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# Emerging disinfection byproducts: a review on their occurrence and control in drinking water treatment processes

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## Abstract

The occurrence of disinfection byproducts (DBPs) is related both to drinking water treatment (DWT) processes and to raw water's characteristics. Emerging pollutants typically occur in low concentrations and are not removed by conventional DWT processes. Emerging DBPs appear within the DWT or in the distribution system due to the combination of disinfection agents (especially chlorine) with precursors as: natural organic matter (NOM), algal organic matter (AOM), anthropogenic contaminants (pesticides, pharmaceuticals, detergents etc.), brominated and iodinated compounds.

This study has as main goal a consistent analysis of the major problems caused by emerging DBPs to drinking water supplies. It presents a comprehensive review of the research efforts related to emerging DBPs considering three viewpoints: 1. an overview of their classification, legislative framework, methods of analysis, disinfection operational

27 conditions and removal processes; 2. their occurrence, fate, health effects and impacts; 3.  
28 the analysis of the advanced DWT processes that might be used for the removal and control  
29 of precursors and DBPs with a focus on pilot and full-scale installations. All presented case  
30 studies considered pollutants removed, process conditions and efficiencies, and a critical  
31 assessment of processes based on membranes, advanced oxidation and adsorption on  
32 activated carbon or other materials. The main challenges of the control and removal of  
33 emerging DBPs are their low concentrations and the technical and economic sustainability of  
34 the application at full-scale of the AOPs, which need to be carefully adapted to local  
35 boundary conditions.

36

37 *Keywords:* disinfection byproducts, emerging pollutants, drinking water, water treatment  
38 process

39

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50

## 51 List of abbreviations

Abbreviations/ Chemical formulas	Complete name
ADWT	Advanced Drinking Water Treatment
AOM	Algal Organic Matter
AOPs	Advanced Oxidation Processes
BCAA	Bromochloroacetic acid
BDCAA	Bromodichloroacetic acid
BDCM	Bromodichloromethane
Br-DBPs	Brominated-Disinfection byproducts
C/F	Coagulation/Flocculation
C-DBPs	Carbonaceous disinfection byproducts
CHCl <sub>3</sub>	Chloroform
Cl-DBPs	Chlorinated-Disinfection byproducts
DBAN	Dibromoacetonitrile
DBCM	Dibromochloromethane
DBPs	Disinfection byproducts
DCAA	Dichloroacetic acid
DCAcAm	2,2-dichloroacetamide
DCAN	Dichloroacetonitrile
DWT	Drinking Water Treatment
EDCs	Endocrine Disrupting Chemicals
EPs	Emerging pollutants
GAC	Granular Activated Carbon
GC-ECD	Gas Chromatography- Electron Capture Detector
GC-MS	Gas Chromatography- Mass Spectrometry
HAA5	Haloacetic Acids

Abbreviations/ Chemical formulas	Complete name
HNM	Halonitromethane
HPLC	High-Performance Liquid Chromatography
HS-SPME	Solid-phase Microextraction in the Headspace
IC-MS/MS	Ion Chromatography-tandem Mass Spectrometry
I-DBPs	Iodinated disinfection byproducts
IER	Ion-exchange Resin
LLE-ETAC	Liquid-Liquid Extraction with Ethyl Acetate
MCAA	Monochloroacetic acid
MCL	Maximum Concentration Level
MLLE	Micro Liquid-Liquid Extraction
N-DBPs	Nitrogenous disinfection byproducts
NDMA	N-nitrosodimethylamine
NOM	Natural Organic Matter
PAC	Powdered Activated Carbon
PIE	Precursor Ion Elimination
TBAA	Tribromoacetic acid
TCAA	Trichloroacetic acid
TCAcAm	Trichloroacetamide
TCAN	Trichloroacetonitrile
TCNM	Trichloronitromethane
THM	Trihalomethane
TTHM	Total Trihalomethanes
UPLC	Ultra-Performance Liquid Chromatography

52

## 53 1. Introduction

54 A major accomplishment for human health is the production of safe drinking water from raw  
 55 surface and ground waters. Drinking water sources have thus become a priority for  
 56 environmental communities worldwide. Water disinfection plays a pivotal role in reducing  
 57 serious illnesses associated with waterborne diseases. Disinfection is used to eliminate  
 58 pathogenic microorganisms during the drinking water treatment (DWT) and to ensure in the  
 59 distribution network the quality parameters for a safe drinking water consumption (Chau et  
 60 al., 2015). DWT plants' managers and water suppliers must assume the responsibility for  
 61 water safety and at the same time for the implementation of the most efficient technologies  
 62 to warrant that all qualitative indicators are below the limits imposed by the regulations.  
 63 Most common chemical disinfectants are chlorine-based (e.g. chlorine, chlorine dioxide,  
 64 hypochlorite salts), having the advantages of low cost and easy manipulation, high efficiency

65 towards different pathogens, taste improvement and persistence in the distribution network.  
66 However, disinfection byproducts (DBPs) often occur after chlorine-based disinfection  
67 processes (Gupta and Ali, 2013), implying possible adverse effects and risks to human  
68 health. Many precursors, as natural organic matter (NOM), algal organic matter (AOM),  
69 anthropogenic contaminants (e.g. pesticides, pharmaceuticals, detergents, etc.), brominated  
70 and iodinated compounds, as well as upstream wastewater discharges, and DWT  
71 operational parameters (disinfection agent type and/or dose, pH, contact time, temperature)  
72 may contribute to the development of DBPs (Alexandrou et al., 2018). Precursors' presence  
73 and amount could be subjected to seasonal variations (e.g. for AOM or contaminants related  
74 to agricultural activities, as ammonium and pesticides). DBPs are classified "harmful for  
75 human health" and their occurrence in raw water sources imposes special monitoring and  
76 efforts for the water suppliers (WHO, 2017).

77 In order to avoid DBPs formation, new disinfection processes and technologies were  
78 developed (e.g. ozone, ultraviolet, silver ion, electrochlorination, ferrate), involving high costs  
79 related to equipment and energy consumption (Zainudin et al., 2018). However, DBPs  
80 occurrence was demonstrated also for disinfection processes using non-chlorinated reagents  
81 (Ding et al., 2019).

82 Many members of the drinking water protection community have been actively working to  
83 clearly understand the possible negative effects of DBPs on human health. Meanwhile, state  
84 and federal governments have taken steps to protect the public from the potential health  
85 risks of DBPs by conducting research on their toxicological effects, strengthening drinking  
86 water regulations and supporting improvements in water treatment technology (Bereskie et  
87 al., 2017). Many authors focused their research on DBPs and disinfection processes under  
88 the following directions: (i) occurrence and removal of DBPs precursors deriving from raw  
89 water sources and optimization of DWT operational parameters and (ii) improved removal of  
90 DBPs and residual microorganisms at the end of DWT process and within the distribution  
91 network. Advanced drinking water treatment (ADWT) technologies are commonly

92 implemented for the removal of emerging pollutants (including DBPs) and to reduce the  
93 concentrations of organic/inorganic precursors (Teodosiu et al., 2018).

94 This study has as main goal a consistent analysis of the major problems caused by  
95 emerging DBPs to drinking water supplies, providing a foundation for future research and  
96 highlighting the strengths and weaknesses of DBPs' control processes. To our knowledge,  
97 most research focused so far on the identification of DBPs and their precursors rather than  
98 on removal technologies. This study has the aim to present a comprehensive review of the  
99 research efforts related to emerging DBPs considering three objectives: 1) an overview of  
100 their classification, legislative framework, methods of analysis, disinfection operational  
101 conditions and removal processes; 2) their occurrence, fate, health effects and impacts; 3) a  
102 critical assessment of the ADWT processes that might be used for the removal and control of  
103 emerging DBPs and their precursors with a specific focus on pilot and full-scale installations.

104

## 105 **2. Methodology**

106 The analysis of the scientific literature considered for this review was based on the following  
107 selection criteria:

108 ✓ *The relevance of articles and international information databases.* This study was based  
109 on 202 documents: articles found in Science Direct, Scopus, Web of Science, Springer,  
110 Wiley Online Library, and reports downloaded from the European Commission or other  
111 international reference databases (162 scientific papers, 23 review papers, 17  
112 books/technical reports/regulations);

113 ✓ *Publication period.* The references are mostly from 2010-2020 (93.77 %);

114 ✓ *Relevant keywords.* The following keywords have been used in different combinations:  
115 *drinking water treatment, disinfection, disinfection byproducts, treatment technologies, pilot*  
116 *scale, full scale, analysis of disinfection by products from drinking water, regulations;*

117 ✓ *Selection of references based on content analysis.* On the grounds of the above-  
118 mentioned criteria, 202 references were finally analysed as abstracts or full text documents.  
119

### 120 **3. Emerging DBPs of concern for drinking water treatment processes**

121 DBPs are formed in drinking water from the reaction of disinfection agents with other  
122 compounds (*precursors*) occurring in raw water as: NOM, bromide and iodide, anthropogenic  
123 compounds (pharmaceuticals, antibacterial agents, textile dyes, pesticides, surfactants and  
124 cyanotoxins, etc.) (Papageorgiou et al., 2016; Barcelo, 2012). In recent years, great efforts  
125 were made to study the fate, occurrence and ecotoxicology of byproducts of drinking water  
126 disinfection processes. During chlorination, a large number and variety of DBPs are formed  
127 (Bond et al., 2014), as trihalomethanes (THMs), haloacetic acids (HAAs), haloacetonitriles,  
128 halophenols and halopropanoles (Farré et al., 2008, Liu et al., 2017; Young et al., 2018).  
129 Some DBPs, like THMs, HAAs, chlorites and bromates, are acknowledged by USEPA, WHO  
130 and international regulations as they are considered to have high impacts on human health.  
131 However, other DBPs (Table 1) are unregulated, even if their presence was detected during  
132 disinfection (with chloramines, chlorine, chlorine dioxide, ozone or ferrate) or if raw water  
133 contains natural bromide (Jiang et al., 2016). Unregulated DBPs are less known and studied,  
134 having very low concentrations, but they can be more toxic for humans and can rapidly  
135 increase the risks for cancer or other diseases (Li et al., 2015).

136

#### 137 *3.1. Definition, classifications and regulations*

138 Most DBPs could be defined *emerging pollutants* (EPs) (see Table 1), according to the  
139 Norman Network classification (Norman Network, 2016). EPs are chemical compounds with  
140 risks to human health, which appear in drinking water as a consequence of disinfection  
141 treatment (Table 1). Over 600 DBPs were identified (Richardson and Postigo, 2011),  
142 however the studies concerning their occurrence and formation mechanisms, human health

143 effects and treatment alternatives (Hebert et al., 2010; Kimura and Ortega-Hernandez, 2019;  
 144 Chaukura et al., 2020; Sorlini et al., 2014) are much less. Usually, DBPs are related to the  
 145 use of chlorine-based disinfectants (chlorine gas, chloramines, chlorine dioxide,  
 146 hypochlorite/hypochlorous acid) or ozone and to the raw water physicochemical features.  
 147 The most common DBPs classes, their sources and health effects are presented in Table 1.

148 **Table 1.** DBPs classes, sources and health effects

DBPs Classes	Recognized as EPs	Sources				Effects	References
		Cl	NH <sub>2</sub> Cl	ClO <sub>2</sub>	O <sub>3</sub>		
<b>Total Trihalomethanes (TTHMs or THM4)</b>							
Bromodichloromethane	yes					Possible carcinogenic for liver, kidney, intestine; central nervous and reproductive system diseases; also causing bladder, colon, rectal, or pancreatic cancer	Hebert et al., 2010; Pardakhti et al., 2011; Kogevinas et al., 2016; Lodhi et al., 2017; Han et al., 2017
Bromoform							
Dibromochloromethane							
Chloroform		x	x	x			
<b>Haloacetic acids (HAAs or HAA5)</b>							
Dichloroacetic acid		x	x			Possible carcinogenic for liver, kidney, lung, causing leukaemia; effects on reproductive system, skin and eyes irritation	Chowdhury et al., 2011; Dad et al., 2018
Trichloroacetic acid							
Chloroacetic acid	yes						
Bromoacetic acid							
Dibromoacetic acid							
<b>Bromate</b>				x	x	Possible carcinogenic for thyroid, kidney and breast	Dongmei et al., 2015; Xiao et al., 2017
<b>Chlorite</b>				x		Anaemia, thyroid gland and kidney diseases; mutagenic and teratogenic effects on nervous system	Feretti et al., 2008; Garcia-Villanova et al., 2010; Chhetri et al., 2017
<b>Chlorate</b>	yes			x		Possible carcinogenic for thyroid; immune system hypersensitivity	Feretti et al., 2008; Hebert et al., 2010; Ibrahim and Abu-Shanab, 2013
<b>Hydrazine</b>	yes		x			Possible effects upon the nervous and cardiovascular system, liver and kidneys; causing vomiting and hepatotoxicity; carcinogenic and with chronic toxicity	Bond et al., 2011; Matsumoto et al., 2016; Guyton et al., 2018;

DBPs Classes	Recognized as EPs	Sources				Effects	References
		Cl	NH <sub>2</sub> Cl	ClO <sub>2</sub>	O <sub>3</sub>		
<b>Nitrosamines (NDMA)</b> N-Nitrosodimethylamine N-Nitrosopyrrolidine N-Nitrosomorpholine N-Nitrosopiperidine N-Nitrosodiphenylamine N-nitrosomethylethylamine N-nitrosodiethylamine N-nitroso-di-n-butylamine	yes	x	x			Possible carcinogenic for liver, stomach, esophagus, bladder, lung, breast and brain	Hebert et al., 2010; Hanigan et al., 2017 Linge et al., 2017;
<b>Aldehydes</b> Chloroacetaldehyde Dichloroacetaldehyde Bromochloroacetaldehyde Tribromoacetaldehyde	yes	x		x	x	Possible carcinogenic for stomach and lung; reproductive system toxicity; causes depression and DNA damage	Hebert et al., 2010; Xie et al., 2016
<b>Unregulated THMs</b> Dibromomethane Bromochloromethane Tetrachloromethane Dibromodichloromethane	yes	x	x			Possible carcinogenic for liver, kidney, intestine; central nervous and reproductive systems diseases;	Hebert et al., 2010 Yang et al., 2019
<b>Unregulated HAAs</b> Bromochloroacetic acid Bromodichloroacetic acid Dibromochloroacetic acid Tribromoacetic acid	yes	x	x			Possible carcinogenic for liver, lungs, kidney and reproductive system diseases; leukemia	Richardson et al., 2007; Hebert et al., 2010; Zhang et al., 2011
<b>Tribromopyrrole/ Halopyrrole</b>	yes	x	x	x	x	Possible carcinogenic for breast	Hebert et al., 2010; Yang and Zhang, 2014
<b>Cyanogen chloride</b>	yes	x	x		x	Possibly causes eyes and skin irritation, respiratory and cardiovascular deficiencies; may be lethal at high concentration	Hebert et al., 2010; Weng and Blatchley, 2013;
<b>Haloanisoles</b> 2,4,6-trichloroanisole 2,4,6-tribromoanisole 2,4-dibromoanisole 2,4-dichloroanisole 2,6-dichloroanisole 2-bromoanisole 4-bromoanisole	yes	x				N/A	Hebert et al., 2010; Barceló and Alastuey, 2012; Xie et al., 2016
<b>Chlorophenols</b> 2-chlorophenol 2,4-Dichlorophenol 2,4,6-Trichlorophenol 2,3,4,6-Tetrachlorophenol 2,4,5-Trichlorophenol 4-chlorophenol	yes	x	x			Possible carcinogenic for liver, lymphoma and lungs; immune system hypersensitivity; hepatic and renal toxicity	Gopal et al., 2007; Hebert et al., 2010; Igbinosa et al., 2013; Zhai et al., 2014
<b>Haloamides</b> Dichloroacetamide Bromochloroacetamide Dibromoacetamide Bromiodoacetamide Trichloroacetamide Diiodoacetamide Tribromoacetamide	yes	x	x			Possible carcinogenic effects on reproductive system, liver or leukemia	Hebert et al., 2010; Bull et al., 2011; Shah et al., 2012

DBPs Classes	Recognized as EPs	Sources				Effects	References
		Cl	NH <sub>2</sub> Cl	ClO <sub>2</sub>	O <sub>3</sub>		
<b>Halonitromethanes</b> Chloronitromethane Bromonitromethane Dichloronitromethane Dibromonitromethane Bromochloronitromethane Trichloronitromethane Dibromochloronitromethane Tribromonitromethane	yes	x	x	x	x	Possible carcinogenic for breast	Liviac et al., 2009; Hebert et al., 2010; Ceretti et al., 2016;
<b>Haloacetonitriles (HANs)</b> Chloroacetonitrile Bromoacetonitrile Dichloroacetonitrile Bromochloroacetonitrile Trichloroacetonitrile Iodoacetonitrile Dibromoacetonitrile	yes	x	x	x	x	Reproductive system toxicity; possible carcinogenic for breast	Richardson et al., 2007; Hebert et al., 2010; Ileka-Priouzeau et al., 2015;
<b>Haloketones</b> Hexachloropropanone 1,1-Dibromopropanone 1,1,3-Trichloropropanone 1,1,1,3-Tetrachloropropanone 1,1,3,3-Tetrachloropropanone 1,1,1,3,3-Pentachloropropanone	yes	x	x			Possible carcinogenic or mutagenic effects	Hebert et al., 2010; Linge et al., 2013; Zhai et al., 2014
<b>Iodoacids</b>	yes		x			Possible cytotoxic and genotoxic effects, causing breast cancer	Hebert et al., 2010; Xiao et al., 2016
<b>IodoTHMs</b> Dichloroiodomethane Bromochloroiodomethane Bromodiiodomethane Dibromoiodomethane Chlorodiiodomethane Iodoform	yes	x	x	x		Possible genotoxic and cytotoxic effects, causing bleeder cancer and endocrine diseases	Hebert et al., 2010; Luo et al., 2014; Han et al., 2017
<b>HEX &amp; TEX</b> Hexachlorocyclopentadiene Tetrachlorocyclopentadiene	yes		x			Possible carcinogenic for liver, causing skin and eyes irritation	Hebert et al., 2010; TOXNET, 2018
<b>MX &amp; halofuranones</b> Mutagen X Mucochloric acid	yes	x	x	x		Possible carcinogenic for thyroid, bile duct, pancreas and lungs	Hebert et al., 2010; Richardson and Postigo, 2016

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**Note:** N/A – not applicable/not available

152 Apart of the identification of DBPs and their adverse health effects, international regulations  
153 and guidelines also defined maximum amounts of DBPs allowed in drinking water. The  
154 World Health Organization (WHO) introduced Drinking Water Guidelines with maximum  
155 contaminant levels for DBPs (WHO, 2006; WHO, 2008; WHO, 2017). The Environmental  
156 Protection Agency (USEPA, 2009) regulates DBPs like THMs, HAAs, bromate and chlorite.  
157 USEPA specifically regulates five HAAs, defined HAA5 (Table 1). In Europe, only THM4

158 (Table 1) and bromate are regulated as DBPs through the Drinking Water Directive, but  
 159 there are standards and guidelines for each species of THMs (Karanfil et al., 2008). Wang et  
 160 al. (2015) compared the maximum contaminant levels (MCL, expressed as  $\mu\text{g L}^{-1}$ ) for  
 161 regulated DBPs in drinking water according to international regulations and guidelines (Table  
 162 2). However, MCL values do not characterize DBPs from the point of view of toxicity,  
 163 carcinogenicity and occurrence (Hebert et al., 2010).

164

165 **Table 2.** Maximum contaminant levels for DBPs as in current directives and guidelines  
 166 (adapted from Wang et al., 2015)

DBPs	MCL ( $\mu\text{g L}^{-1}$ )							
	WHO guidelines (a)	USA Guidelines (b)	Canadian Guidelines (c)	Australian guidelines (d)	EU Directive (e)	Japanese standards (f)	Chinese regulations (g)	Egyptian standards (k)
TTHM(h)	<b>1000</b>	<b>80</b>	<b>100</b> LRAA(i)	<b>250</b>	<b>100</b>	<b>100</b>	<b>1000</b>	<b>100</b>
Cloroform(l)	<b>300</b>	-	-	-	-	<b>60</b>	<b>60</b>	<b>1000</b>
BDCM (n)	<b>60</b>	-	<b>16</b>	-	-	<b>30</b>	<b>60</b>	<b>500</b>
DBCM (n)	<b>100</b>	-	-	-	-	<b>100</b>	<b>100</b>	<b>500</b>
Bromoform (l,n)	<b>100</b>	80	-	-	-	<b>90</b>	<b>100</b>	<b>200</b>
HAA5(j)	<b>60</b>	<b>60</b>	<b>80</b> LRAA	-	-	-	-	-
MCAA	20	-	-	<b>150</b>	-	<b>20</b>	-	-
DCAA	50	-	-	<b>100</b>	-	<b>30</b>	<b>50</b>	-
DCAN(k)	20	-	-	N.D.	-	-	-	<b>1000</b>
DBAN	70	-	-	N.D.	-	-	-	<b>1000</b>
TCAA	200	-	-	<b>100</b>	-	<b>30</b>	<b>100</b>	-
TCAN	-	-	-	-	-	-	-	<b>100</b>
CH (k)	-	-	-	<b>100</b>	-	-	<b>10</b>	<b>200</b>
Bromate (l,p)	10	<b>10</b>	<b>10</b>	20	<b>10</b>	<b>10</b>	<b>10</b>	-
Formaldehyde	-	-	-	<b>500</b>	-	<b>80</b>	<b>900</b>	-
Chlorite (l,m)	700	<b>1000</b>	<b>1000</b>	800	-	-	<b>700</b>	-
Chlorate (m)	700	700	<b>1000</b>	N.D.	-	-	<b>700</b>	-
CHCl (as CN) (l)	-	-	-	80	-	<b>10</b>	<b>70</b>	-
2-chlorophenol (o)	-	-	-	<b>300</b>	-	-	-	-
2,4-dichlorophenol (o)	-	-	<b>900</b>	<b>200</b>	-	-	-	-
2,4,6-dichlorophenol (o)	200	-	2	<b>20</b>	-	-	<b>200</b>	-
2,3,4,6-Tetrachlorophenol(o)	-	-	<b>100</b>	-	-	-	-	-
NDMA (nitrosamines)	0.1	-	<b>0.04</b>	0.1	-	-	-	-
Chloropicrin (q)	-	-	-	-	-	-	-	<b>500</b>
1,1,1-Trichloropropane (q)	-	-	-	-	-	-	-	<b>500</b>

167 **Notes and abbreviations:** (a) World Health Organization guidelines – (WHO, 2017); (b) United States of  
 168 America guidelines– (US EPA, 2006); (c) Canadian Guidelines 2019 - (Canadian DWQ, 2019); (d) Australian  
 169 guidelines – (NWQMS, 2016); (e) Drinking Water Directive of European Council 2012 - (Directive 98/83/EC,  
 170 1998); (f) Ministry of Health, Labour and Welfare (Japanese Standards, 2016, JWVA, 2016); (g) Chinese  
 171 regulation (GB 5749-2006, 2007); (h) TTHM - Sum of all THMs contaminants; (i) LRAA - Local Running Annual  
 172 Average; (j) HAA5 - Haloacetic Acids (k) Ibrahim and Abu-Shanab, 2013; (l) Richardson, 2005; (m) Al-Otoum  
 173 et al., 2016; (n) Pérez Pavón et al., 2008; (o) Hebert et al., 2010; (p) Winid, 2015; (q) Ali, 2013

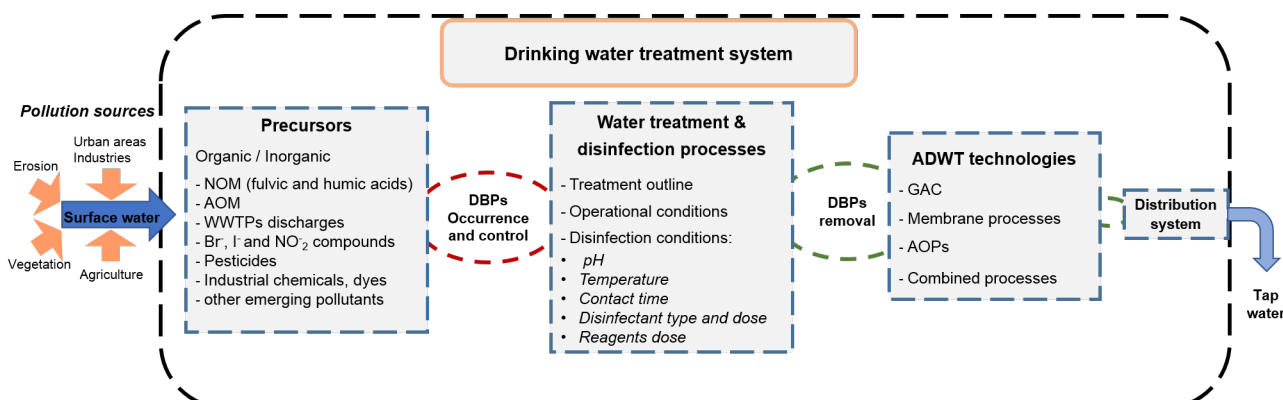
174 **Explanation:** Values in bold refer to DBPs included in the international guidelines, regulations, standards and  
 175 directives specified in brackets from point (a) to (g) and (k). Notes in brackets as (h), (i) and (j) are  
 176 abbreviations explained. The rest are references for DBPs not included in the international  
 177 regulations/guidelines.

178

179 3.2. DBPs sources and occurrence

180 To state the importance of the disinfection processes in providing safe drinking water for  
 181 human consumption, the disinfectants used, with their main characteristics and effectiveness  
 182 should be considered, along with the byproducts formed and consequent health risks. Figure  
 183 1 presents the sources and technologies that may remove both precursors and already  
 184 formed-DBPs, these technologies being largely influenced by the outline of the DWT process  
 185 scheme and of the distribution system.

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Fig. 1. Disinfection byproducts in a DWT system – from occurrence to removal.

190 DBPs formation mechanisms are strongly influenced by operational parameters such as:  
 191 type of disinfectant used, dose and contact time, and by the physico-chemical features of  
 192 raw water (pH, temperature, NOM, ammonium, metals) (Shahi et al., 2019; Jiang et al.,  
 193 2018; Gougoutsa et al., 2016; Ibrahim and Abu-Shanab, 2013; Wei et al., 2010). Raw water  
 194 characteristics are crucial, and many substances were identified as DBPs precursors: NOM  
 195 (Sillanpää, 2014; Neale et al., 2019), pharmaceuticals (Ternes and von Gunten, 2010; Zhou  
 196 et al., 2016), antibacterial agents (Barceló, 2012), textile dyes (Alves de Lima et al., 2007),  
 197 pesticides (Duirk and Collette, 2006; Mehrsheikh et al., 2006), bisphenol A (Hu et al., 2002),  
 198 alkylphenol ethoxylate surfactants (Petrovic et al., 2010) and cyanotoxins ( Lee et al., 2017;  
 199 Corbe et al., 2014 ). It is more effective to remove DBPs precursors before disinfection or to  
 200 control its operational parameters than removing the already formed DBPs (Lin et al., 2016).  
 201 NOM (especially humic substances), AOM, bromide, iodide and anthropogenic pollutants

202 have been widely investigated in raw water sources as precursors (López-Roldán et al.,  
203 2016; Chaukura et al., 2020).

204 Water sources are often affected by industrial discharges, agriculture runoffs, algal blooms,  
205 municipal wastewater discharges, storm water runoffs, wildfires and elevated bromine and  
206 iodine concentration, each with high or moderate impact on DBPs occurrence (Sgroi et al.,  
207 2018). Pharmaceuticals reach drinking water due to inadequate wastewater treatment,  
208 exaggerated human consumption and veterinary use, even through leaks from the  
209 agricultural lands. Even if they are present in low concentrations, pharmaceuticals can resist  
210 conventional DWT (e.g. flocculation, sedimentation, filtration) and chemical disinfection  
211 (through chlorine, chloramines, ozone or chlorine dioxide) (Kaplan, 2013) and produce DBPs  
212 (Postigo and Richardson, 2014; Kimura and Ortega-Hernandez, 2019). Advanced DWT  
213 systems that use UV or UV/H<sub>2</sub>O<sub>2</sub> disinfection processes can interact with pharmaceutical  
214 substances and form NDMA or increased levels of DBPs after the addition of chloramine  
215 (Radjenovic et al., 2012; Postigo and Richardson, 2014).

216 Disinfectants like chlorine, chlorine dioxide, chloramine, sodium hypochloride or ozone  
217 interact with NOM, microorganisms or bromide/iodide to produce a different class of DBPs  
218 with a high toxicity (Zhao et al., 2012; Jiang et al., 2018).

219 In the following subsections an overview of the main disinfection agents used in DWT  
220 processes and the generated DBPs will be presented.

### 221 3.2.1. Chlorine

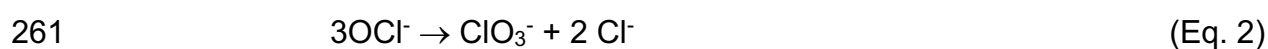
222 Within chlorine disinfection, TCAA and DCAA are the major DBPs formed, followed by THMs  
223 (Zhai et al., 2017; Di Cristo et al., 2015). NOM presence in raw water has a high potential to  
224 generate THMs and HAAs during the chlorination process and it is correlated with seasonal  
225 variations and water quality parameters (Alver et al., 2018). AOM is a precursor as well and  
226 during chlorination leads to C-DBPs and N-DBPs (Qi et al., 2016; Goslan et al., 2017),  
227 depending both on AOM concentration and on the chlorine dose and contact time.

228 Coagulation/flocculation process is essential to achieve high NOM and AOM removal  
229 efficiencies, while a well-adjusted coagulant dose influences the performances of suspended  
230 solids removal (Gad-Allah et al., 2012; Zaleschi et al., 2012). Recent studies indicated that  
231 chlorine might attack NOM and form intermediate DBPs (including nonhalogenated and  
232 halogenated aromatic DBPs) (Jiang et al., 2017), and finally form halogenated aliphatic  
233 DBPs (including regulated THMs and HAAs) (Jiang et al., 2020, Li et al., 2020). Similarly,  
234 nitrogenous DBPs formed during chlorination depend on disinfectant dose and pH (5 to 6),  
235 while the high temperature increases the formation potential of TCNM (Chen et al., 2017).  
236 A study involving water disinfection with chlorine, UV, UV/chlorine and UV/H<sub>2</sub>O<sub>2</sub> (Wang et al.,  
237 2015), showed that DBPs formation during UV/chlorine AOPs depended on chlorine dose,  
238 exposure time and quality of water sources. The formation of THM and HAA during  
239 disinfection was low and constant for the three types of disinfection processes and depended  
240 on pH, temperature and season.

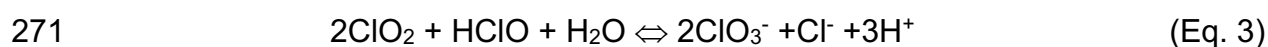
241 Chlorine used as disinfection agent in special conditions (seasonal variation, temperature,  
242 dose or drinking water quality) can form Haloacetamides (Kimura et al., 2019) or  
243 Dimethylamine (de Vera et al., 2017). The formation of CHCl<sub>3</sub> or N-DBPs during UV/chlorine  
244 disinfection occurred, as demonstrated by a study of Kimura et al. (2019). HAAs as TCAA,  
245 hydroquinone, DCAA presented a high formation potential during chlorination under short-  
246 term and long-term contact time conditions (Cordero et al.,2020). N-nitrosodimethylamine  
247 formation occurred when chlorination was applied to a surface DWT plant (Orak et al., 2019).  
248 Also, dichloroacetonitrile (DCAN) and dichloroacetamide (DCAcAm) occurred after the  
249 chlorination process (Chen et al., 2019). AOM is a major precursor of halogenated  
250 carbonaceous and nitrogenous byproducts (C-DBPs and N-DBPs) in the chlorination  
251 process (Wu et al., 2020; Maeng et al., 2019).

### 252 3.2.2. Chlorine dioxide

253 The increased application of chlorine dioxide in DWT, particularly for surface water, avoids  
254 the formation of THMs but can generate other DBPs: chlorite and chlorate, both supposed to  
255 act as endocrine disruptors affecting human thyroid (Snyder et al., 2009). Chlorite ( $\text{ClO}_2^-$ )  
256 and chlorate ( $\text{ClO}_3^-$ ) are well-known degradation products of hypochlorite solutions.  
257 Hypochlorite ion ( $\text{OCl}^-$ ) is unstable and naturally undergoes two independent paths of self-  
258 decomposition: one generating oxygen and chloride (Eq. 1), and the other chlorate and  
259 chloride ions (Eq. 2):



262 Chlorite is rapidly formed as an intermediate between hypochlorite and chlorate in (Eq. 2)  
263 (Snyder et al., 2009). Chlorine dioxide ( $\text{ClO}_2$ ), in contact with organic and inorganic  
264 oxidizable substances (DBPs precursors), degrades to chlorite, chlorate and chloride ions,  
265 thus reducing the subsequent possibility of THMs and HAAs developing in the distribution  
266 networks (Al-Otoum et al., 2016; Ye et al., 2019). Chlorite levels may vary between 10-20 %  
267 of  $\text{ClO}_2$  dose in winter and 40-70 % in summer, while chlorate levels usually are between 10  
268 and 30 % of the  $\text{ClO}_2$  dose (Gates et al., 2009). Chlorine dioxide, in the presence of  
269 hypochlorite, may undergo a disproportionation reaction to form chlorate (Eq. 3) (Gates et  
270 al., 2009):



272 The factors commonly assumed to be of main influence in chlorite and chlorate generation  
273 are: temperature and sunlight, which may catalyze the reactions presented in Eqs. 1- 3;  
274 hypochlorite and chlorine dioxide doses; characteristics of raw water, particularly the  
275 presence of transition metals and other oxidizable substances (Gates et al., 2009; Snyder et  
276 al., 2009; Garcia-Villanova et al., 2010). It was found that DBPs occurrence during sodium  
277 hypochlorite disinfection was affected by bromide ions (Rong et al., 2018).

278 DBPs formation (anhydroerythromycin, N-desmethyl clarithromycin and N-desmethyl  
279 clarithromycin), occurred after chlorine dioxide treatment in the raw water of a DWT plant

280 located in Barcelona. Anhydroerythromycin was removed by both conventional (ozonisation)  
281 and advanced (reverse osmosis) processes, and N-desmethyl clarithromycin was removed  
282 only after ADWT (Rubirola et al., 2019).

### 283 3.2.3. Chloramination

284 NOM is considered the main precursor of DBPs, it is usually associated with surface waters  
285 and interacts with chloramine forming THMs, HAAs and HANs. Dissolved fractions of organic  
286 nitrogen and carbon (DON and DOC) play an essential role in the formation of C-DBPs and  
287 N-DBPs in the pre-chloramination stage during summer (Chu et al., 2011). NOM with higher  
288 aromaticity in the reaction with chloramine increased phenolic I-DBPs and decreased polar  
289 aliphatic I-DBPs (Pan et al., 2016).

290 I-DBPs formation in the presence of zero-valent iron (ZVI) during chloramination is strongly  
291 influenced by the same operational parameters (initial pH,  $\text{IO}_3^-$  concentrations and  
292 disinfectant dose). However, ZVI presence, iron corrosion scale, phosphate concentration  
293 and  $\text{Br}^-/\text{IO}_3^-$  molar ratio accelerated the formation potential of I-THMs (Xia et al., 2017).  
294 THMs and HAAs are the most abundant DBPs generated by hypochlorite, while HANs may  
295 derive from chloramines.

296 ADWT systems that use UV or UV/ $\text{H}_2\text{O}_2$  disinfection processes can interact with  
297 pharmaceutical substances and form NDMA or increased concentrations of DBPs after the  
298 addition of chloramine (Radjenovic et al., 2012; Postigo and Richardson, 2014). During the  
299 chloramination process used for DWT some DBPs are formed, especially Nitrosamines  
300 (Selles et al., 2018), N-DBPs and Chloroform (Kimura et al., 2019) or NDMA (Ding et al.,  
301 2019). Chloramine disinfection reagent contribute to the formation of NDMA and THMs,  
302 respectively, both of which are defined as disinfection byproducts (Orak et al., 2019). Iodo-  
303 THMs (DCIM, DBIM, BCIM, CDIM and TIM) occurred in 65 water treatment systems in  
304 Canada during drinking water disinfection process with chloramine, and their concentration  
305 ranged from 0.02  $\mu\text{g/L}$  to 21.66  $\mu\text{g/L}$ , due to various water quality parameters, seasonal

306 variations and relevant precursors content (DOC, bromide, iodide and iodine) (Tugulea et al.,  
307 2018).

#### 308 3.2.4. Ozone

309 When ozonation is followed by chlorination, the concentration of brominated and iodinated  
310 THMs and HAAs increases, but nitrosamines formation is inhibited at high concentrations of  
311 bromide (Zha et al., 2014). It was reported that in surface waters, bromides converted to  
312 brominated DBPs has a potential hazardous impact on living organisms, especially on the  
313 endocrine system, and on the aquatic environment (Watson et al., 2015; Winid, 2015).

314 In the presence of chlorine, chloramine or ozone, bromide and iodide halogenated ions  
315 influence the amount of Cl-DBPs, Br-DBPs or I-DBPs and not only THMs, HAAs and N-  
316 nitrosodimethylamine (Pan et al., 2015). Iodide compounds form hypoiodous acid/iodine by  
317 disinfection processes with chlorine, monochloramine and ozone, or iodine-radicals by  
318 chlorine dioxide. Bromate formation can be inhibited during water treatment process by  
319 adapting ozonation to electro-peroxone process by a carbon-based cathode and production  
320 of H<sub>2</sub>O<sub>2</sub> from O<sub>2</sub>. In this case, Br-DBP is reduced, while NOM removal in the drinking water  
321 treatment process increases (Li et al., 2015).

322 Carboxylic acids may occur after disinfection, especially when chlorine and ozone are used  
323 (Richardson et al., 2000), their amount varying due to raw water quality and DWT processes.  
324 Jurado-Sánchez et al. (2014) detected 35 aliphatic and aromatic acids in two DWT plants  
325 characterized by different process outlines. The first plant used chlorination/ chloramination  
326 while the second one used ozonation and chlorination; the presence of the carboxylic acids  
327 being 6 times higher in the second plant, the number of generated carboxylic acids  
328 increasing especially in autumn and winter seasons. NDMA (Kimura et al., 2019) and  
329 Hydroxylamine (Heeb et al., 2017) are some DBPs formed during drinking water disinfection  
330 with ozone especially under various conditions (ozone dosages, pH) and different

331 components in water (bromide ion ( $\text{Br}^-$ ), bicarbonate ion ( $\text{HCO}_3^-$ ), sulfate ion ( $\text{SO}_4^{2-}$ ), and  
332 humic acid (HA), as well as NOM from a lake (Shen et al., 2019).

### 333 3.2.5. Ferrate

334 A new disinfection agent is ferrate (Fe IV), which can successfully be applied on DWT  
335 processes given its oxidation, coagulation (Prucek et al., 2013) and disinfection (Alsheyab et  
336 al., 2009) abilities. It also limits DBPs generation and contributes to NOM and EPs  
337 elimination (Jiang, 2013). DBPs formation through chlorine dioxide or ferrate used for  
338 surface water treatment show that the production of THM, haloketones, haloacetonitriles and  
339 trichloronitromethane is higher for chlorine dioxide (Yang et al., 2013, Liu et al., 2020). Few  
340 studies regarding DBPs formation reported that high doses of ferrate (21 mg/L) can reduce  
341 nitrosamines up to 84 % (Lee et al., 2008) or react with amino acids, resulting in aldehydes  
342 (Sharma, 2010). Halogenated DBPs (including THMs, HAAs, haloketones, chloral hydrate,  
343 haloacetonitriles and trichloronitromethane) are the byproducts resulted from the disinfection  
344 of water with ferrate in presence of an important precursor such as AOM (Dong et al., 2019).

345

### 346 3.2.6. Multi stage disinfection processes and the influence of the distribution system

347 Other crucial issues about DBPs generation refer to the type and dose of disinfectants,  
348 related to raw water quality and size of DWT plant. While in small DWT plants, particularly  
349 those dedicated to groundwater treatment, a single-phase disinfection is usually adopted,  
350 large DWT plants generally perform multi-stage disinfection processes. In such systems,  
351 primary disinfection targets the removal of pollutants and pathogens (usually by means of  
352 ozone or chlorine dioxide). While secondary disinfection has the main goal of assuring  
353 drinking water quality along the water distribution network up to the final users. As a result,  
354 primary and secondary disinfectants are both considered DBPs' potential sources and their  
355 interaction should be taken into account. Hypochlorite is usually employed as secondary  
356 disinfectant because of its persistence in the distribution network, therefore most

357 international regulations call for a residual chlorine concentration in drinking water (e.g. EU  
358 requires 0.2 mg/L through Directive 98/83/CE) (EC, 1998). Another relevant factor that can  
359 influence DBPs formation is the seasonal variation of the surface water characteristics.  
360 Higher temperature determines the use of an increased dose of disinfectant (aimed to  
361 control the increase in pathogens growth) and supports its reaction with NOM, leading to  
362 high DBPs formation in summer and autumn (Ibrahim and Abu-Shanab, 2013; Guilherme  
363 and Rodriguez, 2014), while HAAs concentration is higher in the cold season (Wei et al.,  
364 2010). The occurrence of DBPs in the drinking water network is variable, and their  
365 concentration range does not depend on the size of the network itself (Guilherme and  
366 Rodriguez, 2015). At the same time, DBPs concentration can fluctuate within the DWT, so  
367 the difficulty of their removal consists in choosing an appropriate process outline, taking into  
368 account: spatial location of the treatment system, raw water extraction quality, seasonal and  
369 short-term variations (Mercier-Shanks, et al., 2013). The fluctuating levels of THMs and  
370 HAAs across the treatment system show a temporal and spatial variability of their  
371 concentrations, which hinders the monitoring process and makes it difficult to choose a  
372 period or point of sampling (Guilherme and Rodriguez, 2015). Recent studies emphasized  
373 the role of different chemical agents and treatment/piping materials of the DWT or  
374 distribution system on DBPs formation. Besides additional DBPs precursors (phenolic  
375 structures, amino acids, oligopeptides and nitrogen-containing contaminants), another  
376 concern is that certain treatment stages have the potential to transform DBPs into more toxic  
377 compounds. For example, activated carbon, ion-exchange resins or membrane treatments  
378 can increase the ratios of Br-DBPs to Cl-DBPs, resulting in higher mammalian cell  
379 cytotoxicity and genotoxicity, although the overall DBPs concentrations decrease (Ding et al.,  
380 2019). Also, in the distribution systems the main challenges include the maintenance of  
381 stable concentrations of residual disinfectant and the control of microbial content that may  
382 form DBPs as consequence of residual decay processes. Microbial activity is a complex  
383 process and many factors (temperature, water age, piping material, corrosion products,

384 nutrients, natural organic matter, hydraulic condition and disinfectant residual type and  
385 dosage) could influence it. Disinfectant types and dose were found to be among the most  
386 important factors driving the occurrence of DBPs in distribution networks (Li et al., 2019).

387

### 388 3.3. *Methods of analysis*

389 Most common methods for DBPs analysis include (Table 3): LC/ESI-MS/MS (liquid  
390 chromatography-electrospray ionization tandem mass spectrometry) for determination of  
391 chlorinated and brominated DBPs (Nollet and De Gelder, 2014) and also used to detect  
392 polar halogenated DBPs, especially aromatic DBPs (Yang et al., 2019); capillary  
393 electrophoresis (CE) (Kubáň et al., 2012) and high-field asymmetric waveform ion mobility  
394 spectrometry (FAIMS-MS) for HAA analysis (Barceló et al., 2012); membrane-introduction  
395 mass spectrometry (MIMS) for CNX (cyanogen halide) (Barceló et al., 2012); FT-ICR MS  
396 (Fourier transform ion cyclotron resonance mass spectrometry) for bromate and brominated  
397 DBPs (Yang and Zhang, 2016); SPE-LC-MS/MS (solid phase extraction-liquid  
398 chromatography-tandem mass spectrometry) for haloquinones investigation from WTPs (Li  
399 et al., 2011). Gas chromatography coupled with Orbitrap-based on mass spectrometry was  
400 used for I-DBPs analysis (Cojocariu et al., 2016), GC-ECD was reported to identify the DBPs  
401 formed by UV/chlorine disinfection (Huang et al., 2017) and GC-MS was able to detect  
402 specific DBPs with low molecular weight (Yang and Zhang, 2016). Other specific analytical  
403 methods for emerging DBPs detection include: High-Resolution Mass Spectrometry (HR-  
404 MS) (Richardson and Postigo, 2016); Ion Chromatography-Mass Spectrometry (Bruzzoniti et  
405 al., 2019), combined SPE, DCLCHR-LCMS and PIE (Zulkifli et al., 2018). HNMs HANs,  
406 HAcAms, NDMAAs and other emerging N-DBPs (Ding and Chu, 2017) are commonly  
407 analyzed using GC (Pozzi et al., 2011; Chen et al., 2015; Ma et al., 2014; Montesinos and  
408 Gallego, 2012), HPLC (Chu et al., 2012; Kodamatani et al., 2016) and UPLC (Müller et al.,  
409 2012; Ripollés et al., 2011).

410

411 **Table 3.** Analytical techniques used for DBPs measurement

Analysis method	DBPs analyzed	Reference
SPE-HPLC-MS/MS	I-HAAs and aromatic I-DBPs	Hu et al., 2018
FT-ICR MS	Br-DBP	Yang and Zhang, 2016
GC-O/MS	I-DBP	Cojocariu et al., 2016
GC MS	HNMs HANs, HAcAms, NDMA and emerging N-DBPs	Ding and Chu, 2017; Pozzi et al., 2011; Chen et al., 2015; Ma et al., 2014; Montesinos and Gallego, 2012
HPLC MS		Chu et al., 2012; Kodamatani et al., 2016
UPLC		Müller et al., 2012; Ripollés et al., 2011
LC/ESI-MS/MS	Chlorinated and Brominated DBPs	Nollet and De Gelder, 2014
	halogenated DBPs	Yang et al., 2019
MIMS	CNX HAA	Barceló et al., 2012
CE		
FAIMS-MS		
HPLC-MS	Hydrazine	Susinskis et al., 2018
IC-MS/MS	Nine HAAs, bromate, dalapon and I-HAAs	Wu et al., 2017
UHPLC/QTOF-MS	NDMA	Hanigan et al., 2017
GS-ECD	THMs, THAs, HANs and HKs	Huang et al., 2017
LLE-ETAC	DCAcAm	Lin et al., 2016
HS-SPME/GC-MS	Haloketones	Serrano, M., et al., 2015
LC/HR-MS	N-DBPs	Kolkman et al., 2015
MLLE	Haloketones	Serrano, M., et al., 2014
UPLC/ESI-MS-MS	Halobenzoquinones (HBQs)	Huang et al., 2013
SPE-LC-MS/MS	Haloquinones	Li et al., 2011

412

413 **3.4. Environmental and health impacts**

414 Human organisms can be exposed to DBPs through three conventional pathways: dermal  
415 contact, ingestion or inhalation (Nieuwenhuijsen et al., 2009; Chowdhury et al., 2011;  
416 Chaves et al. 2019; Chowdhury et al., 2020; Gonsioroski et al. 2020). The presence of DBPs  
417 in drinking water became a human health concern because some epidemiological studies  
418 demonstrated associations between DBPs exposure and increased risk of cancer  
419 development, liver, kidney defects and central nervous system problems, adverse  
420 reproductive outcomes (Legay et al., 2010; Chowdhury et al., 2017; Chen et al., 2019; Wang  
421 et al., 2019; Gonsioroski et al., 2020) and endocrine disruption (Chaves et al., 2020). Urinary  
422 bladder cancer has been the health risk most consistently associated with chlorination DBPs

423 (Hrudey et al., 2015; Regli et al.,2015; Chaves et al. 2019; Diana et al., 2019). Another study  
424 (Jones et al., 2019) evaluated the association between high DBPs levels with increased risk  
425 of colon and rectal cancer, however positive associations with individual THMs and HAAs,  
426 most consistent for rectal cancer, require further investigations.

427 Emerging DBPs formed in distribution system during chlorination and chloramination (THMs,  
428 HAA and HANs) have been reported to cause symptoms such as liver, kidneys and nervous  
429 system diseases and health risks associated with these regulated DBPs can be spontaneous  
430 abortions, births defects, stillbirths and negative reproductive effects (Quintiliani et al., 2018).  
431 Recent research focused on maternal exposure to emerging DBPs (THMs and TCAA) during  
432 pregnancy, and some DNA anomalies were observed in cord blood (Yang et al., 2017; Salas  
433 et al., 2015) causing foetal growth restriction (Cao et al., 2016) or other adverse reproductive  
434 outcomes (Wang et al., 2016; Holmes et al., 2017). Rivera-Nunez et al. evaluated the  
435 association between maternal exposure to THMs and DCAA. In a study of 2460 cases in  
436 Massachusetts from 1997 to 2004, chloroform, BDCM (Lodhi et al., 2017) and DCAA  
437 exposures (Dad et al., 2018) were associated with stillbirths (Rivera-Nunez et al., 2018). In a  
438 study of 7438 singleton term babies in United Kingdom, THMs and HAA% exposure during  
439 pregnancy was associated with reduced birth weight, but suggested differences by ethnicity  
440 (Smith et al., 2016). Halomethanes, iodo-trihalomethanes, nitrosamines, halobenzoquinones,  
441 NDMA, DCAN, DBAN exposure has been shown to be more genotoxic and cytotoxic than  
442 some regulated compounds, reinforcing their potential toxicological effect to humans  
443 (Chaves et al., 2020), and posing higher cancer risks for children than for adults (Luo et al.,  
444 2020).

445 Emerging DBPs may be classified as carcinogenic substances (Di Cristo et al., 2015; Ng et  
446 al., 2016) or may affect human health due to effects like: alteration of pregnancy duration,  
447 menstrual cycle or pregnancy loss, foetal development and congenital malformations or  
448 cancer (Villanueva et al., 2015). Long-term human exposure to various types of DBPs  
449 triggers many forms of cancer or other diseases (as presented in Table 1) (Hebert et al.,

2010; Yang and Zhang, 2014; Xiao et al., 2017; Chhetri et al., 2017). The risk to develop cancer (López-Roldán et al., 2016) or diseases caused by THMs ingestion is higher than the risk caused by inhalation during shower or dermal exposure (Dyck et al., 2015). Therefore, water contaminated with DBPs (mostly THMs and HAAs) is highly dangerous for human health and can present high risks if the raw water originates from surface water, and less risks if the source is mixed (e.g., surface water and groundwater or only groundwater). The variety of DBPs known today, caused by the multitude of sources that influence their formation and occurrence, is reflected in the variety and complexity of the negative effects they can have on the environment and humans (Table 1). Some DBPs are considered EPs due to their persistence, low biodegradability and although they can be found in low concentrations, they may have the most devastating effects as compared to other contaminants (Yang and Zhang, 2016).

Because emerging N-DBPs resulted after chlorination or chloramination present cytotoxicity and genotoxicity risks, treatment processes should be applied as described in section 4 (Hu et al., 2018; Chen et al., 2017). With the same toxic characteristics, Br-DBPs resulting during water chlorination and chloramination in the presence of bromide, have a high potential to generate illnesses as compared to Cl-DBPs (Zhang and Yang, 2018). Some classes of DBPs (N-nitrosamines) are classified at international level as substances with a potential risk to develop cancer if the daily ingestion rate is exceeded (Fan and Lin, 2018).

Recent studies focused on formation mechanisms, concentrations and adverse health effects have been conducted those I-aldehydes, cyanides, halonitromethanes, haloketones, haloacetamides, iodinated-DBPs and N-nitrosamines presented in drinking waters are harmful for human health being more cytotoxic, genotoxic and mutagenic than their brominated and chlorinated equivalents (Chen et al., 2018; Andersson et al., 2019, Chaukura et al., 2020). Among N-DBPs, NDMA have received significant attention because low ng/L levels in drinking water are associated with  $10^{-6}$  lifetime excess cancer risks. Epidemiological studies highlight that exposure via inhalation and dermal contact may be

477 more dangerous than via ingestion of drinking water, because many emerging DBPs are  
478 sufficiently volatile such that skin absorption or inhalation during showering can be harmful  
479 (Li et al., 2018, Li et al., 2020).

480

#### 481 **4. Advanced DWT technologies for DBPs prevention and control**

482 Given the number of known DBPs and its continuous growth, to prevent their occurrence and  
483 development, operational parameters and drinking water characteristics should be carefully  
484 monitored. Implementing technologies that have the ability to remove DBPs and to prevent  
485 their re-emergence in the distribution network is highly necessary (López-Roldán et al.,  
486 2016). Conventional DWT processes needs to be completed with ADWT processes (Du et  
487 al., 2017; Ohar et al., 2014; Chaukura et al., 2020) and/or the use of a strong oxidant as final  
488 disinfectant (Zainudin et al., 2018). In order to mitigate DBPs formation, dissemination  
489 activities oriented to a reduction of precursors or unnecessary pharmaceuticals use among  
490 the population could also be effective (Plewa et al., 2010).

491 Recent studies at pilot and full-scale showed the high performances of ADWT technologies  
492 (Hu et al., 2018; Wang et al., 2015; Jurado-Sanchez et al., 2014) (see Table 4), such as:  
493 membrane processes, advanced oxidation processes or adsorption on activated carbon and  
494 other materials, which were studied from the points of view of: technological performances  
495 (mechanism, pollutants removal efficiency), economic, social and environmental  
496 performances (Zainudin et al., 2018; Bui et al., 2016). From the chemical, biological and  
497 technological point of views, the prevention of DBPs formation may be possible through  
498 preventive actions (disinfectant agents used/ removing DBPs precursors through adequate  
499 DWT processes) or treatment actions (removing DBPs after their formation). Each treatment  
500 stage, like adsorption (GAC, PAC, CNTs, IER), coagulation/flocculation- C/F, AOPs,  
501 membrane filtration or integrated technologies), has its own contribution to the ADWT  
502 process efficiency. However, combined technologies ensure better drinking water quality,  
503 while preventing the development of certain risks from the recurrence of DBPs.

504 **Table 4.** Overview of DBPs removal by ADWT at pilot and full-scale

Treatment process	DBPs monitored	Operational parameters (PS/FS)	Removal efficiency (%)	References
C/F; S; SF PO (KMnO <sub>4</sub> /O <sub>3</sub> /K <sub>2</sub> FeO <sub>4</sub> / ClO <sub>2</sub> ); C/F; S; SF PO (KMnO <sub>4</sub> /O <sub>3</sub> /K <sub>2</sub> FeO <sub>4</sub> / ClO <sub>2</sub> ); C/F; S; SF; GAC filtration/O <sub>3</sub> - GAC advanced treatment	THMs, CH; DCAcAm; TCAcAm; TCNM and DCAN	(FS) pH: 6.5 ± 0.2 Contact time: 24 h	17.6%, 23.6%, 19.6%, and 14.5%; 45.3%, 51.1%, 49.3%, and 46.1%;	Hu J. et al., 2018
UV/TiO <sub>2</sub> UV/Pt/TiO <sub>2</sub> Fe(0)/Cu(II) UV-L UV-M	Bromate	(PS) Reaction time/pH: 90-150 min/1.5-13.5 90 min/ 8.1; 30 min/6-6.5; 60 min/5.1-9.2; 50 min/ 6.8	50-60%; 95-99%; 100%; 35%–45%; 100%	Xiao et al., 2017
1. PO (Cl <sub>2</sub> ); C/F; SF (sand/anthracite) 2. PO (Cl <sub>2</sub> ); C/F; GAC filtration	36 DBPs	(PS) Contact time: 7.5/15 min; Cl <sub>2</sub> dose:3-6 mg/L	THMs 20%; Halonitroalkanes- 58.50%; Haloaldehydes 33.62%; HAAs 28.13%; Haloalkanes 20.46%; Haloketones 13.46%; Nitrosamines 10.23%; Halonitriles -8.82%; Haloalkenes -9.84%; N-DBPs 4.82%; Chlorite 84.95% (GAC), Chlorite 19.55 % (SF)	Fu et al., 2017
Membrane nanofiltration: ESNA 1-LF2; TS80; NF270	NDMA, HNM, and THM	(PS) pH: 6-9; Ionic strength: 0.005- 0.05 M; Ca <sup>2+</sup> : 6-60 mg/L	57-83%; 48-87% and 72-97%	Ersan et al., 2016
RWE, PO, pH adjustment (H <sub>2</sub> SO <sub>4</sub> ); C/F (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ); SF; O <sub>3</sub> , GAC filtration; Cl <sub>2</sub> ; pH adjustment (H <sub>2</sub> SO <sub>4</sub> )	THMs (CHCl <sub>3</sub> and BDCM); HAAs (MCAA, MBAA, DCAA, DBAA, BCAA, BDCAA, TCAA, TBAA, H)	(FS) O <sub>3</sub> dose: 5 - 0.8 mg/L; T: 7.4°C to 21°C; pH 8.02 - 8.8;	79% for CHCl <sub>3</sub> ; 21%for BDCM; 71% for DCAA and TCAA	Papageorgiou et al., 2016
1) C; S; SF; Cl <sub>2</sub> 2) C; S; SF; O <sub>3</sub> , BAC, Cl <sub>2</sub>	DCAcAm	(FS) pH: 8,5; Cl <sub>2</sub> dose: 25.5 mg/L; Disinfection time: 24 h	9% in CDWT plant and 76% in ADWT plant	Lin et al., 2016
1) RWE, PO (ClO <sub>2</sub> ), C,F,S,SF; UF, RO,REM, Cl <sub>2</sub> , Tanks, Cl <sub>2</sub> 2) RWE, PO (ClO <sub>2</sub> ), C,F,S,SF; O <sub>3</sub> , GAC filtration, Cl <sub>2</sub> , Tanks, Cl <sub>2</sub>	THMs	(FS)	-	López-Roldán et al., 2016
RWE; F (Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub> ); pH adjustment and activated silica; S; SF (anthracite/sand); NH <sub>2</sub> Cl; pH correction and FI	TTHM and HAA5	(FS) Contact time 20 min; Bed layer: anthracite 0.56m and sand 0.3m	75%overall reduction for TTHM and HAA5	Delatolla et al., 2015
RWE; PO (Cl <sub>2</sub> ); C/F (Al <sub>2</sub> Cl <sub>3</sub> ); S; SF; NH <sub>2</sub> Cl	10 THMs, 13 HAAs, 6 HNMs, 6 HANs and 11 aldehydes	(FS) Cl <sub>2</sub> dose: 0.6–1.0 mg/L; NH <sub>2</sub> Cl dose: 2.1–2.6 mg/L; Contact time 24/48 h; pH 3.0–3.5 (H <sub>2</sub> SO <sub>4</sub> )	Increased concentration in summer seasons along the distribution network: 50% for HAAs and 350% for THMs. Cl <sub>2</sub> , NH <sub>2</sub> Cl dose, C/F, and S steps increase DBPs level in warmer seasons	Serrano et al., 2015
RWE; PO (Cl <sub>2</sub> ); C/F (Al <sub>2</sub> Cl <sub>3</sub> ); S; SF (sand/anthracite); UV (Cl <sub>2</sub> ; UV; UV/Cl <sub>2</sub> ;UV/H <sub>2</sub> O <sub>2</sub> )	THM, HAA, DCAN, BCAN; AOX	(FS) Cl <sub>2</sub> dose: 5–10 mg L <sup>-1</sup> as free chlorine; UV dose: 1800Mj/m <sup>2</sup> ; UV dose: 1800Mj/m <sup>2</sup> ; Contact time: 30/40	UV/Cl <sub>2</sub> : 27 µg L <sup>-1</sup> for THM and HAA; < 6 µg L <sup>-1</sup> for DCAN and BACN; 70 µg Cl L <sup>-1</sup> for AOX. UV/Cl <sub>2</sub> : 50 µg L <sup>-1</sup> for	Wang et al., 2015
RWE; C/F (Al <sub>2</sub> Cl <sub>3</sub> ); S; SF				

Treatment process	DBPs monitored	Operational parameters (PS/FS)	Removal efficiency (%)	References
(sand/anthracite); UV (Cl <sub>2</sub> ; UV; UV/Cl <sub>2</sub> ;UV/H <sub>2</sub> O <sub>2</sub> )		seconds; pH: 6.5-8.5	THM and 19-24 for HAA; 20-70 µg Cl L <sup>-1</sup> for AOX	
RWE, PO (O <sub>3</sub> ), pH adjustment, C/F, SF, O <sub>3</sub> , GAC filtration, Cl, pH adjustment	Carbonyl compounds	(FS) T: 7.4 - 17.7°C; pH: 8.02 – 8.80 O <sub>3</sub> dose: 0.29-0.52 mg/L;	~80% (15 - 62% on GAC filtration)	Papageorgiou et al., 2014
RWE, PO (KMnO <sub>4</sub> ), C (Al <sub>x</sub> Cl <sub>3</sub> ), S, SF (sand + anthracite), NH <sub>2</sub> Cl, Tanks, Cl+FI	35 Carboxylic acids	(FS) T:9-28°C; NH <sub>2</sub> Cl dose 0.7 mg/L; Contact time 20 min; pH 6-8	6.3 µg/L	Jurado-Sanchez et al., 2014
RWE, C, O <sub>3</sub> , UF (ceramic membrane), GAC filtration, Cl	4 THMs, 6 HAAs	(FS) Cl <sub>2</sub> dose 1.5 mg/L; O <sub>3</sub> dose 2–5 mg/L; Contact time 120 min;	73% for THMs and 75% for HAAs	Fan et al., 2014
4 DWTPs: RWE; PO (Cl <sub>2</sub> ), C/F, S, SF, Cl <sub>2</sub>	THM4, HAA5	(FS) pH: 7.4-8.1;	64.38, 43.94, 52.41 and 51.72 µg/L for THM4; HAA5;	Gad-Allah et al., 2012

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**Notes and abbreviations:** RWE – raw water extraction; PO – pre-oxidation; pH adjustment; C/F – coagulation/flocculation; SF – sand filtration; O<sub>3</sub> – ozonisation; GAC filtration; Cl – chlorination; S – sedimentation; FI – fluorination; NH<sub>2</sub>Cl – chloramination; UF – ultrafiltration; Cl<sub>2</sub> – Chlorination; BAC – Biological Activated Carbon; REM – Remineralisation; RO - Reverse Osmosis; UV - Ultraviolet Disinfection; PS – Pilot scale; FS – Full scale.

512 Adsorption is often applied due to its low cost and operational simplicity, providing a high  
513 quality of treated water. The majority of DWT processes (Table 4), applied at pilot or full  
514 scale, include adsorption on GAC with high removal efficiencies for DBPs. For nitrosamines  
515 removal from DW sources, GAC filtration shows an increased efficiency of DBPs precursor  
516 removal or DBPs formed after the disinfection process (Leavey-Roback et al., 2016; Zhang  
517 and Yang, 2018). A new approach has been studied to control the formation of THMs, HAAs,  
518 and total organic halogen (TOX) in chlorinated drinking water by targeting intermediate  
519 aromatic halogenated DBPs instead of NOM by using GAC adsorption, the applicability of  
520 this method being influenced by the source water matrices and disinfection conditions. With  
521 different bromide levels in source waters and different chlorination contact times, the TOX  
522 removals with this new approach (chlorine disinfection prior to GAC adsorption) were always  
523 doubled or more than doubled as compared to those of the traditional approach (GAC  
524 adsorption prior to chlorine disinfection) (Jiang et al., 2018). However, the potential to  
525 remove some DBPs through GAC obviously decreases with the saturation of the adsorbent

526 (Zainudin et al., 2018). Another disadvantage consists in the need to manage the  
527 wastewater resulting from back-washing the GAC filters (Bhatnagar and Sillanpaa, 2017).

528 In a recent study, Lin et al. (2016) showed that biological activated carbon (BAC) filtration is  
529 the most popular technology in DWT process, followed by ozonation. They proved that  
530 combined advanced processes ( $O_3$ +BAC) significantly improve DCACAm removal. Few full-  
531 scale studies on control and removal of DBPs through BAC processes show that HAAs and  
532 THMs formation potential was minimized at 57 % and 45 %, respectively, since bromide  
533 concentration was higher after biofiltration. This required a higher capacity to inactivate the  
534 microorganisms responsible for the DBPs occurrence (Liu et al., 2017). Different  
535 coagulation/flocculation processes ( $KMnO_4$ ,  $O_3$ ,  $K_2FeO_4$ , and  $ClO_2$ ) applied after pre-  
536 treatment resulted in DCAN removal efficiencies higher with 12.9 %, 18.0 %, 16.3 % and  
537 10.1 %, respectively, compared to conventional DWT. When  $O_3$ -GAC was added to the DWT  
538 process, the removal efficiencies increased to 48.6 %, 53.0 %, 51.0 %, and 48.5 %,   
539 respectively (Hu et al, 2018).

540 Photocatalytic technologies applied for bromate degradation from post-ozonation showed  
541 good reduction efficiency, stable performance and easy combination with UV disinfection  
542 techniques ( $UV/TiO_2$ ;  $UV/SO_3^{2-}$ ;  $UV/Pt/TiO_2$ ) (Xiao et al., 2017). AOPs with ozone ( $O_3$ ),  
543 hydrogen peroxide ( $H_2O_2$ ), UV radiation or combinations of these are often used for DBPs  
544 removal and recording positive results in DWT processes. Advanced disinfection techniques  
545 such as UV or UV/hydrogen peroxide can interact with pharmaceuticals and form  
546 hydroxylated compounds and NDMA or can increase some levels of DBPs after chloramine  
547 addition (Postigo and Richardson, 2014; Radjenovic et al., 2012).

548 Membrane filtration is often applied for DBPs removal due to the high removal efficiency  
549 especially towards THMs precursors (Sutherland et al., 2015). Ultrafiltration (UF), reverse  
550 osmosis (RO), advanced oxidation water treatment by post-ozonation and biological  
551 activated carbon (BAC) were demonstrated to have a significant contribution in THMs and  
552 HAAs removal from drinking water (Lou et al., 2010; Fan et al., 2014). The removal of DBP

553 precursors is influenced by the presence of bacteria, micropollutants and membrane  
554 characteristics. For example, chloroform is easily removed using a UF membrane rather than  
555 NF or RO membranes (Bodzek et al., 2002). Different membrane technologies (MF, NF, UF,  
556 RO) (Fan et al., 2014; Ersan et al., 2016; López-Roldán et al., 2016) proved their efficiency  
557 in DBPs removal (see Table 4), however there is a lack of data regarding the application of  
558 membrane processes at pilot or full scale and the operational costs (imposed by the rapid  
559 fouling of the membranes), high electricity consumption etc. are not available (Zainudin et  
560 al., 2018).

561 Considering a critical assessment of the ADWT processes analysed (see Table 4),  
562 integrated technologies involving membrane processes present higher removal rates for  
563 DBPs or precursors compared to single DWT processes. In the case of THM removal, the  
564 efficiency sequence follows the order:  $O_3/BGAC > GAC$ ;  $O_3/UF > UF$  and  $GAC/SF > SF$   
565 (Zainudin et al., 2018). However, as a general principle, the benefits of a particular  
566 technology applied in a DWT process outline are related to the operational parameters  
567 adopted. Once applied a certain technology, a number of challenges arise, especially  
568 regarding the quality of water supplied to consumers, which must comply with legislation. At  
569 the same time, the performance of a treatment technology may be different from plant to  
570 plant. Depending on where any specific technology is implemented within the treatment  
571 process, it may influence positively or negatively the quality of the water required for human  
572 consumption.

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## 574 **5. Conclusions**

575 DWT plants monitoring and management may not fully cover the regulated DBPs occurrence  
576 and the further exposure of humans through drinking water consumption. DBPs and their  
577 precursors removal from treated water is the key to supply safe drinking water. Unlike the  
578 removal of commonly known precursors, which has been greatly developed, up-to-date  
579 treatment processes may be not efficient in removing emerging DBPs because of the lack of

580 data on their trace concentrations and the risks associated with these compounds. DBPs  
581 removal using a single treatment technology is likely not the best approach for their removal  
582 from drinking water. It is necessary to investigate the use of coupled/integrated systems,  
583 which can improve the effectiveness of single technologies for the removal of these complex  
584 contaminants. The key findings of this study are summarised below.

585 Firstly, operational parameters such as pH, temperature, disinfectant dose, reagents used,  
586 and raw water quality should be carefully analysed and optimized to reduce or remove  
587 emerging DBPs in order to comply with current standards and regulations. Secondly, the  
588 occurrence of DBPs in drinking water due to their persistence, low biodegradability and  
589 concentrations, and the pathways related to population exposure may have the most  
590 devastating effects against the human body (particularly, the nervous and reproductive  
591 systems) for a long time.

592 This study represents a solid foundation for further research, and it highlights the strengths  
593 and weaknesses of DWT systems, according to their operational parameters. Due to the  
594 multitude of DBPs known and their newly discovered health effects, the analytical methods  
595 must be continuously improved. A particularity of this review paper is that only the ADWT  
596 technologies for DBPs removal from drinking water sources applied at the pilot or full-scale  
597 were considered. It was found that a limited number of studies have been performed in this  
598 direction. Various treatment technologies, including membrane processes, advanced  
599 oxidation processes and adsorption on activated carbon or other materials have found  
600 application at pilot and full-scale, and their selection among available existing mature  
601 technologies was mainly based on technical operation conditions and economic  
602 considerations. According to the studied literature, the treatment processes that are efficient  
603 for DBPs removal from drinking water sources are ADWTs combining different treatment  
604 technologies, because every raw water source has individual characteristics and from this  
605 point of view, the technological treatment scheme should be specifically adapted to improve  
606 the DBPs' removal efficiencies.

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614 editing: C. Teodosiu and S. Fiore; data curation, investigation, writing-original draft: A.F.  
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