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Review

Chlorination byproducts, their toxicodynamics and removal from drinking water

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Abstract

No doubt that chlorination has been successfully used for the control of water borne infections diseases for more than a century. However identification of chlorination byproducts (CBPs) and incidences of potential health hazards created a major issue on the balancing of the toxicodynamics of the chemical species and risk from pathogenic microbes in the supply of drinking water. There have been epidemiological evidences of close relationship between its exposure and adverse outcomes particularly the cancers of vital organs in human beings. Halogenated trihalomethanes (THMs) and haloacetic acids (HAAs) are two major classes of disinfection byproducts (DBPs) commonly found in waters disinfected with chlorine. The total concentration of trihalomethanes and the formation of individual THM species in chlorinated water strongly depend on the composition of the raw water, on operational parameters and on the occurrence of residual chlorine in the distribution system. Attempts have been made to develop predictive models to establish the production and kinetics of THM formations. These models may be useful for operational purposes during water treatment and water quality management. It is also suggested to explore some biomarkers for determination of DBP production. Various methods have been suggested which include adsorption on activated carbons, coagulation with polymer, alum, lime or iron, sulfates, ion exchange and membrane process for the removal of DBPs. Thus in order to reduce the public health risk from these toxic compounds regulation must be inforced for the implementation of guideline values to lower the allowable concentrations or exposure.

Keywords: Adsorbent; Chlorination byproducts; Disinfection byproducts; Toxicodynamics; Natural organic matter

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1. Introduction

There are many sources of contamination in drinking water. These contaminants in ground and surface water are natural substances leaching from soil, runoff from agricultural activities,

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discharges from sewage treatment/industrial plants and hazardous chemicals from landfill sites. It is reported that nearly half of the population in the developing countries suffers from health problems associated with lack of potable drinking water as well as the presence of microbiologically contaminated water [1]. Disinfection by chlorination is the most important step in water treatment for public supply as chlorine remains in the water as long as it is not consumed. However, chlorine also reacts with the natural organic matter (NOM) present in the water and produces a number of byproducts with harmful long-term effects.

Chlorine and its compounds are the most commonly used disinfectants for the treatment of water and its popularity is due to higher oxidizing potential, provides a minimum level of chlorine residual throughout the distribution system and protects against microbial recontamination [2]. Use of chlorination reduces the risk of pathogenic infection but may pose chemical threat to human health due to disinfection residues and their byproducts. DBPs will be produced upon chlorination only if the water contains DBP precursors. During chlorination of water containing natural organic matter a complex mixture of chlorine byproducts is formed and more than 300 different types of DBPs have been identified [3]. The formation of these compounds in drinking water depends on several other factors such as temperature, pH, dose, contact time, inorganic compounds and natural organic matter present in the drinking water supply.

The formation of chlorinated DBPs in drinking water like trihalomethanes has emphasized the need for exploring alternate disinfectants and new treatment technologies. The trihalomethanes viz. chloroform, dichlorobromomethane, dibromochloromethane and bromoforms are the major byproducts of chlorination. In addition to trihalomethanes, haloacetic acids and haloacetonitriles are the products of both chlorination and chloramination. Chloramination also leads to the production of cyanogen chloride and *N*-organochloramine. Chlorine dioxide is a strong water disinfectant over a wide pH range. It is most efficient in killing bacteria and especially successful in deactivating viruses [4]. It produces mainly oxidized products and does not form THMs. The disadvantage is the potential production of the undesired inorganic species, i.e. chlorite and chlorate ions [5]. Chlorine dioxide will produce low levels of organic byproducts as compared to chlorine disinfection [6].

Formation of chlorinated acetic acid is also reported from organic material during the chlorination. The typical level of this chlorinated acetic acid in finished drinking water supply is reported from 30 to 160 μg/l [7]. Another byproducts of water chlorination is chloral hydrate which has been detected from 0.01 to 100 μg/l in the drinking water. Chloramination is the most commonly alternative to chlorine resulted a significant decrease in the byproduct formation than with the use of chlorine [6], however its use requires longer contact time because it is less effective than chlorine. Moreover, it tends to produce volatile products responsible for tastes and odors.

It is suggested that the spectrum of these byproducts mostly depends upon humic acid content and total organic carbon (TOC) as well as the quantity of inorganic chemicals in the water supply [8]. The use of chlorine dioxide is of a particular interest because it does not produce the organic disinfection byproducts common to chlorination practices, Further it has also been found effective for inactivation of chlorine resistant pathogen species such as *Giardia* and *Cryptosporidium* [9]. Recently it has been used for the control of taste/odour and removal of iron, manganese and hydrogen sulfide [10].

Disinfectants have varying capacities to inactivate or kill the pathogens and its action depends on the type and nature of organisms as well as the process conditions including temperature, pH contact time and dose. The influence of heating on the formation and behaviour of disinfection byproducts showed thermal cleavage of larger halogenated species subsequently leads to the formation of smaller chlorinated molecules and dechlorination of organics [11].

Aside from the regulated disinfection byproducts (DBPs), there are thousands of other compounds formed from the reaction of disinfectant with the substances in the water. Products from the reaction between oxidizing disinfectants and NOM or naturally occurring inorganic constituents are bonafide DBPs. A wide range of PPCPs has been detected in a variety of environmental samples at levels ranging from ng kg^{-1} upto g kg^{-1} [12]. During the wastewater treatment process, the parents PPCPs, conjugates and metabolites may be: (i) completely transformed to CO_2, (ii) partially transformed producing metabolites or (iii) unchanged [13]. Nonylphenol polyexthoxylated (NPnEOs) and nonylphenol (NP), a metabolite of NPnEOs have been detected at concentrations as high as 981 mg/kg (dry mass) and 1380 mg/kg (dry mass), respectively [14,15]. Brominated diphenyl ethers, commonly used fire retardants, have been detected at 32–4890 μg/kg (dry mass) in biosolid from several wastewater treatment plants [16].

Some PPCPs are thought to cause abnormal sexual development in fish [17]. PPCPs are being studied for their potential to cause endocrine disrupting behavior. These can be prescription and non-prescription drugs, steroids, insect repellents, detergent metabolites, disinfectants, plasticizers, fire retardants, antioxidants, fragrances, polyaromatic hydrocarbons and solvents [18].

2. Predictive models

In the recent years much attention has been paid to develop predictive models in order to determine the production and the kinetics of DBP formation [19–21]. It is reported that the chlorination serves as oxidant in addition to kill the pathogens. As such, it is also used for the removal of taste and odour, the oxidation of some metals thereby enhancing the efficiency of coagulation and filtration, the prevention of algal growth and re-occurrence of microorganisms in the water distribution system [22]. In this process chlorine gas (Cl_2) is bubbled into pure water, which leads to the formation of hydrochloric acid (HCl) and hypochlorous acid (HOCl), by rapid hydrolysis [23]. HOCl undergoes to subsequent reactions with dissolved organics resulting in the formation of trihalomethanes (THM). HOCl oxidizes the bromide (Br^-) present in the water, which reacts readily with natural organic matter (NOM) to form brominated halomethanes. Therefore, trihalomethane formation is influenced by chlorine dose, concentration and nature of NOM, contact time, pH, temperature of water bromide ions

occurrence and concentration [24]. In temperate environments, trihalomethane levels in drinking water are significantly affected by seasonal variations [25–28] suggested that chlorine decay rate follows a second order kinetic law that includes simultaneous THM formation. This model was designed and applied successfully to the rechlorination.

Since chlorine is the most popular and traditional disinfectant, most of modeling has been focused on THMs. Recently attempts have been made to combine the mechanistic and empirically based approaches on the modeling [2]. Multiple linear and non-linear regression technique is found to be the most common in developing DBPs predictive models [2]. These DBPs models can be useful for operational purposes during water treatment and water quality management for the evaluation of water treatment facilities, exposure assessment in epidemiological studies, health risk assessment and also for estimating the impacts of DBP regulation. The development of models on the toxicodynamics of DBPs may be helpful to deal with the fate of the species present in drinking water. The DBP models can also be useful to determine the human exposure of these compounds through various contact routes. Three exposures routes viz. ingestion, inhalation and dermal contact are considered in this analysis. The exposure assessment results can be further used in human health risk assessment. It is noticed that aqueous bromine reactions with NOM are much faster than aqueous chlorine. The speciation and concentrations of DBP formation in chlorinated process are mainly dominated by the rate of bromide to reactive NOM. During chlorination of water containing bromide, the typical mode rate of applied chlorine to the ambient concentration of bromide is approximately in the order of 10 [29–31]. More recently it has been demonstrated that the presence of copper in water enhances THMs formation as copper is known to catalyse a number of reactions that are similar to the conventional haloform reactions [32].

3. Health effects and epidemiological studies

The addition of oxidants to water generates a variety of disinfection byproducts, which have been found to be associated with adverse health effect [33,34]; Glaze et al., 1986. Chlorination is the most widely used cost effective method of disinfection practiced throughout the world and leads to control of water borne diseases [35]. Association between the ingestion of chlorinated drinking water in excess with risk of bladder and rectal cancer followed by mortality have been reported in several epidemiological studies [36]. An apparent association between bladder cancer, reproductive disorders and trihalomethane occurrence has also been established [37]. The most precise information about risk of cancer and contamination of drinking water comes from water quality surveillance. A study was conducted to determine the types of cancer associated with surface water and strength and consistency of such association [38]. Cancer of the colon, rectum, and urinary bladder was noticed to be linked with many settings of water sources containing the elevated level of chlorine byproducts. In addition, several other cancer sites namely stomach, brain, pancreas, lung and liver were also found to be linked with chlorinated byproducts (CBPs). There have been some epidemiological evidences of a relationship between the exposure to DBPs and adverse reproductive outcomes in human beings and animal studies [39,40]. It is being considered that the introduction of some biomarkers could be a specific measure of DBP production. Because this is a complete mixture of the chemical species which can be quite variable over the time due to range of other factors such as seasonality [41]. However, the epidemiological evidences showed that the main groups are trihalomethanes and haloacetic acid, which are the risk of some cancers [42]. Studies conducted on mammals revealed that THMs induces neurotoxicity, hepatotoxicity, reproductive toxicity and nephrotoxicity [43] (Table 1).

4. Different techniques for the removal of CBPs

Various options are applicable for the removal of water pollutants included reverse osmosis, ion exchange, coagulation, co-precipitation, catalytic reduction, herbal filtration, electrodialysis and adsorption [44]. Chlorine based treatment techniques have the property to remain active in the water as long as they are not consumed by either inactivation or competitive reaction. Water treatment designs and operators have only alternatives either limit the formation of disinfection byproducts by innovative chlorination strategies or to develop process for the removal of organic and other chlorine sensitive compound [45]. Synthetic and natural organic contaminants are mostly found in drinking water. These compounds include taste and odor causing synthetic organic chemicals, pesticides, herbicides color and trihalomethane precursors. NOM is produced in the biological degradation of organic substances viz. amino acids, fatty acids, phenols, steroids, sugars, hydrocarbons, urea, porphyrines and

Table 1
Chlorination byproducts and its health effects

Class of DBPs	Compounds	Health effects
Trihalomethanes (THM)	Chloroform	Cancer, liver, kidney and reproductive effects
	Dibromochloromethane	Nervous system, liver, kidney and reproductive effects
	Bromodichloromethane	Cancer, liver, kidney and reproductive effects
	Bromoform	Cancer, liver, kidney and reproductive effects
Haloacetonitrile (HAN)	Trichloroacetonitrile	Cancer, mutagenic and clastogenic effects
Halogenated aldehydes and ketones	Formaldehyde	Mutagenic
Halophenol	2-Chlorophenol	Cancer and tumor promoter
Haloacetic acids (HAA)	Dichloroacetic acid	Cancer and reproductive and developmental effects
	Trichloroacetic acid	Liver, kidney, spleen and developmental effects

polymers. The polymers include polypeptides, lipids, polysaccharides and humic substances [46]. Drinking water sources contain 2–10 mg/l of NOM, although much higher levels are also reported [47]. The dissolved and colloidal fraction of NOM or DOM serves as major precursors in these reactions. DOM is a heterogeneous mixture of various organic molecules originated from aquagenic (biota in water body) and pedogenic processes (soil and terrestrial vegetation). Earlier a number of methods have been developed ranging from sorption by synthetic resin to separation by membranes for DOM isolation or fractionation.

Removal of DBPs and their precursors is very low when using coagulation by organic polymers, physical processes like settling, deep bed filtration or pressure driven porous membrane processes.

Therefore the introduction of adsorption process is found to be the most suitable and significant for its removal. Granular activated carbon (GAC) and powdered activated carbon (PAC) have been used to remove these organic compounds from the water. It is suggested that the formation of the compounds with NOM and chlorine are referred as THM precursors and the complete reaction is known as trihalomethane formation potential (THMFP) and can be determined directly or by measuring total organic carbons (Fig. 1).

Prechlorination is commonly practiced for the control of bacterial and algal growth in the treatment plants. In this case THM formation reaction starts prior to addition of PAC and therefore the oxidation–reduction reaction between the chlorine and surface of activated carbon inhibited the adsorption capacity of the activated carbon for phenolic compounds [48]. Therefore the removal of THM precursor and THMs from drinking water by PAC is variable. The fraction of NOM that produces THMs varies in the different water sample. However the data indicate that there is no correlation between the level of THM precursor and TOC in the different type of water.

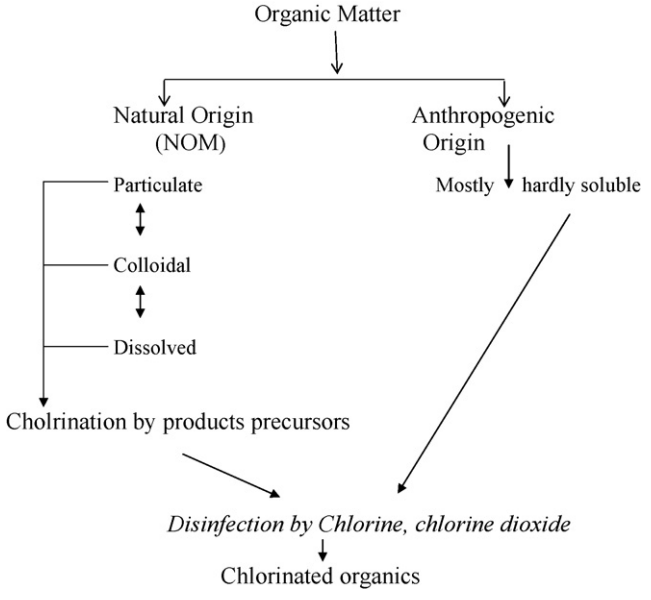

Fig. 1. Suitable diagram showing the formation of THM precursor and complete reaction.

Granular activated carbon has been suggested due to its greater efficiency in the removal of NOM, THMs, odor, color containing compound and other toxic chemicals [49,50]. The addition of Ag^+/H_2O_2 was found to reduce the formation of DBPs and H_2O_2 act as an quenching agent in addition to its property of disinfection with Ag^+ [51]. Iron oxide coated sand was found to be useful as sorbent for accumulating NOM from water source and for removing DBP precursor from water supply. Humic substances are known to be DBP precursors, a substantial fraction of its formation potential can be removed by sorption of NOM on to oxide surfaces which form during coagulation with iron and aluminum salt. The use of Fe(II) noticed to be a most promising of excess ClO_2 removal technique and has been successfully used in pilot and full scale studies [52,53]. Other alternatives have been studied including adsorption on activated carbon, coagulation with polymer, alum, lime or iron sulfate, ion exchange and membrane process for the removal of DBPs [54,55].

Bacterial growth potential and trihalomethane formation potential (THMFP) were investigated in relation to DOM before and after alum treatment, and results indicated that DBP of terrestrially derived DOM can be high in comparison with that of overall DOM in natural water and alum coagulation was not found to be sufficient to produce microbiologically stable drinking water [56]. The efficiency of NOM removal by adsorption in the column is significantly greater than in conventional coagulation particularly at the beginning of a fresh absorbent surface. It is established that the activated carbon obtained from apricot stones by pyrolysis can be used for removal of trihalomethanes from water treated with chlorine [57].

5. Guideline values

A guideline value represents the concentration of a constituent that does not result in any significant risk to health of consumer over a lifetime of consumption. Identification of disinfection byproducts and concern over the possible adverse health effects of these compounds has promoted considerable research activity in USA and Europe in order to minimize the risk of cancers. United State Environmental Protection Agency (USEPA), World Health Organization (WHO) and the European Union (EU) introduced regulations for THMs in drinking water. In 1979 the United States EPA initiated a regulatory standard of 100 μg/l for THMs under the safe drinking water act. A level of 60 μg/l has also been introduced for sum of five haloacetic acid and 10 μg/l for bromate [58]. In addition to DBP regulation, requirement was imposed to reduce DBP precursors through the use of coagulation and granular activated carbon adsorption. In Canada the regulatory situation is comparable to Australia and accordingly standards are laid down. In Germany the guideline value for total THMs is 10 μg/l. The European Communities (EC) drinking water quality standard for total THMs of 100 μg/l is currently under review [59]. To assure consumers that drinking water is safe and can be consumed without any risk, guidelines or standards have to be set up giving maximum allowable concentrations of compound in drinking water (Table 2).

Table 2
Chlorination byproducts and guideline values in drinking water

Chlorination byproducts	WHO guideline value μg/l
Trihalomethanes	
Chloroform ($CHCl_3$)	200
Bromodichloromethane ($CHBrCl_2$)	60
Bromoform ($CHBr_3$)	100
Dibromochloromethane (CHBr2Cl)	100
Haloacetic acid	
Dichloroacetic acid ($Cl_2CHCOOH$)	50
Trichloroacetic acid (Cl_3CCOOH)	100
Haloacetonitriles	
Dichloroacetonitrile (Cl_2CHCN)	90
Halocarbonyl compounds	
Chloral (CCl_3CHO, H_2O)	10

6. Toxicological measures

In several areas toxicological evaluations can be used to assess the exposure in environmental epidemiology. Toxicokinetics provides information on the fate of environmental chemicals and development of more sensitive method in the laboratory with lower detection limit. There have been considerable developments in this area over the years as epidemiologists have to introduce more biomarkers of exposure [60]. It is a complimentary discipline to toxicology where the data relate to the effect of chemical hazards on animals usually at much higher doses and under more controlled conditions than occur in human situation. These developments have given the environmental epidemiologist a larger range of tools for inclusion in human studies and should increase the statistical power in the studies [61]. Chromosomal aberrations of water treated with disinfectant and its byproduct have been studied with reference to nitro- and carbonyl groups. The contribution of the carbonyl group to activity inducing abbreviation is larger. In case of chlorinated water organochlorine compounds contribute to activity inducing aberrations in addition to the carbonyl group.

Epidemiology studies have been suggested that bladder, rectal and colon cancer are potentially associated with the drinking chlorinated water [62]. The evaluation of genetic alterations in rodent tumors may help to provide better understanding of molecular mechanisms underlying chemical carcinogenesis and consequently it is important for the accurate extrapolation of the cancer models to humans. Small fish models are considered short-term models that can be used to provide comparative toxicity information for the members of each family of chemicals and chemical mixtures [63]. Small fish models have previously been used to examine chlorination byproducts. THMs may cause colon cancer in humans and bromodichloromethane causes a high incidence of colon cancer in rats [60].

7. Conclusion

It is recognized that chlorination is not the ideal final disinfection process prior to water distribution. However, most of its drawbacks can be alleviated by a better knowledge of the reactions leading to the formation of DBPs and the application of more precise methods to estimate toxicity potential of these byproducts. This becomes a crucial issue when considering present health problems related to improperly disinfected or not disinfected waters that are distributed in sensitive areas. In this case, chlorination might be the only reliable enough disinfection process, provided that it is applied in conditions that minimize the impact of DBPs. In this regard, toxicodynamics may reveal itself a very useful tool for the investigation of chlorination and the associated DBPs.

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