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REVIEW

pH effect on the formation of THM and HAA disinfection byproducts and potential control strategies for food processing



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Abstract

Chlorine-based sanitizers have seen wide spread use in food sanitation. The reaction of chlorine species with organic matter is a concern for two reasons. Available chlorine can be “used up” by organic compounds resulting in a lower amount of chlorine available for disinfection. Another concern is that some forms of chlorine can react with some organic compounds to form toxic halogenated disinfection byproducts (DBPs). Many studies have been conducted to evaluate the role of hypochlorous acid (HOCl) and hypochlorite ion (OCl⁻) in the production of DBPs with a particular interest in the production of trihalomethanes (THMs) and haloacetic acids (HAAs). Since most of the chlorine reactions are pH dependent, pH is found to have a significant effect on the formation of chlorine DBPs. In many cases, the concentration of THMs decreases and HAAs increases as pH decreases. pH also plays an important role in the determination of the type and amount of DBPs formed, with lower, more acidic, pHs resulting in the formation of less chloroform. This review summarizes the information from the literature on the role of chlorine-based sanitizers as affected by pH in the formation of different types of DBPs. Alternative novel strategies to minimize the formation of DBPs are also discussed.

Keywords: sanitizer, chlorine, disinfection byproducts, pH, hypochlorite

1. Introduction

Chlorine has long been used for the treatment of drinking water in many countries around the world. Also, chlorine has been used to treat food preparation surfaces as well as

the food itself (Wei *et al.* 1985). Chlorine-based sanitizers such as chlorine gas (Cl₂), sodium hypochlorite (NaOCl), calcium hypochlorite (Ca(OCl)₂), and chlorine dioxide (ClO₂) have seen widespread use in food sanitation. For example, fruits, vegetables, and meats can be rinsed with chlorine to help control microbial load (Wei *et al.* 1985).

Chlorine, in its pure form, is a poisonous, yellow-green gas. Chlorine is an excellent sanitizer. However, its efficacy is highly dependent on the water quality, pH, presence of inorganic salts and natural organic matter, etc. Solutions of sodium hypochlorite (bleach) are commonly-used sanitizers in commercial and domestic settings. Typically, bleach solutions of 3–6% NaOCl are used in home applications, and this is often diluted by the user prior to use. As per the United States Department of Agriculture (USDA), Food Safety and Inspection Service (FSIS) guidelines, the poultry

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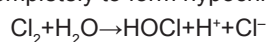
industry is allowed to use up to 50 mg L⁻¹ hypochlorite in poultry chiller water. The FSIS also requires that chlorinated water containing a minimum of 20 mg L⁻¹ available chlorine be used to sanitize surfaces that carcasses have come in contact with. While produce industry most commonly use hypochlorite solutions with a federally-mandated limit of chlorine in wash water at 200 mg L⁻¹. However, the uses of chlorine as a water disinfectant has come under scrutiny because its potential to react with natural organic matter and form chlorinated disinfectant byproducts (DBPs).

Chloramines (e.g., NH₂Cl, monochloramine) and chlorine dioxide (ClO₂) have shown promise as an alternative disinfectant to free chlorine to avoid the formation of DBPs. However, the biocidal efficacy of chloramines is significantly lower than Cl₂, and the formation of organic chloramines have raised concerns. On the other hand, ClO₂ is an effective antimicrobial agent and less sensitive to water quality and pH changes than Cl₂ (WHO 2000). Although ClO₂ is less stable than Cl₂ and must be generated on-site and used as soon as it is made, some stabilized solution forms of ClO₂ are also available.

Several of the recent reports suggest that the chlorine-based sanitizers have a tendency to form various types of DBPs such as trihalomethanes (THMs), haloacetic acids (HAAs), chlorites, and halo ketones, etc., in the presence of food organic matter (Olmez and Kretzschmar 2009; Luo *et al.* 2011; Van Haute *et al.* 2013; Shen *et al.* 2016). Several pH dependent chlorine reactions were believed to be the major factor responsible for the formation of these DBPs. This review presents an overview of various pH dependent chlorine reactions with organic matter, the formation of DBPs and their health concerns as well as novel strategies to minimize the risk.

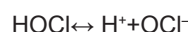
2. Chlorine chemistry and the effect of pH

Chlorine is a highly reactive gas under normal pressure and temperature. Chlorine is also available in granular or powdered form as calcium hypochlorite (Ca(OCl)₂) and in liquid form as sodium hypochlorite (NaOCl). Most elementary reactions involving chlorine compounds are second-order in nature, and the kinetics are pH dependent. Since chlorine has pH-dependent aqueous chemistry, several chlorine species including HOCl, OCl⁻, and Cl₂, will be formed in water (Doré 1989). Chlorine gas hydrolyses in water almost completely to form hypochlorous acid (HOCl):



Where, HOCl is a weak acid with a pKa of about 7.46 at 25°C. The hypochlorous acid readily decomposes upon exposure to light (Molina and Molina 1978). In the process of decomposition, the formation of Cl₂ is possible if the pH is sufficiently low (pH < 4). Above pH 7.5, HOCl dissociates

into hydrogen ions (H⁺) and hypochlorite ions (OCl⁻) in the reversible reaction:



This change of chlorine species with pH is shown in Fig. 1 (Deborde and von Gunten 2008) and HOCl is the predominant species at pH 3–7 (Gordon and Tachiyashiki 1991) and OCl⁻ dominates above pH 7.5. Hypochlorite ion is most commonly found in the form of sodium hypochlorite or calcium hypochlorite.

HOCl is a strong oxidant, and it freely oxidizes many organic compounds (Folkes *et al.* 1995; Wyman 1996; Winterbourn and Brennan 1997; Hawkins *et al.* 2003). Cell membranes are mostly impermeable to charged substances because ions have a high affinity to water molecules, which develop dipoles by nature. The hydrophobic portion of the cell membrane contains no water, so charged particles cannot pass through it. As a weak acid, HOCl can diffuse through cell membranes and acidify the interior of cells (Gutknecht and Tosteson 1973). Being both uncharged and small, HOCl is free to diffuse across the cell membrane. In addition to hydrolysis of saccharide and peptide bonds, hypochlorite can also interact with disulfide bonds. Disulfide bonds are formed by sulfur-containing amino acids such as cysteine and methionine, and they play an important role in determining a proteins folded structure. Hypochlorite readily oxidizes these bonds, resulting in their cleavage (Wyman 1996). These properties help make HOCl an effective antimicrobial agent.

3. Chlorine and its reactions with organic compounds

Chlorine reactions with organic matter are a cause of concern for two main reasons. First, available chlorine can be “used up” by organic compounds not associated with microorganisms, resulting in a lower amount of chlorine available

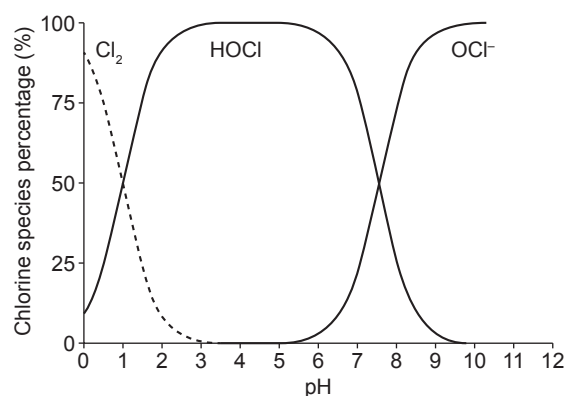


Fig. 1 The relative percentages of three different chlorine species in solution as a function of pH (Deborde and von Gunten 2008).

for disinfection. This may be alleviated by prewashing a food product to remove extraneous organic matter on the surface, or a greater concentration of chlorine can be used to help ensure that more chlorine is available for disinfection. Second, chlorine species react with organic matter to form potential carcinogenic halogenated DBPs (Olmez and Kretzschmar 2009) such as trichloromethane (TCM), THMs, and HAAs (Luo *et al.* 2011; Van Haute *et al.* 2013; Shen *et al.* 2016).

Shen *et al.* (2016) studied the effect of free chlorine and organic load on the generation of HAAs and THMs in simulated produce wash water. Monobromoacetic, tribromoacetic, chlorobromoacetic and trichloroacetic acid (HAAs) as well as chloroform (THMs) were found to be major DBP components formed. López-Gálvez *et al.* (2010) detected the formations of THMs in lettuce wash water only when sodium hypochlorite was used under extreme concentrations, high chemical oxygen demand (COD) levels and long contact times. In addition, chlorine disassociates gradually into chloride and chlorate. Gil *et al.* (2016) reported that the levels of chlorate residues increased with increasing organic load and sodium hypochlorite concentration in the lettuce wash waters. Kaufmann-Horlacher *et al.* (2014) reported that post-harvest treatment of carrots using chlorinated water resulted in the formation of chlorate residues. ClO_2 on the other hand is found to be less prone to DBPs formation and is an efficient disinfectant over a wide pH range (Gordon *et al.* 1990; Pereira *et al.* 2008). Liyanage *et al.* (1997) reported that the pH has less of an influence on pathogen inactivation for viruses and cysts with ClO_2 than with chlorine in the pH range 6 to 8.5 (Liyanage *et al.* 1997). However, ClO_2 reduces in large part into chlorite (ClO_2^-) and chlorate (ClO_3^-) ions (Hua and Rechow 2007).

Fan and Sokorai *et al.* (2015) compared the levels of TCMs formation in chlorinated wash waters prepared with Cl_2 and ClO_2 , respectively, as sanitizers for washing cut-lettuce and diced onions. Higher levels of TCMs were detected in Cl_2 solutions compared to ClO_2 solutions. The type of cut-vegetable and its constituent organic compounds, as well as the type of buffering agents used to adjust the pH of sanitizers (citric acid vs. sodium phosphate) were found to have a significant effect on the TCMs formation. Huang and Batterman (2009) have measured THMs (chloroform) in 11 foods and 17 beverages prepared in chlorinated water. Their studies showed that tea formed the highest chloroform levels (up to $67 \mu\text{g L}^{-1}$), followed by coffee, rice, soups, and vegetables ($51 \mu\text{g L}^{-1}$).

4. Formation and chemistry of chlorine DBPs

Trihalomethanes are the most prevalent DBPs in drinking

water (Boorman *et al.* 1999). Chloroform (CHCl_3) is the chief THM formed by HOCl. THMs, especially chloroform, are the most studied of all DBPs due to their prevalence. Formation of chloroform has been studied in the reaction of chlorine with humic substances (Folkes *et al.* 1995; Iriarte-Velasco *et al.* 2006), triclosan (Rule *et al.* 2005), citric acid (Larson and Rockwell 1979) and resorcinol (Özbelge 2001). When in contact with chlorine species, aldehydes and ketones are converted to chloroform *via* a base-catalyzed reaction pattern (Deborde and von Gunten 2008). These reactions start with a rate-limiting enolization step that converts ketones to enols. For example, 3-ketoglutaric acid is rapidly converted into an enol because of the increased enol stability conferred by carboxylate groups within the compound (Larson and Rockwell 1979). After enolization, rapid chlorination steps yield a trichloromethyl ketone. Finally, hydroxide nucleophilic substitution in the compound results in chloroform and a carboxylic acid.

For compounds such as acetylacetone, the conversion to THM process is very similar to that of 3-ketoglutaric acid. The main difference is the early cleavage of the compound to form acetate and a chlorinated compound, which is further chlorinated before undergoing nucleophilic substitution to yield acetate and chloroform. In aromatic compounds such as humic acids, enolization is followed by chlorination. In most cases, 2-chloro, 2,4-dichloro, and 2,4,6-trichloro phenolic and resorcinolic compounds are among the first compounds formed when aromatic phenols are the substrates (Rule *et al.* 2005; Deborde and vonGunten 2008). The chlorination continues until the methyl group is trichlorinated. Finally, hydroxide attacks the resulting trihaloketone to yield chloroform and a carboxylic acid. Alcohols are other potential substrates that will react with chlorine to form chloroform. It is suggested that chloroform formation can occur through the mixing of bleach and alcohols such as isopropanol and ethanol. Because the reactions are very slow, only a few studies on the chlorine reactivity with alcohols have been undertaken (Prütz 1996).

Chloramines can be categorized as another class of DBPs formed by chlorine. The formation of chloramines involves the addition of chlorine to amine groups or ammonia to form monochloramine, dichloramine or trichloramine (Abia *et al.* 1998). Monochloramine is the most common haloamine. Many animals are sensitive to chloramines, and chloramines are toxic to some animals, such as fish. The particular chloramine yielded from the reaction of HOCl and amines is dependent on chlorine-to-nitrogen ratio, pH, temperature and contact time (Donnermair and Blatchley 2003). If there is significantly more nitrogen than chlorine, monochloramine will be the dominant product (White 1999). HOCl reacts with both organic and inorganic amines in solution, but the distribution depends on affinity for the

organic and inorganic nitrogen compounds, distribution of the nitrogen compounds and pH (Yoon and Jensen 1993). The reactivity of chlorine to NH_4^+ (species present at acidic pHs) is negligible. Most of the chlorination of amines happens at pH 8–9 ($K_{\text{app}} > 10\,000 \text{ mol L}^{-1} \text{ s}^{-1}$) (Deborde and von Gunten 2008).

HAAAs are a class of organic halides that are formed when halogenation takes place at methyl hydrogens in acetic acid. The formation of haloacetic acids is pH-dependent; however, more is known about THMs than HAAAs. Among the nine HAAAs known, five of them are regulated in drinking water; they are monochloro-, monobromo-, dichloro-, dibromo-, and trichloroacetic acid (Liang and Singer 2003). Like THMs, HAAAs can be produced from reactions of humic and fulvic substances with chlorine. Dichloro- and trichloroacetic acids account for the greater majority of HAAAs formed, with dichloroacetic acid being the most common (Shin *et al.* 1999; Fahimi *et al.* 2003). The rates of these reactions are pH dependent. In the presence of resorcinolic compounds, pH below 7 yield mostly HAAAs while pH above 7 yields mostly THMs (Liang and Singer 2003). Therefore, pH appears to play a large role in the determination of DBP species formed.

5. Health concerns of chlorinated disinfection byproducts

Chloroform causes carcinogenesis by nongenotoxic methods, namely decreased methylation which leads to overexpression of proto-oncogene (Coffin *et al.* 2000). The authors speculate that THMs, in general, cause a decreased methylation of the c-myc proto-oncogene, which leads to the gene's subsequent uncontrolled expression. It has also been speculated that THMs present in disinfected drinking water leads to an increase in bladder and colorectal cancers

(Villanueva *et al.* 2006). Despite this, it is uncertain if the health risks are serious enough to warrant stricter regulation. Although direct contact by oral gavage with CHCl_3 has been shown to cause tumorigenesis in mice, CHCl_3 administered in drinking water has not (Coffin *et al.* 2000). The reason for this is that the incremental ingestion of chloroform through water does not promote the liver's detoxification mechanisms as opposed to oral gavage, which provides more chloroform at once. This is the reason why chloroform has been classified as a threshold carcinogen (US EPA 1994). A threshold carcinogen is defined as a substance that is only carcinogenic after a certain level, or threshold, is reached. Since this is the case, it is unlikely that food, which is not expected to contain a substantial amount of chloroform, would be a major route of chloroform exposure.

Among HAAAs, trichloroacetic acid and dichloroacetic acid have been shown to cause liver tumors in rats and mice (Boorman *et al.* 1999). It is speculated that these two compounds have different modes of carcinogenicity. Dichloroacetic acid is thought to modify intracellular signaling pathways (US EPA 1998), while trichloroacetic acid is thought to be related to peroxisome proliferation (Bull *et al.* 1990). Chloramines, as stated previously, are useful as disinfection agents. However, chloramines lack the effectiveness of other chlorine-based disinfectants, as they showed little to no effect on *Escherichia coli* (Donnermair and Blatchley 2003). Trichloroamine may be linked to an increase in asthma. Chloramine toxicity is mostly a concern in water rather than food. Chlorite and chlorate can cause anemia in some animals, and high levels are harmful to thyroid function (Hebert *et al.* 2010).

For toxicity comparison, Table 1 summarizes the acute toxicity of chlorinated compounds and the US EPA control limits on these compounds in drinking water. This govern-

Table 1 Toxicity and regulatory information of several chlorine compounds (US EPA 1998)

Compound	LD ₅₀ ¹⁾ (rat, oral) (mg kg ⁻¹)	US government control ²⁾
Hypochlorous acid	>3 000	MRDL=4 mg L ⁻¹
Sodium hypochlorite	5 000 (at 12.5%)	None
Chloroform	908	MCL=0.08 mg L ⁻¹ for total trihalomethanes
Bromodichloromethane	916	MCL=0.08 mg L ⁻¹ for total trihalomethanes
Chlorodibromomethane	1 186	MCL=0.08 mg L ⁻¹ for total trihalomethanes
Monochloroacetic acid	76	MCL=0.06 mg L ⁻¹ for total haloacetic acids
Dichloroacetic acid	2 820	MCL=0.06 mg L ⁻¹ for total haloacetic acids
Trichloroacetic acid	5 000	MCL=0.06 mg L ⁻¹ for total haloacetic acids
Chloramine	935	MRDL=4 mg L ⁻¹ for total chloramines
Dichloramine	No data available	MRDL=4 mg L ⁻¹ for total chloramines
Trichloramine	No data available	MRDL=4 mg L ⁻¹ for total chloramines
Chlorine dioxide	292	MRDL=0.8 mg L ⁻¹
Chlorite	292	MCL=1 mg L ⁻¹
Chlorate	1 200–7 000	Not specified

¹⁾ LD₅₀, the median lethal dose required to kill half the members of a tested population after a specified test duration.

²⁾ MRDL, maximum residual disinfectant level; MCL, maximum contaminant level.

ment control limits information help illustrate the relative toxicity of each compound.

6. pH of chlorine on the reaction rate of DBP formation

Many studies have been conducted to evaluate the role of HOCl and OCl⁻ in the production of DBPs with a particular interest in the production of THMs such as chloroform. Tables 2 and 3 are a summary of information from the literature that has examined the role of different species of chlorine as affected by pH in the formation of DBPs with different substrates.

The use of aromatic compounds in the study by Chaidou *et al.* (1999) is significant because these compounds approximate humic substances, which are ubiquitous in water. Also, the decision to use chlorinated phenols as reactants in some of the tests is probably to approximate partially-chlorinated intermediates of humic substances. In many cases, the concentration of chloroform DBP decreases as

pH decreases, as indicated in the literature (Tables 2 and 3). The work done by Özbelge (2001) indicated the same trend of reduced chloroform formation as pH decreases. In that study, the pH of hypochlorite was adjusted to pH 4, 7 and 10. The hypochlorite was then added to resorcinol in a ratio of 1:1 (resorcinol:chlorine), 1:3 and 1:10. No chloroform was formed at pH 4 for the 1:3 ratio, but 50% of the resorcinol was converted to chloroform and 95% of the resorcinol was converted to chloroform at pH 7 and 10, respectively. At the 1:10 ratio, 32 and 82% of the resorcinol was converted to chloroform at pH 4 and 7, respectively. Chloroform formation at pH 10 was not studied at this ratio.

Waters and Hung (2014) measured the THM reaction products from chlorinated water at different pHs with resorcinol. They found chloroform accounted for the largest percentage of THMs detected in total samples, indicating that it was the main THM product when chlorine reacted with resorcinol. The data indicated that the chlorinated water at pH 6 and 9.3 when reacting with resorcinol produced 10 times more chloroform (9.54 and 12.4 mg L⁻¹, respectively) than chlorinated water at pH 2.5 (1.1 mg L⁻¹). Total other THM products (dichloromethane, dibromochloromethane, dibromomethane, bromoform) and haloacetic acids (chloroacetic acid, bromoacetic acid, dichloroacetic acid, trichloroacetic acid, bromochloroacetic acid, dibromoacetic acid) were at µg L⁻¹ level and were about 10 times less than chloroform. They also found the pH of chlorinated water affected the yield of chlorine byproduct formation, as well as the type of chlorine byproduct formed. Increases in pH led to the formation of chloroform as a chief THM product.

Flischacker and Randtke (1983) found that the yield of organic chlorine formation has increased dramatically with decreasing pH while the opposite trend has been observed in the case of chloroform formation during the reaction of free and combined chlorine with pelvic acid. No observable inflection in the formation of organic chlorine has been detected at pH 7.6 to explain the effect of HOCl and OCl⁻. The presence of other chlorine species such as molecular chlorine, protonated HOCl and chlorine monoxide (Cl₂O) at acidic pH (~3) was believed to be the contributing factors for high organic chlorine formation. Further, the changes in organic molecules with pH were found to have a significant effect on chloroform formation.

Obolensky and Singer (2008) studied the THMs formation models and found that the pH has an effect on 3 of the 8 DBPs. They found that pH has a significant effect on CHCl₃ formation, but pH is not identified as an important predictor for the brominated THMs. Trihaloacetic acids were found to have an inverse correlation with pH while dihaloacetic acids were not affected by pH changes. Cowman and Singer (1996) found that elevated pH has a greater negative impact on brominated HAAs than on chlorinated HAAs.

Table 2 The formation of chloroform from the reactions of organic compounds with sodium hypochlorite at different pH

Substrate	Chloroform formed (mg L ⁻¹)		Reference
	pH≤7	pH≥8	
Hexane	0.15	0.1	Chaidou <i>et al.</i> (1999)
Benzene	0.1	0.12	Chaidou <i>et al.</i> (1999)
Toluene	0.11	0.2	Chaidou <i>et al.</i> (1999)
Ethylbenzene	0.3	0.3	Chaidou <i>et al.</i> (1999)
Styrene	0.35	0.4	Chaidou <i>et al.</i> (1999)
<i>m</i> -Xylene	0.55	0.55	Chaidou <i>et al.</i> (1999)
2-Ethyltoluene	0.1	0.12	Chaidou <i>et al.</i> (1999)
Naphthalene	0.23	0.25	Chaidou <i>et al.</i> (1999)
Acetone	2.2	3.6	Chaidou <i>et al.</i> (1999)
Isopropanol	0.7	1	Chaidou <i>et al.</i> (1999)
1,2-Dichlorobenzene	0	0.1	Chaidou <i>et al.</i> (1999)
1,4-Dichlorobenzene	0	0.1	Chaidou <i>et al.</i> (1999)
Aniline	3	3.2	Chaidou <i>et al.</i> (1999)
Nitrobenzene	0.2	0.3	Chaidou <i>et al.</i> (1999)
2-Chloroaniline	3	3.2	Chaidou <i>et al.</i> (1999)
3-Chloroaniline	1	1.3	Chaidou <i>et al.</i> (1999)
4-Chloroaniline	3	3	Chaidou <i>et al.</i> (1999)
Phenol	0.7	1	Chaidou <i>et al.</i> (1999)
Hydroquinone	0	1.7	Chaidou <i>et al.</i> (1999)
Resorcinol	11	11.2	Chaidou <i>et al.</i> (1999)
Catechol	0	0.2	Chaidou <i>et al.</i> (1999)
Pyrogtotalol	0	0.2	Chaidou <i>et al.</i> (1999)
Phloroglucinol	7.5	13	Chaidou <i>et al.</i> (1999)
2-Chlorophenol	2	2.2	Chaidou <i>et al.</i> (1999)
3-Chlorophenol	2.2	2.4	Chaidou <i>et al.</i> (1999)
4-Chlorophenol	1	1.2	Chaidou <i>et al.</i> (1999)
<i>o</i> -Cresol	1	1.8	Chaidou <i>et al.</i> (1999)
<i>m</i> -Cresol	1	1.5	Chaidou <i>et al.</i> (1999)
<i>p</i> -Cresol	0	0.4	Chaidou <i>et al.</i> (1999)
Humic acid	0.074	0.095	Dowd (1994)
Resorcinol	1.10	9.54	Waters and Hung (2014)

Table 3 Reaction rates of different organic substrates with free chlorine for the formation of chlorinated DBPs as a function of pH

Substrate	Rate of product formation (mol L ⁻¹ s ⁻¹)		Product formed	Reference
	pH≤7	pH≥8		
Bromide	1.55×10 ³	9×10 ⁻⁴	OBr ⁻	Kumar and Margerum (1987)
Sulfite	7.6(±0.4)×10 ⁸	2.3(±0.2)×10 ⁴	ClSO ₃ ⁻	Fogelman <i>et al.</i> (1989)
Cyanide	1.22(±0.03)×10 ⁹	2.3(±0.2)×10 ⁴	ClCN	Fogelman <i>et al.</i> (1989)
Cysteine	1.2×10 ⁹	1.9×10 ⁵	Cystiene disulfide	Armesto <i>et al.</i> (2000)
Methionine	3.3×10 ⁸	5.5×10 ⁵	Sulfoxides	Pattison and Davies (2001)
N-Methylformamide	1.70×10 ⁻³	1.82×10 ⁻²	Chlorinated amide	Thomm and Wayman (1969)
N-Methylacetamide	1.70×10 ⁻²	9.20×10 ⁻³	Chlorinated amide	Thomm and Wayman (1969)
(N,N)-Dimethylurea	0.82	0.0083	Chlorinated amide	Thomm and Wayman (1969)
Gemfibrozil	10 ⁰	10 ^{-0.75}	Chloramines, chloroform	Deborde and Gunten (2008)
Naproxen	10 ^{0.5}	10 ⁰	Chloramines, chloroform	Deborde and Gunten (2008)
Trimethoprim	10 ^{1.75}	10 ^{1.5}	Chloramines, chloroform	Deborde and Gunten (2008)
Indometacine	10 ^{1.75}	10 ¹	Chloramines, chloroform	Deborde and Gunten (2008)
Enrofloxacin	10 ^{2.5}	10 ^{2.5}	Chloramines, chloroform	Deborde and Gunten (2008)
sulfamethoxazole	10 ³	10 ^{2.75}	Chloramines, chloroform	Deborde and Gunten (2008)
Sulfamedimethoxine	10 ⁴	10 ^{3.75}	Chloramines, chloroform	Deborde and Gunten (2008)
Ciprofloxacin	10 ^{5.75}	10 ⁶	Chloramines, chloroform	Deborde and Gunten (2008)
Acetaminophen	10 ¹	10 ^{1.25}	Chloramines, chloroform	Deborde and Gunten (2008)
4- <i>n</i> -Nonylphenol	10 ¹	10 ^{1.25}	Chloramines, chloroform	Deborde and Gunten (2008)
Triclosan	10 ^{2.5}	10 ³	Chloramines, chloroform	Deborde and Gunten (2008)
Bisphenol A	10 ^{1.75}	10 ²	Chloramines, chloroform	Deborde and Gunten (2008)
Estrogenic steroid hormones	10 ²	10 ^{2.5}	Chloramines, chloroform	Deborde and Gunten (2008)
<i>p</i> -Iodophenol	8	30	Chloroform	Gtotalard and Gunten (2002)
<i>p</i> -Methylphenol	10	50	Chloroform	Gtotalard and Gunten (2002)
<i>p</i> -Cyanophenol	8	10	Chloroform	Gtotalard and Gunten (2002)
Phenol	10	50	Chloroform	Gtotalard and Gunten (2002)
<i>p</i> -Chlorophenol	8	20	Chloroform	Gtotalard and Gunten (2002)
Ammonia	8.9×10 ³	2.6×10 ⁴	Monochloramine	Qiang and Adams (2004)

7. Minimizing safety and health concerns on using chlorine-based sanitizers

It is clear that the DBPs' formation is influenced by various factors such as the type of chlorine based sanitizers, the concentration of chlorine, contact time, type of organic matter, organic load, and physico-chemical properties of sanitizers and organic molecules. It was reported that the levels of added chlorine, and its reaction with organic matter with time and concentration are the significant factors contributing to the DBPs formation. Further, pH of the chlorine-based sanitizers is found to have a significant effect on the yield of DBPs formation. By considering these factors, four potential strategies can be implemented to minimize the formation of DBPs in various food processing operations.

7.1. Use of numerical models to predict chlorine demand

One established practice in the food processing industry is to replenish the treatment solutions with excess amounts of chlorine to meet the required chlorine demand and target levels of disinfection. To maintain sufficient levels of free chlorine, replenishing chlorine using continual fixed dosing,

automated demand-based injection systems, and manual demand-based periodic dosing is a common practice (Shen *et al.* 2016). However, studies reported that repeatedly adding an additional amount of chlorine into washing solutions that are high in organic loads can also lead to the formation of toxic chlorine byproducts such as THMs, HAAs, halo ketones and chloropicrin (Connell *et al.* 1996; Gil *et al.* 2009). Empirical models to predict the chlorine demand for specific process treatment were made available. Chen and Hung (2016) developed equations to predict the chlorine demand for different produce items based on wash water quality parameters. According to their models, chlorine demand of a specific produce item can be predicted based on factors such as phenolic to protein ratio (PPC) and the ultraviolet absorbance at 254 nm (UV254). Industry can make use of such predictive models to avoid the use of excessive amounts of chlorine and minimize potential DBPs formation.

7.2. Use of sanitizers at near neutral pH

The antimicrobial efficacy of chlorine largely depends on the pH and the amount of organic material in the water, and to a limited extent, on the temperature of water (Suslow 2001). Both highly acidic and basic conditions are not found to be

beneficial in achieving high antimicrobial efficacy, equipment protection, and minimizing the formation of toxic DBPs. Therefore, it is desirable to maintain the pH of solutions at near neutral conditions (6 to 7) to maximize the HOCl levels for optimal disinfection efficacy and minimize the potential DBPs formation. Several recent studies (Abadias *et al.* 2008; Guentzel *et al.* 2008; Jadeja and Hung 2014; Fang *et al.* 2016) demonstrated the disinfection potential of near neutral electrolyzed oxidizing (NNEO) water at around pH 6–7. Hence, NNEO water can be used as one promising alternative to chlorine to minimize DBPs formation and achieve target disinfection. A study by Gómez-López *et al.* (2013) reported that process wash waters treated with sodium hypochlorite showed higher levels of THMs compared to EO water treatment. However, treatment of EO water with added salts showed increased THM levels. They concluded that hyper chlorination might lead to the formation of THMs.

7.3. Use of post-rinsing treatment of fresh produce

Several studies reported that the levels of DBPs on actual foods such as fresh produce were greatly minimized by introducing a rinsing step followed by chlorine treatment. For example, Klaiber *et al.* (2005) found that chlorine by-products due to chlorination in minimally processed carrots were negligible after rinsing with tap water. Gómez-López *et al.* (2013) found that THMs were detected at low levels in spinach washed with sodium hypochlorite solution because the rinsing step reduced them to levels below the detection limit. However, the post-rinsing step does not help to avoid the risk associated with the presence of DBPs in process wash water.

7.4. Use of combination treatments

Advanced oxidation processes (AOPs) involving the use of oxidants such as H₂O₂, O₃, Cl₂, ClO₂, and metal oxides along with UV light have been used for several disinfection and decontamination applications (Sun *et al.* 2016a). AOPs take advantage of highly reactive species such as hydroxyl radicals (\cdot OH) for degradation of toxic chemical contaminants. Studies have been reported that combination of UV/chlorination is more efficient in degradation of various chemical compounds, contaminants, pesticides, herbicides, fungicides, and etc. (Kent *et al.* 2011; Sichel *et al.* 2011; Guo *et al.* 2016). In large, the resultant reactive chlorine and oxygen species such as Cl \cdot , Cl₂ \cdot^- , ClO \cdot and \cdot OH radicals during pH dependent UV/chlorination combination treatments were believed to be the main contributors for the enhanced degradation of targeted compounds when compared to individual treatments (Sun *et al.* 2016b).

Sichel *et al.* (2011) reported that contaminants such as

estrogen, benzotriazole, and tolytriazole were degraded by AOPs following the order of UV/HOCl>UV/H₂O₂>UV/ClO₂ with the formation of low levels of THMs and little *N*-nitrosodimethylamine (NDMA) by UV/HOCl. However, contrasting results were reported on the effect of DBPs formation of the UV/chlorine processes. For example, Guo *et al.* (2016) and Ben *et al.* (2016) found that UV/chlorination treatment enhanced the formation of DBPs due to the generation of transient, more reactive precursors. Meanwhile, Wu *et al.* (2016) reported that UV/choline treatment resulted in a less degree of halogenated DBPs formation compared to chlorination alone. Thus, the effect of the UV/chlorine process on the formation of chlorinated products and alteration is still uncertain and needs further evaluation (Wu *et al.* 2016). On the other hand, treatments such as the combination of UV and TiO₂ removed most HAA and some THM precursors from natural organic matter in water (Kent *et al.* 2011).

8. Conclusion

With the incidence of foodborne pathogens in food on the rise, it becomes increasingly important for consumers to adopt measures to ensure food safety. Chlorine-based sanitizers are still the most popular chemicals to ensure food safety. There is a concern that the production of toxic byproducts will negate the health benefits of treating food or drinking water with the chlorine-based disinfectants. Based on the information in this review, pH plays an important role in the determination of the type and amount of DBPs formed, with lower, more acidic, pHs resulting in the formation of less chloroform. This review also stresses the need for more research on the link between pH and THM as well as HAA production, particularly in food-related sanitation.

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