risk of microbial disease at as low levels as possible. This review summarizes information including formation, properties, actual measurements and regulation of DBPs.

FACTORS AFFECTING DBP FORMATION - PROPOSED PREDICTIVE EQUATIONS

The main factors affecting DBP formation (Singer, 1993; Singer, 1994; Pourmoghaddas and Stevens, 1995; Lekkas, 1996) are pH, contact time, temperature and season, concentration and properties of Natural Organic Materials (NOM), concentration of chlorine and residual chlorine and concentration of bromide.

pH. With increasing pH, trihalomethane formation increases, whereas haloacetic acid formation decreases (Krasner et al., 1989; Pourmoghaddas and Stevens, 1995). At high pH values, hydrolysis of many halogenated DBPs occurs (Krasner et al., 1989). As a result, total organic halide (TOX) concentration is lower at pH>8 (Singer, 1994).

Contact time. With increasing contact time, trihalomethane and haloacetic acid formation increases. On the other hand, DBPs such as haloacetonitriles and haloketones, which were initially formed, decay as a result of hydrolysis and reactions with residual chlorine (Singer, 1994).

Temperature and season. When temperature increases, reactions are faster and a higher chlorine dose is required, leading to higher formation of DBPs. Subsequently, DBP concentrations are expected to be higher in summer than in winter (Williams et al., 1997; Golfinopoulos et al., 1993; Golfinopoulos et al., 1996a,b; LeBel et al., 1997). However, influence of season and weather on the nature of NOM and on bromide concentration must be taken into account.

Concentration and properties of NOM. With increasing NOM concentration, DBP formation increases. Properties of NOM play an important role as well, since activated aromatic content of NOM increases DBP formation (Reckow et al., 1990). In addition, NOM contains hydrophobic and hydrophilic materials, the nature and distribution of which may vary with different types of vegetation in the watershed and different species of algae in water. This results in varying influence of NOM on DBP formation (Singer, 1994).

Concentration of chlorine and residual chlorine. With increasing chlorine dose and residual, formation of haloacetic acids becomes greater than trihalomethane formation. Also, more trihalogenated than mono- and di-halogenated species and more chlorinated than brominated species are formed (Singer, 1994). Depletion of the free chlorine residual ceases THM and haloacetic acid formation. However, limited formation of some other DBPs continues due to hydrolysis reactions (Singer, 1994).

Concentration of bromide. In the presence of bromide ion (Br-), more brominated and mixed chloro-bromo derivatives are formed (Krasner et al., 1989; Peters et al., 1991; Heller-Grossman et al., 1993; Pourmoghaddas and Stevens, 1995). This is the result of bromide oxidation to hypobromous acid (HOBr) by chlorine and the reaction of HOBr and residual HOCl with NOM. In waters with very high bromide concentration, the brominated species -bromoform, dibromoacetic acid- may be the major species formed (Singer, 1994).

Some predictive equations for the formation of DBPs based on the previous factors are give below.

$$C_{TTHM}=0.00309(TOC\cdot UV_{254})^{0.440}C_{Cl_2}^{0.409}t^{0.265}T^{1.06}$$

$$(pH-2.6)^{0.715}(C_{Br}+1)^{0.03} \tag{1}$$

(Harrington et al., 1992)

$$C_{TTHM}=52.13+10.73\ln(C_{chla})-20.95pH+$$

$$+243.23C_{Br}-147.45C_{Br}^{2}-27.77\mathrm{S}+$$

$$+120.01Sp-7.37T\cdot Sp+1.52TC_{Cl_2} \tag{2}$$

(Golfinopoulos et al., 1998)

$$THMFP=10+17Act_{Ar\text{-}R} \tag{3}$$

(Rechow et al., 1990)

$$C_{CHCl_3}=0/278(TOC\cdot UV_{254})^{0.616}C_{Cl_2}^{0.391}t^{0.265}T^{1.15}$$

$$(pH-2.6)^{0.800}(C_{Br}+1)^{-2.23} \tag{4}$$

(Malcolm Pirnie Inc., 1992)

$$C_{HCl_3}=k_1k_2TOC^{0.95}\left(\frac{C_{Cl_2}}{TOC}\right)^{0.28} t^z \qquad (5)$$

(Engerholm and Amy, 1983)

$$C_{CHBrCl_2}=0.863(TOC\cdot UV_{254})^{0.177}C_{Cl_2}^{0.309}t^{0.271}T^{0.720}$$

$$(pH-2.6)^{0.925}(C_{Br}+1)^{0.722} \qquad (6)$$

(Malcolm Pirnie Inc., 1992)

$$C_{CHBr_2Cl}=2.57\left(\frac{UV_{254}}{TOC}\right)^{-0.184} C_{Cl_2}^{-0.0746}t^{0.252}T^{0.57}$$

$$(pH-2.6)^{1.35}\ C_{Br}^{2.08} \qquad (7)$$

(Malcolm Pirnie Inc., 1992)

$$D_{DCAA}=0.605TOC^{0.291}UV_{254}^{0.726}C_{Cl_2}^{0.480}t^{0.239}T^{0.665}$$

$$(C_{Br}+1)^{-0.568} \qquad (8)$$

(Malcolm Pirnie Inc., 1992)

$$C_{TCAA}=87.102TOC^{0.335}UV_{254}^{0.901}C_{Cl_2}^{0.881}t^{0.264}pH^{-1.732}$$

$$(C_{Br}+1)^{-0.679} \qquad (9)$$

(Malcolm Pirnie Inc., 1992)

$$\frac{TCAAFP}{TTHMFP}=0.6+14UV_{254} \qquad (10)$$

(Reckow et al., 1990)

$$DCANFP=-2.3+0.053C_{Org\text{-}N} \qquad (11)$$

(Reckow et al., 1990)

The applicability of equations (1), (4), (6), (7), (8) and (9) has not been tested in waters with relatively high bromide concentrations, where the distribution of the species formed will probably be different. Additionally, high ammonia concentration also may affect DBP formation, due to consumption of chlorine (Harrington et al., 1992). Finally, season influence has not been taken into account, although it may have a significant effect on DBP formation. Season alinfluences has been incorporated into equation (2), which however does not include contact time (Golfinopoulos et al., 1998). For equation (5), the limitation is that only one type of humic acid has been considered as DBP precursor, which is not true for natural waters (Engerholm and Amy, 1983). Further investigation of the influence of natural organic materials is necessary in order to improve the applicability of the above equations for predicting DBP formation.

Comparison of Malcolm Pirnie Inc. predictions by the numerical models shown above and measured concentrations in California and North Carolina drinking waters (Greiner et al., 1992) has shown similar percentage deviations and central tendencies. The same is true for the comparison of Golfinopoulos et al., model predictions and measured concentrations in Athens drinking water (Golfinopoulos et al., 1998).

CATEGORIES AND MEASUREMENTS OF DBPS

In order to minimize DBP formation, it is necessary to study and analyze their occurrence and

Table 1. Values of constants in predictive equation for chloroform formation (Engerholm and Amy, 1983)

Constant	pH Value	Temperature (°C)	Value of constant
x	All	All	0.95
y	All	All	0.28
z		10	0.19
		20	0.22
		35	0.28
k_1	5.5		0.82
	7.0		1.00
	8.5		1.22
k_2		10	13.59
		20	16.75
		35	23.35

behaviour in water. Concentrations of compounds belonging to different categories of DBPs have been measured in several countries. Results of such measurements are presented in this section. Health effects are also mentioned. Physical - chemical properties and environmental fate of DBPs need to be taken into account as well and are summarized in Annex I (Sittig, 1985; Ehrenfeld et al., 1986; Merck, 1989; Vogel, 1989; Howard 1991a,b; Mackay et al., 1992a,b; Lyde, 1993-94; Shiu et al., 1994; Bowden et al., 1998).

A. Trihalomethanes

This category includes the following substances: Bromodichloromethane ($CHBrCl_2$), Bromoform ($CHBr_3$), Chloroform ($CHCl_3$) and Dibromochloromethane ($CHBr_2Cl$).

Additional lifetime cancer risk is 1/100000 at a concentration of 1.9 μg/l for bromodichloromethane, bromoform and chloroform. According to EPA, these three substances belong to carcinogens and Priority Toxic Pollutants. Bromoform, chloroform and dibromochloromethane belong to the category of Hazardous Wastes. Dibromochloromethane has shown positive results for mutagenesis in *Salmonella typhimurium.*

Measurements of THM concentrations in drinking waters are given in Table 2.

B. Haloacetic acids

The DBPs belonging to this category are Bromoacetic acid (MBA) ($BrCH_2COOH$), Bromochloroacetic acid (BCA) (BrClCHCOOH), α-, β- and γ-Chloroacetic acid (MCA) (ClCH2COOH), dibromoacetic acid (DBA) ($Br_2CHCOOH$), dibromochloroacetic acid (DBCA) ($Br_2ClCCOOH$), dichloroacetic acid (DCA) ($Cl_2CHCOOH$), dichlorobromoacetic acid (DCBA) ($Cl_2BrCCOOH$), tribromoacetic acid (TBA) (Br_3CCOOH) and trichloroacetic acid (TCA) (Cl_3CCOOH).

Monochloroacetic acid attacks skin, eyes and respiratory system. Dichloroacetic acid and trichloroacetic acid are animal carcinogens (Bull and Kopfler, 1991; Regli et al., 1992).

Measurements of haloacetic acid concentrations in drinking waters are given in Table 3.

C. Haloaldehydes

Bromoacetaldehyde ($BrCH_2CHO$), chloroacetaldehyde ($ClCH_2CHO$), dibromoacetaldehyde (Br_2CHCHO), dichloroacetaldehyde (Cl_2CHCHO), tribromoacetaldehyde (Br_3CCHO), trichloroacetaldehyde (Cl_3CCHO) and chloral hydrate ($C_2H_3Cl_3O_2$) are the main members of this category of DBPs.

Chloroacetaldehyde attacks eyes, skin and mucous membranes. According to EPA, chloral hydrate belongs to possible human carcinogens.

Measurements of chloral hydrate concentrations in drinking waters are given in Table 4.

D. Haloketones

1,1-dichloroacetone ($C_2H_4Cl_2CO$), 1,3-di-

Table 2. Measured concentrations of THMs in chlorinated drinking water

	$CHCl_3$ (μg l^{-1})	$CHCl_2Br$ (μg l^{-1})	$CHClBr_2$ (μg l^{-1})	$CHBr_3$ (μg l^{-1})
United States (Krasner et al., 1989)				
Spring	15	6.9	2.6	0.33
Summer	15	10	4.5	0.57
Fall	13	5.5	3.8	0.88
Winter	9.6	4.1	2.7	0.51
Israel (Heller-Grossman et al., 1993)				
Summer	10.5	39	203	353
Winter	20	25.5	196	265
Greece (Golfinopoulos et al., 1993)				
Spring	7.0	7.0	9.0	7.0
Summer	7.0	5.0	5.0	4.0
Range (yearly)	2-11	3-24	6-30	4-50
Canada (LeBel et al., 1997)				
Yearly mean	38.5	4.2	0.3	<0.1
Range	10.7 - 92.1	2.3 - 6.7	<0.1 - 0.5	<0.1 - 0.3

Table 3. Measured concentrations of haloacetic acids in chlorinated drinking water

(μg l^{-1})	MCA (μg l^{-1})	DCA (μg l^{-1})	MBA (μg l^{-1})	BCA (μg l^{-1})	TCA (μg l^{-1})	DBA (μg l^{-1})	BDCA (μg l^{-1})	CDBA (μg l^{-1})	TBA (μg l^{-1})
United States (Krasner et al., 1989)									
Spring	<1.0	7.3	<0.5	NM	5.8	0.9	NM	NM	NM
Summer	1.2	6.8	<0.5	NM	5.8	1.5	NM	NM	NM
Fall	<1.0	6.4	<0.5	NM	6.0	1.4	NM	NM	NM
Winter	1.2	5.0	<0.5	NM	4.0	1.0	NM	NM	NM
Israel (Heller-Grossman et al., 1993)									
Summer	NM	12	NM	36	6	90	NM	90	80
Winter	NM	18	NM	29	5	69	NM	90	50
The Netherlands (Peters et al., 1991)									
Range	NM	0.2-3	0.1-0.5	0.1-2.5	0.1-1.4	0.1-6.5	0.1-1.7	0.2-1.6	0.3-2.1
Canada (LeBel et al., 1997)									
Yearly mean	1.5	12.7	<0.01	NM	12.7	<0.01	NM	NM	NM
Range	0.2-7.8	2.1 -26.2	<0.01	NM	1.7 - 36.8	<0.01	NM	NM	NM

NM = not measured

chloroacetone ($C_2H_4Cl_2CO$) and 1,1,1-trichloroacetone ($C_2H_3Cl_3CO$) are the haloketones identified in water as by-products of chlorination.

Carcinogenic and mutagenic effects of the haloketones 1,1 - dichloropropanone and 1,1,1 - trichloropropanone on mice have been reported (Bull & Robinson, 1986).

Measurements of haloketone concentrations in drinking waters are given in Table 5.

E. Haloacetonitriles

The substances included in the present category are bromoacetonitrile (BAN) ($BrCH_2CN$), bromochloroacetonitrile (BCAN) (BrClCHCN), chloroacetonitrile (CAN) ($ClCH_2CN$), dibromoacetonitrile (DBAN) (Br_2CHCN), dichloroacetonitrile (DCAN) (Cl_2CHCN), tribromoacetonitrile (TBAN) (Br_3CCN) and trichloroacetonitrile (TCAN) (Cl_3CCN).

Dichloroacetonitrile has been shown to be mutagenic in bacterial assays (Simmon et al., 1977). Trichloroacetonitrile attacks eyes and skin. Carcinogenic and mutagenic effects of the haloacetonitriles BAN, DBAN and BCAN on mice have been reported (Bull & Robinson, 1986). Haloacetonitriles have also been found to damage DNA of mice (Pereira et al., 1986).

Measurements of haloacetonitrile concentrations in drinking waters are given in Table 6.

F. Chloropicrin: CCl_3NO_2

Chloropicrin attacks respiratory system, lungs, eyes and skin.

Measurements of chloropicrin concentrations in drinking waters are given in Table 7.

G. Chlorophenols

This category includes 2-, 3- and 4-chlorophenol (ClC_6H_4OH), 2,3- , 2,4- , 2,5- , 2,6- , 3,4- and 3,5- dichlorophenol ($Cl_2C_6H_3OH$), 2,3,4-, 2,3,5-,

Table 4. Measured concentrations of chloral hydrate in chlorinated drinking water

	Chloral hydrate (μg l^{-1})
United States (Krasner et al., 1989)	
Spring	1.8
Summer	3.0
Fall	2.2
Winter	1.7
Canada (LeBel et al., 1997)	
Yearly mean	3.4
Range	0.6 - 10.5

Table 5. Measured concentrations of haloketones in chlorinated drinking water

	1,1- Dichloroacetone (μg l^{-1})	1,1,1- Trichloroacetone (μg l^{-1})
United States (Krasner et al., 1989)		
Spring	0.52	0.80
Summer	0.46	0.35
Fall	0.52	0.60
Winter	0.55	0.66
Canada (LeBel et al., 1997)		
Yearly mean	0.7	2.4
Range	0.4 - 1.3	<0.1 - 5.7

2,3,6- , 2,4,5- , 2,4,6- and 3,4,5- trichlorophenol ($Cl_3C_6H_2OH$) and pentachlorophenol (Cl_5C_6OH).

According to EPA, 2-chlorophenol, 2,4-dichlorophenol and 2,4,6-trichlorophenol belong to Priority Toxic Pollutants and Hazardous Wastes. Additional lifetime cancer risk is 1/100000 at a concentration of 12 μg l^{-1}.

Measurements of chlorophenol concentrations in drinking waters are given in Table 8.

REGULATION OF DISINFECTION BY-PRODUCTS

In November 1979, USEPA set a Maximum Contaminant Level (MCL) for total THMs of 0.100 mg l^{-1} as an annual average (USEPA, 1979). In November 1992, a Regulatory Negotiation Process started in order to ensure the microbiological safety of drinking water and simultaneously reduce the risk deriving from exposure to disinfectants and DBPs. For this goal to be achieved, a large amount of necessary information was not available. Nevertheless, the negotiations resulted in the Disinfectants/DBPs Stage 1 rule proposed by USEPA in 1994. According to this rule (USEPA, 1998a):

- the MCL for total THMs was lowered from 0.100 to 0.080 mg l^{-1}
- a MCL of 0.060 mg l^{-1} for the sum of concentrations of five haloacetic acids (monochloro-, dichloro-, trichloro-, monobromo- and dibromo-acetic acid) was added.

Epidemiological studies published after the 1994 proposed rule were evaluated by EPA (McGeehin et al., 1993; Vena et al., 1993; King et al., 1996; Doyle et al., 1997; Freedman et al., 1997; Cantor et al., 1998; Hildesheim et al., 1998). Better evidence for an association between exporure to chlorinated water and cancer derives from bladder cancer studies than from colon and rectal cancer studies (USEPA, 1998b). EPA estimates that an upper bound range of 1100-9300 potential bladder cancer cases per year could be associated with exposure to DBPs in chlorinated surface water (USEPA, 1998a). Several studies of association of possible reproductive and developmental effects with exposure to DBPs in chlorinated water have also been published (Savitz et al., 1995; Kanitz et al., 1996; Klotz and Pyrch, 1998; Swan et al., 1998; Waller et al., 1998). EPA continues an epidemiology and toxicology research program to this direction and is considering (USEPA, 1997; USEPA, 1998 a,c):

- establishing a Maximum Contaminant Level Goal (MCLG) of zero for $CHCl_3$,

Table 6. Measured concentrations of haloacetonitriles in chlorinated drinking water

	$CHCl_2CN$ (μg l^{-1})	CCl_3CN (μg l^{-1})	CHClBrCN (μg l^{-1})	$CHBr_2CN$ (μg l^{-1})
United States (Krasner et al., 1989)				
Spring	1.2	<0.012	0.50	0.54
Summer	1.1	<0.012	0.58	0.48
Fall	1.1	<0.029	0.70	0.51
Winter	1.2	<0.029	0.59	0.46
Canada (LeBel et al., 1997)				
Yearly mean	2.2	<0.1	0.2	<0.1
Range	0.4 - 5.2	<0.1	<0.1 - 0.5	<0.1

Table 7. Measured concentrations of chloropicrin in chlorinated drinking water

	Chloropicrin (μg l^{-1})
United States (Krasner et al., 1989)	
Spring	0.16
Summer	0.12
Fall	0.10
Winter	0.10
Canada (LeBel et al., 1997)	
Yearly mean	0.3
Range	<0.1 - 0.5
Utah (Nieminski et al., 1993)	
Yearly mean (for 14 water utilities)	0.10 - 0.61

Table 8. Measured concentrations of 2,4,6 -trichlorophenol in chlorinated drinking water

	2,4,6 -Trichlorophenol (μg l^{-1})
Utah (Nieminski et al., 1993)	
Yearly mean (range for 14 water utilities)	0.52 - 1.10
California (Fu et al., 1994)	
Yearly mean (simulated distribution system)	1.96

$CHCl_2Br$ and $CHBr_3$ due to sufficient evidence of carcinogenicity in animals

- establishing a MCLG of 0.06 mg l^{-1} for $CHClBr_2$, due to liver toxicity and limited evidence of carcinogenicity in animals.

Based on the International Life Sciences Institute panel report in 1997 (ILSI, 1997), EPA is considering changing the MCLG for $CHCl_3$ from zero to 0.3 mg l^{-1} and leaving the MCLG for dichloroacetic acid at zero, because it is considered as a "likely" cancer hazard to humans (USEPA, 1998a,d). Parametric values for THMs, based on the additional lifetime cancer risk 1/1000000, have also been set by the European Commission in the COM (94) 612 Council Directive, adopted in 1995. The latter derived from revision of the 80/778/EEC Directive about water quality, in which values for THM concentrations in water had not been mentioned. The values set in the revised Directive are 40 and 15 μg l^{-1} for $CHCl_3$ and $CHCl_2Br$ (Premazzi et al., 1997). In the interests of operating convenience the parametric value for dibromochloromethane can be increased to 25 μg l^{-1} provided that the parametric value for chloroform is reduced to 30 μg l^{-1}.

Qualitative target levels for DBPs have been set by the World Health Organization WHO as well, based on the additional cancer risk of 1/100000, and are presented in Table 9.

TECHNOLOGIES FOR DBP CONTROL

A. Precursor Removal

Enhanced coagulation. Total organic carbon concentration and alkalinity of the water affect the effectiveness of this technique. When the alkalinity is low, alum addition may be needed in order to lower the pH and achieve more effective coagulation of NOM. When alkalinity is high, excessive amount of alum is demanded or an acid (sulfuric) may be needed. The hydrophobic/hydrophilic distribution of TOC plays an important role, since hydrophobic organic carbon is more susceptible to coagulation than hydrophilic organic carbon (Singer and Harrington, 1993).

Granular activated carbon adsorption. It is a relatively expensive process. In most cases a separate postfiltration bed is needed. Empty-bed contact times above 15 min are required and regeneration frequencies are between three and six months. Decreasing pH of water or increasing alum dosages during pretreatment increase effectiveness of the method (Semmens et al., 1986).

Membrane filtration. It is a relatively expensive process. Nanofilters with membranes having molecular weight cutoffs of 200-500 daltons are needed (Laine et al., 1993).

Powder activated carbon adsorption. Dosage and contact time are the main factors affecting the efficiency of the method. An increase beyond 60 min in contact time or beyond 30 mg l^{-1} in dosage is not convenient (Sandrucci et al., 1995).

Use of activated carbon adsorption and membrane filtration, especially when control of pesticide contamination is needed as well, has been reported in the Netherlands (Premazzi et al., 1997). Preozonation on the basis of low doses in order to enhance flocculation is a new technique being developed (Premazzi et al., 1997).

B. Alternative disinfectants

Monochloramine. It does not produce appreciable amounts of known DBPs, except for DCA and cyanogen chloride (Cowman and Singer, 1994). However, it is a poor primary disinfectant and oxidant and not effective for taste and odor control.

Ozone. It is the most effective oxidant and disinfectant, but since molecular ozone is unstable, a persistent disinfectant residual is not available. Ozone reacts with NOM, forming the following categories of by-products: aldehydes, aldo- and ketoacids, hydrogen peroxide. When bromide is present, brominated DBPs such as bromoform, brominated acetic acids, brominated acetonitriles are formed (Singer, 1994).

Chlorine dioxide. It is an effective oxidant and disinfectant and appropriate for taste and odor control. It does not produce significant concentrations of halogenated DBPs, except for chlorite. However, when chlorine dioxide reacts with NOM, oxidation by-products similar to those formed by ozone are produced (Singer, 1994).

Permanganate. It is an effective oxidant and suitable for taste and odor control, but a poor disinfectant. Additionally, it leads to formation of $MnO_{2(s)}$, which is insoluble and creates operational problems at the treatment plant (Singer, 1994).

UV light. It is an effective disinfectant (Wolfe, 1990), but requires low-turbidity water with a low concentration of UV absorbing substances (groundwater or filtered surface water). Residual is not available.

Table 9. Qualitative target levels of DBPs (WHO, 1995)

DBP	Target level (200μg l^{-1})
2,4,6 Trichlorophenol	
$CHBr_3$	100
$CHClBr_2$	100
$CHCl_2Br$	60
$CHCl_3$	200
DCA	50
TCA	100
DCAN	90
DBAN	100
TCAN	1

Ozone and chlorine dioxide have been widely used as alternative disinfectants in the United States and in Europe (Richardson et al., 1994; Singer, 1994). The same is true for the use of monochloramine as a preoxidant (USEPA, 1998e).

Combinations of technologies have been considered, such as ozone-monochloramine, hydrogen peroxide-ozone, UV-ozone, UV-hydrogen peroxide, in order to produce highly reactive short-lived free radical species which can oxidize contaminants. It has also been shown (Richardson et al., 1996) that TiO_2/UV photocatalysis can kill microorganisms and simultaneously lead to degradation of chlorinated organic compounds and DBP precursors. However, this technique does not provide a disinfectant residual. Further research is necessary in order to determine the effectiveness of each technology and the related chemical mechanisms.

C. DBP removal

Air stripping. DBPs, which have already been formed, can be removed with the methods of packed column air stripping (packed towers) or diffused air stripping (compressed air). Application of air stripping in Italy has been reported (Premazzi et al., 1997). Air contamination or residual disinfectant removal could be the main negative points of these techniques.

Reverse osmosis. This method can remove 85-90% of all organic compounds. As membrane technology improves and costs decrease, the procedure seems more attractive for DBP removal (Premazzi et al., 1997).

Granular activated carbon. With this technique many categories of organic compounds can be removed. However, regular maintenance is necessary and microbiological contamination might take place (Premazzi et al., 1997).

SUMMARY AND CONCLUSIONS

Chlorination of drinking water leads to the formation of a variety of disinfection by-products that may have adverse health effects on humans. Relative data is still limited, especially for the non-trihalomethane DBPs: haloacetic acids, haloaldehydes, haloketones, haloacetonitriles, chloropicrin and chlorophenols. The objective of minimizing both risks of microbial disease and exposure to DBPs in drinking water is a critical point. In order to solve this problem further research is required, based on chemistry related to DBP formation, their physical and chemical properties and environmental fate. Relative information is given in this review. Reference to actual measurements of DBPs in different countries has been made and proposed changes in regulation of DBPs by EPA and WHO have been presented. Epidemiological and toxicological studies are still continued so that the adverse health effects of DBPs on humans can be specified. Technologies for controlling DBPs in drinking water have also been summarized. Extensive studies of the chemical mechanisms occurring in each case are needed in order to determine the most effective DBP controlling technology or even creating a new one.

SYMBOLS

C_{Br}	Bromide ion concentration, mg l^{-1}
C_{CHBrCl_2}	Bromodichloromethane concentration, μg l^{-1}
C_{CHBr_2Cl}	Bromochloromethane concentration, μg l^{-1}
C_{CHCl_3}	Chloroform concentration, μg l^{-1}
C_{chla}	Chlorophyll-a concentration, mg m^{-3}
C_{cl_2}	Chlorine dose, mg l^{-1}
C_{DCAA}	Dichloroacetic acid concentration, μg l^{-1}
$C_{Org\text{-}N}$	Organic nitrogen concentration, μg l^{-1}
C_{TCAA}	Trichloroacetic acid concentration, μg l^{-1}
C_{TTHM}	Total trihalomethane concentration, μmol l^{-1}
$Act_{Ar\text{-}R}$	Activated aromatic content, mM (g C)$^{-1}$
DCANFP	Dichloroacetonitrile formation potential, μg l^{-1}
TCAAFP	Trichloroacetic acid formation potential, μg (mg of C)$^{-1}$
THMFP	Trihalomethane formation potential, μg (mg of C)$^{-1}$
TOC	Total organic carbon concentration, mg l^{-1}
UV_{254}	Ultraviolet radiation absorption at 254 nm wavelength, cm^{-1}
t	Time, h
T	Temperature, °C
S, Sp	Summer, Spring (pseudo variables which expresses the season infuence: S=1 for samples collected in summer, S=0 for samples collected during other seasons, Sp=1 for samples collected in spring, Sp=0 for samples collected during the other seasons)
k_1	Function of pH (Table 1)
k_2	Function of temperature (Table 1)
z	Function of temperature (Table 1)

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Annex I. *Physical - chemical properties and environmental fate of Disinfection By-Products*

Compound	M W	Physical characteristics (^{o}C)	Boiling point (^{o}C)	Melting point	Soluble in	Water Solubility	Vapor Pressure	Reactions in water
Chloroform	119.39	Clear, colorless liquid with a characteristic odor	61.7	-63.5	Organic solvents	7950 mg l^{-1} at 25 ^{o}C	246 mm Hg at 25 ^{o}C	Evaporation Biodegradation
Dichlorobromomethane	163.83	Liquid	90	-57.1	Organic solvents	4700 mg l^{-1} at 25 ^{o}C	50 mm Hg at 25 ^{o}C biodegradation	Evaporation Anaerobic
Dibromochloromethane	208.28	Clear, colorless liquid	119-120	-20	Organic solvents	4400 m l^{-1} at 25 ^{o}C	15 mm Hg at 10.5 ^{o}C biodegradation	Evaporation Anaerobic
Bromoform	252.77	Colorless - yellow liquid with chlorofomlike odor	149.5	7.70	Organic solvents	3974 mg l^{-1} at 20 ^{o}C	7.36 mm Hg at 25 ^{o}C	Evaporation Biodegradation
Monochloroacetic acid (MCA)	94.50	Colorless - white crystalline solid	188 for α-, 187.9 for β- and 187.8 for γ- isomer	63 for α-, 56.2 for β- and 52.5 for γ-isomer	Water Ether Benzene Alcohols	1.09 x 106 mg l^{-1} at 25 ^{o}C	-	-
Dichloroacetic acid (DCA)	128.94	Liquid with pungent odor	194	13.5	Water Acetone Ether Alcohols	Miscible	-	-
Trichloroacetic acid (TCA)	163.39	Colorless, crystalline solid with characteristic odor, very corrosive	197.5	58 for α- and 49.6 for β-isomer	Ether Alcohols	1.50 x 106 mg l^{-1} at 25 ^{o}C	-	-
Monobromoacetic acid (MBA)	138.95	-	208	50	Water Alcohols Ethers Acetone Benzene	1.75 x 106 mg l^{-1} at 25 ^{o}C	-	-
Dibromoacetic acid (DBA)	217.84	Crystalline solid	195	48	Water Ether Alcohols	2.11 x 106 mg l^{-1} at 25 ^{o}C	-	-
Tribromoacetic acid (TBA)	296.78	Crystalline solid	245	135	Water Ether Alcohols	2.00 x 106 mg l^{-1} at 25 ^{o}C	-	-
Bromochloroacetic acid (BCA)	173.39	-	103-104	38	Water Ether Acetone Alcohols	-	-	-
Monochloroacetonitrile	75.50	-	126-127	-	Ether Alcohols	-	-	-
Dichloroacetonitrile	109.94	-	112-113	-	Alcohols	-	-	-
Bromoacetonitrile	119.95	Pale yellow color	150-151	-	Ether	-	-	-
Trichloroacetonitrile	144.39	Liquid	84.6	-42	-	-	-	-
1,1 - Dichloroacetone	126.98	Oily liquid	120	-	Acetone	Very low	-	-
1,3 - Dichloroacetone	126.98	-	173	45	Water Ether Alcohols	-	-	-
Chloroacetaldehyde	78.5	Clear, colorless liquid with pungent odor	142-144	87	Ethers	7x106 mg l^{-1} at 25 ^{o}C	317 mm Hg at 25 ^{o}C	-
Dichloroacetaldehyde	112.94	-	90-91	-	Alcohols	-	-	-

Annex I. *Physical - chemical properties and environmental fate of Disinfection By-Products* (continued)

Compound	M W	Physical characteristics (^{o}C)	Boiling point (^{o}C)	Melting point	Soluble in	Water Solubility	Vapor Pressure	Reactions in water
Trichloro-acetaldehyde	147.40	Oily liquid with pungent, irritating odor	-	129-135	Water Ether Alcohols	-	-	-
Tribromo-acetaldehyde	280.78	Yellowish, oily liquid	174	-	Water Ether Alcohols	-	-	-
Chloral hydrate	165.42	Pungent odor, caustic taste	98	57	Acetone	-	-	-
Chloropicrin	164.5	Colorless, oily liquid, with pungent odor, causes tears	112	-69.2	Ether Benzene Alcohols	1621 mg l^{-1} at 25 ^{o}C	23.8 mm Hg at 25 ^{o}C	-
2 - Chloro-phenol	128.56	Liquid	175-176	9	Ether Benzene Alcohols	28000 mg l^{-1} at 25 ^{o}C	1.42 mm Hg at 25 ^{o}C	Adsorption on sediments Photolysis Biodegradation
3 - Chloro-phenol	128.56	-	214	33	Ether Benzene Alcohols	26000 mg l^{-1} at 25 ^{o}C	0.119 mm Hg at 25 ^{o}C	Biodegradation Photolysis Adsorption on sediments Evaporation
4 - Chloro-phenol	128.56	-	220	43	Ether Benzene Alcohols	27000 mg l^{-1} at 25 ^{o}C	0.087 mm Hg at 25 ^{o}C	Adsorption on sediments Photolysis Biodegradation
2,3 - Dichloro-phenol	163.00	-	-	57-59	Ether Alcohols	-	-	-
2,4 - Dichloro-phenol	163.00	Colorless, crystalline solid	210	45	Ether Benzene Alcohols	4500 mg l^{-1} at 25 ^{o}C	0.118 mm Hg at 25 ^{o}C	-
2,5 - Dichloro-phenol	163.00	-	211	59	Ether Benzene Alcohols	-	-	-
2,6 - Dichloro-phenol	163.00	White, crystalline solid	219-220	68-69	Ether Benzene Alcohols	172 mg l^{-1} at 25 ^{o}C	0.0165 mm Hg at 25 ^{o}C	-
3,4 - Dichloro-phenol	163.00	-	253.5	68	Ether Benzene Alcohols	-	-	-
3,5 - Dichloro-phenol	163.00	-	233	68	Ether Alcohols	-	-	-
2,3,4 - Trichlo-rophenol	197.45	Liquid	-	83.5	Ether Benzene Alcohols	-	-	-
2,3,5 - Trichlo-rophenol	197.45	-	248-249	62	Ether Alcohols	-	-	-
2,3,6 - Trichlo-rophenol	197.45	-	-	58	Ether Benzene Alcohols	-	-	-
2,4,5 - Trichlo-rophenol	197.45	Strong phenolic odor	253	57	Alcohols	982 mg l^{-1} at 25 ^{o}C	0.0496 mm Hg at 25 ^{o}C	Photolysis Adsorption on suspended solids and sediments
2,4,6 - Trichlo-rophenol	197.45	Strong phenolic odor	246	69	Ether Alcohols	800 mg l^{-1} at 25 ^{o}C	0.0149 mm Hg at 25 ^{o}C	Photolysis Biodegradation Adsorption on sediments Evaporation
3,4,5 - Trichlo-rophenol	197.45	-	271-277	101	Ether	-	-	-