สารอันตรายที่เกิดจากกระบวนการฆ่าเชื้อโรคในน้ำดื่ม Harmful Disinfection By-products in Drinking Water

ทิพวรรณ นิ่งน้อย Tipawan Ningnoi

กองอาหาร กรมวิทยาศาสตร์การแพทย์ ถนนติวานนท์ นนทบุรี 11000 Division of Food
Department of Medical Sciences
Tiwanond Road, Nonthaburi 11000

Interactions between water and human health are complex. Humans use water for many purposes, of which drinking is the most important. We need water in sufficient quantities and of good quality. Human health may be affected by the ingestion of contaminated water, either direct or through food, and by the use of contaminated water for purposes of personal hygiene and recreation. For drinking water, the most important requirement is that it is free from organisms and chemical substances that might be injurious to health. Therefore, treatment of the water supply is necessary to produce adequate water on a continuous basis thus protecting and promoting good health and well being. Terminal disinfection is essential to destroy microbial pathogens and to maintain the microbiological safety of drinking water.

Disinfection involves the use of reactive chemical agents such as chlorine

(Cl) and ozone (O). The use of these disinfectants usually results in the formation of chemical by-products, some of which are potentially hazardous and are included in the WHO Guidelines for Drinking-Water Quality. These guideline values are based on assumptions and scientific principles for the assessment of risks to human health. Chloroform (CHCl), the most common occurring trihalomethane (THM) generated during chlorination and bromate (BrO₃⁻), a possibly carcinogenic by-product of ozonation, were included in the Guidelines in 1993 (WHO, 1993). The health risks due to toxic chemicals in drinking water differ substantially from those caused by microbial agents. The problems associated with chemical constituents of drinking water arise predominantly from their abilities to cause adverse health effects after prolonged period of exposure.

Many researches and studies have

been conducted to find the appropriate techniques to remove or control the byproducts in drinking water (Siddiqui and Amy, 1994; Song et al., 1997; Symons and Zheng, 1997; Viessman and Hammer, 1994; Westerhoff et al. 1998 and Thomson et al., 1997). These include the use of ultraviolet (UV) irradiation, control of treatment variables, removal of by-product precursors and use of alternative disinfectants etc. In Thailand where chlorine is used for disinfection purpose in the water supply system, the potential hazard of its by-products especially THMs has been realized. Efforts have been put in order to get rid of this problem and to find a better alternative to chlorination. Ozonation has been mentioned as a possible alternative. But no effective measures have yet been employed in practice. Therefore, this report aims at reviewing the occurrence of chlorination and ozonation by-products in drinking water, their effects on human health and the processes that can control or remove them. The area of this paper will focus mainly on THMs and bromate in drinking water produced from surface water.

1. Treatment of Drinking Water

The basic role of water treatment is to provide sufficient quantities of water having the appropriate quality for a variety of uses. Drinking water is often expected to be wholesome or palatable. It is essential that drinking water should not give rise to ill health and should be pleasing to the drinker which means it must be free of colour, taste, odour and excessive amounts of mineral and organic matters. Only few raw waters especially from surface water sources meet these requirements. By applying various purification or treatment techniques, it is possible to produce drinking water that meets such requirements.

1.1 General treatment

Water treatment in general involves physical and chemical processes. In the European Communities the treatment includes coagulation, flocculation, sedimentation, filtration, refining and disinfection with chlorine dioxide or ozone (Masschelein, 1992; Anonymous, 1994). In the United States the standard treatment is based on the permissible and desirable criteria. This involves coagulation with the eventual addition of coagulation aids and active carbon, sedimentation, rapid sand filtration and disinfection with chlorine. The treatment is, however, adapted to the particular water under consideration. The potential formation of undesirable or toxic compounds will determine changes in the treatment criteria. In addition, the continuous contamination of sources of drinking-water necessitates new purification techniques, for example techniques for the removal of nitrates, membrane filtration and advanced oxidation techniques to

remove organic micropollutants such as pesticides and humic acids (Masschelein, 1992). In Bangkok, Thailand, the Metropolitan Waterworks Authority uses the treatment processes as natural sedimentation, coagulation, antracite and sand filtration and disinfection with chlorine (Anonymous, 1997).

1.2 Disinfection

Disinfection is essential to ensure that the water supply is microbiologically safe. Disinfection methods may be either physical or chemical. Physical methods include boiling and UV irradiation; chemical methods include the addition of ozone, or most commonly, chlorine or its derivatives. Here only chemical methods will be described and discussed in detail.

1.2.1 Chlorination

Chlorine is the most commonly used chemical for disinfection of water and wastewater. In the United States, liquid chlorine is applied in most water treatment plants (Viessman and Hammer, 1992). In general, chlorine, sodium hypochlorite and bleaching powder are the most important disinfectants applied in water treatment. In water, chlorine is dissociated into hypochlorous acid which has a strong disinfection power. It reacts rapidly with organic and inorganic matter present in water. When it passes the cell wall of microorganisms present in water, it oxidizes the enzymes which are the essential biological catalysts. If adequate disinfection is to be achieved, due allowance must be made for the chlorine consumed in these reactions in addition to that needed for infection. The amount of chlorine required to react with other compounds is termed as the chlorine demand of the water. Thus, the chlorine dose must be sufficient both to satisfy the chlorine demand and to produce unreacted excess known as the residual. A minimum free residual of 0.5 mg/litre is recommended, together with a minimum contact time of 30 minutes and a water turbidity of less than 5 Naphelometric Turbidity Unit (NTU). The chlorine demand of some waters (mainly river water) can increase dramatically at times of heavy pollution, particularly after rain (WHO, 1998a).

The disadvantage of chlorination is that chlorine reacts with other compounds (mainly ammonia, some metal ions, and organic compounds) to form undesirable products. Since Rook and Bellar in 1974 found THMs as chlorination by-products (Bean, 1983), chlorination has become less accepted and studies have been carried out to seek alternative disinfectants.

1.2.2 Ozonation

Ozonation has been increasingly used as an alternative for chlorination in the water treatment plants with the primary reasons as follows (Rapinat, 1982; van der Veer *et al.*, 1994):

Formation of chlorinated byproducts (for example, THMs)

Ineffectiveness of chlorination against zooplankton

Potential increase in mutagenicity of chlorinated water

Taste and odour control

Effectiveness of ozone against viruses and other pathogens (e.g. Giardia)

Pesticide oxidation capabilities of ozone

At the South Holland South Waterworks, (Rotterdam, the Netherlands), an ozonation system has been developed to replace chlorination in the treatment process as well as to remove pesticides and taste and odour compounds (van der Veer *et al.* 1994). In Japan, ozonation is widely employed to remove musty smells in tap water purification (Utsumi *et al.*, 1994).

Use of ozone for the disinfection of drinking water began in 1906 in Nice (France) to eliminate pathogenic microorganisms and indicators of faecal pollution (Escherichia coli. and Streptococcus faecalis) and to reduce pathogenic microorganic population (Masschelein, 1992). Ozone is more active than chlorine. Unlike chlorine, it does not react with water to produce disinfecting species but decomposes in water to produce oxygen and hydroxyl free radicals. The reactions are rapid in inactivating microorganisms. They affect the constituents of cytoplasmic membranes,

enzymatic systems and even nucleic acid; for virus, ozone attacks the proteins of the capside to some extent as the nucleic acid (Block, 1982). The effect is very fast and no resistant water borne strains are known. The residual ozone concentration level in water of 0.4 mg/L for 4 to 6 minutes can effectively deactivate polioviruses. The viral particles are quickly removed (30 seconds) with a very low level of residual ozone (0.05 mg/L). In general a maximum ozone dose of 4 mg/L with a contact time of 10 minutes should be adequate for disinfection for bacteria, viruses, Giardia and Cryptosporidium. The inactivation of Giardia cysts is a criterion for appropriate disinfection purposes as a guideline by USEPA (Masschelein, 1992).

The efficiency of the disinfection depends on various factors such as pH, temperature, organic matter, suspended solids and residual ozone. The presence of residual ozone is generally considered a key parameter in the completion of water disinfection.

However the application of ozonation has some disadvantages. Two main concerns are as follows:

High cost of installation and operation. The application of ozone in the treatment plant is expensive compared with more traditional treatment with chlorination. Since half-life of ozone in water is very short (approximately 10–30 minutes),

it must be generated on site. Thus, ozone generation plant is required which costs a lot of money to install and to operate. In general, an ozonation system consists of an air preparation or oxygen feed, electric power supply, ozone generation, ozone contacting and ozone contactor exhaust gas destruction (Viessman and Hammer, 1992). Moreover, application of ozone requires considerable research and development work and pilot plant studies. If ozonation is used for different purposes a proper evaluation must be made for each purpose (Masschelein, 1992; Holmes *et al.*, 1994).

Unacceptable by-products. Although ozone does not produce many unacceptable by-products, two compounds have been of particular concern: bromates and assimilable organic carbon (AOC). Bromate has been a concern due to its stability and the relatively high carcinogenic risk. Ozone increases the level of biodegradable organic AOC, which has the potential to support bacteriological growth with subsequent problems in the distribution system. Therefore AOC has to be reduced by an additional biological filtration step or by adsorption on activated carbon (Holmes et al., 1994).

2. Disinfection By-products and Effects on Human Health

Twenty two disinfection by-products (DBPs) have been included in the WHO Guidelines for Drinking-Water (WHO, 1993). The guideline values (GV) are established based on assumptions and scientific principles for the assessment of risk to human health. The GV for THMs and bromate are shown in Table 1.

2.1 Trihalomethanes

THMs are hydrogen-substituted single-carbon compounds namely chloroform (CHCl₂), bromodichloromethane (CHBrCl₂), dibromochloromethane (CHBr Cl) and bromoform (CHBr). THMs occur principally as by-products of chlorination of water and accounted for about 50% of the total by-products on the weight basis. The total concentration of these four THMs in drinking-water may vary up to 1000 µg/L, but it is frequently less than 100 μg/L. Among these compounds, chloroform is the most commonly encountered and available information pertains almost exclusively to this substance (WHO, 1984; Viessman and Hammer, 1992).

The levels of chloroform in drinking-water reported in some countries (WHO, 1998b; Ningnoi *et al.*, 1998) are presented in Table 2.

Table 1 WHO Guideline values for THMs and bromate in drinking water

Disinfectant by-products	GV, μg/L	Remarks
Bromate Trihalomethanes	25* (P)	For 7x10 ⁻⁵ excess risk* The sum of the ratio of the concentration of each to their respective GV should not exceed 1
* Bromoform * Dibromochloromethane * Bromodichloromethane Chloroform	100 100 60* 200*	For 10 ⁻⁵ excess risk* For 10 ⁻⁵ excess risk*

(P) Provisional guideline value

Table 2 The levels of chloroform in drinking-water as reported in some countries

Country	Chloroform in $\mu g/L$ (mean value)	Year of study
Canada (53 sites)	<50	1993
Germany (100 cities)	1.3	1977
The Netherlands	up to 8.9	1994
USA (35 sources)	38.9	1988-1989
Thailand (Bangkok)	49.4	1996-1997

^{*} For substances that are considered to be carcinogenic, the GV is the concentration in drinking water associated with an excess lifetime cancer risk of 10–5 (one additional cancer per 100,000 of the population ingesting drinking water containing the substance at the GV for 70 years).

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2.1.1 Trihalomethane formation

In 1974 reports by Rook and Bellar show that chlorination of natural water results in formation of volatile halogenated compounds giving impetus to studies directed toward understanding the reactions of aqueous chlorine with the organic material waters (Bean, 1983). For THMs, fundamentally, the formation of THMs results from the substitution by halogens of the mobile hydrogen atoms carried by a carbon atom, followed by the cleavage of the molecule (Figure 1). Although many factors such as temperature, time, pH and concentrations of bromide and residual chlorine influence formation of THMs, the major variable is the level of humic acids in water.

2.1.2 Effects on human health

Discussion of health effects of THMs deals mainly with chloroform which has received the most intensive study because of its former use as inhalation anaesthetic and because it is the predomi-

nant THM in drinking water. The toxic effect of the other THMs are likely to be similar to those of chloroform (WHO, 1984).

Chloroform is a central nervous system depressant. It also affects liver and kidney functions. The immediate effect of chloroform intoxication is loss of consciousness, which may be followed by coma and death as a result of respiratory failure and cardiac arrhythmia. damage is noted 24-28 hours after exposure and hepatic injury is seen after 2-5 days; thus symptoms of poisoning may occur several days after recovery from chloroform anaesthesia. The application of chloroform as an anaesthesia have been prohibited in many countries (WHO, 1998b). The mean lethal oral dose for an adult is estimated to be about 45 g, but large inter-individual differences in susceptibility occur (WHO, 1998b)

The long-term effects of THMs on human are considered based on the results of laboratory animal studies. It was found

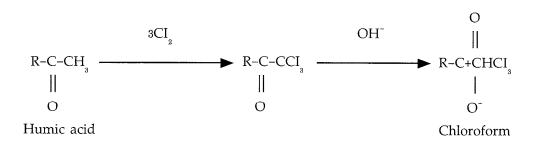


Figure 1 Formation of chloroform during chlorination

from a wide range of studies that chloroform can induce carcinogenicity in rodents. Hepatic tumours and liver tumours were observed in mice fed with gavage in corn oil. Chloroform also induces renal tumours in rats but at lower rates than liver tumours in mice.

Analytical epidemiological studies of potential associations between ingestion of chlorinated drinking-water and colorectal cancer have been conducted in the USA in Wisconsin, New York and Iowa. Similar investigations of bladder cancer have been conducted in Colorado, Iowa and Ontario, Canada. No evidence is available to suggest an increased risk of cancer in both cases (WHO, 1998b).

2.2 Bromate

Bromate ion exists in a number of salts, the most common of which are potassium and sodium bromate. It is used in home wave neutralizing solutions. Small amounts may be added to flour as a maturing agent, to dough as a conditioner, and to fish paste. It may also be added to beer or cheese. Bromate does not volatilize. It is not normally present in water but may be formed from bromide during ozonation. Bromate concentrations of 60–90 µg/L have been reported in ozonated water. For most people, exposure to bromate is unlikely to be significant. If ozone is used to disinfect drinking-water, intake of bromate might range from 120-180 µg/day (WHO, 1996).

2.2.1 Bromate formation

During the ozonation of water, many organic and inorganic disinfection by-products are formed from ozone reactions with compounds dissolved in water. Since some bromide is present in all natural waters, ozonation may yield the formation of bromate, a compound that is possibly carcinogenic to humans and difficult to remove. More than 5 µg/L of bromate may be formed when applying ozone dosage higher than 0.7 mg O₃/L, which result in ozone exposures higher than 5 mg/L min (Orlandini *et al.*, 1994).

Bromate formation depends on ozone residual, bromide concentration, temperature, the contact time, pH and other factors such as the concentration of ammonia and ozone scavengers (i.e., natural organic matter (NOM), bicarbonate/carbonate), and the presence of compounds causing enhanced ozone decomposition to hydroxyl radical (Holmes et al., 1994; Orlandini et al, 1994 and Song et al., 1997). Ammonia delays and may decrease bromate formation as a result of its reaction with HOBr. The presence of NOM and other compounds exerting an ozone demand reduces the amount of ozone available for the reaction with bromide (Orlandini et al., 1994). Furthermore, formation of brominated organic compounds may serve as an additional sink for bromide, reducing its availability for bromate formation.

The mechanism of bromate formation during the ozonation of water containing bromide (Br⁻) can be described in three major pathways (Hoigne, 1982; von Gunten *et al.*, 1996 and Song *et al.*, 1997).

- The direct ozonation pathway. Ozone sequentially oxidises bromide to aqueous bromine (HOBr/OBr⁻) and then bromate. Because OBr⁻ rather than HOBr can be oxidised to bromite (BrO2⁻) at an appreciable rate, bromate formation is favoured at high pH upon continuous ozonation.

- The direct-indirect bromide/ ozonation combination pathway. In this pathway, ozone oxidises bromide to aqueous bromine: followed by hydroxyl radical (HO°) oxidation of aqueous bromine to BrO° which disproportionates to bromite. Bromite is then oxidised by ozone to form bromate.

- The indirect-direct ozonation combination in which bromide is first oxidised by HO° to form Br°, which is further oxidised by O₃ to form BrO°. This continues to form bromate through disproportionation to bromite followed by ozone oxidation to bromate.

This whole process of bromate formation in different pathways is illustrated in Figure 2.

When conventional ozonation is replaced by a combination of ozone and hydrogen peroxide, bromate formation increases if the ozone residual is kept constant but decreases if the ozone dose is kept constant (von Gunten *et al.*, 1996). However a study carried out by Symons and Zheng (1997) showed that when the hydrogen peroxide–UV–visible process was used, bromate formation did not occur. The reduction of any HOBr formed by the excess hydrogen peroxide might be the reason to prevent bromate formation.

2.2.2 Effects on human health

Most cases of human poisoning from bromate are due to the accidental or intentional ingestion of home permanent wave solutions, which normally contain 2–10% bromate. Toxic effects of bromate salts include nausea, vomiting, abdominal pain and diarrhoea, varying degrees of central nervous system depression, seizures, respiratory depression, and pulmonary oedema, most of which are reversible. Irreversible effects include renal failure and deafness, both of which have been observed following the ingestion of 240–500 mg of potassium bromate per kg of body weight (WHO, 1996).

From the results of many researches, WHO has concluded that there is sufficient evidence for the carcinogenicity of potassium bromate in animals and has classified it as a possible human carcinogen. Bromate is mutagenic both in vitro and in vivo.

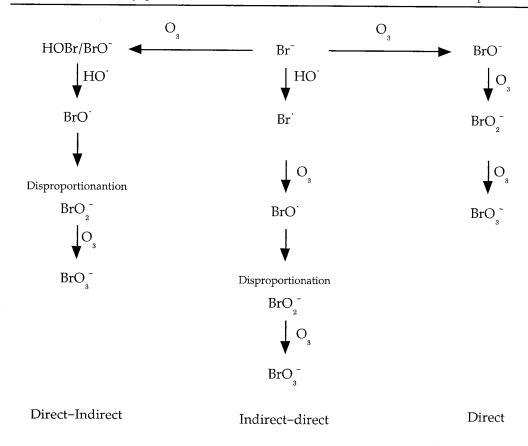


Figure 2 Major bromate formation pathways

3. Removal of Disinfection By-products 3.1 Removal of trihalomethanes

Many studies have been done to find appropriate methods for the reduction or control of THM formation. Brief description of the methods found in the relevant literatures is given below.

3.1.1 Change of the point of chlorination

The easiest method for reducing THM formation is to move the point of chlorine application to later stages in water treatment. If previous control of microbial populations is not necessary, chlorine should not be added until after filtration. The benefits of delayed chlorination are reduction in the required dosage and prior removal of THMs precursors (Viessman and Hammer, 1992).

3.1.2 Removal of THMs precursors prior to chlorination

Removal of THMs precursors can be done during the appropriate treatment process such as enhanced coagulation, activated carbon and enhanced softening. Viessman and Hammer (1992) found that optimising chemical coagulation has the benefits of reducing and removing the precursors. Further study carried out by Vrijenhoek et al., (1998) showed that THM precursors were removed significantly by enhanced coagulation at pH 5.5. At this pH, humic substances are likely to be removed by formation of insoluble aluminumhumate complexes at a low alum dosage and adsorption to aluminum hydroxide precipitates at a high alum dosage. Powdered activated carbon and granular activated carbon applied in the early stages of treatment are found effective for adsorption of humic substances (Viessman and Hammer, 1992; Krupa and Cannon, 1996). Lime-soda ash softening used to remove calcium and magnesium (cause of hardness of water) can remove NOM including humic acids (Thomson et al., 1997).

3.1.3 Use of an alternative disinfectant

The alternative disinfectants can be used to avoid THM formation. These include ozone, chlorine dioxide and chloramines. The study done at Anglican Water, UK (Croll, 1994) showed the level of THM concentrations with effective preoxidants as in Table 3.

Ozone

Ozone is a very effective alternative for chlorine regarding the purpose of removal of THMs. However, if ozone is used as the primary disinfectant, secondary chlorination may be needed to provide a disinfectant residue in the water entering the distribution system. The major disadvantages of ozone are high cost and bromate formation which is harmful to health (Viessman and Hammer, 1992; Croll, 1994).

Chlorine dioxide

The problem associated with the

Table 3 Laboratory THM concentrations with effective pre-Oxidants

Pre-oxidant	THM formation potential $(\mu g/L)$	
	Range	Mean
Free chlorine	52-165	101
Monochloramine	52-104	75
Chlorine dioxide	36-97	55
Ozone	24-75	41

formation of THMs and other organochlorine compounds cast a new light on the potential advantages of the use of chlorine dioxide to disinfect drinking water. It is widely used in several cities in Europe such as Brussels, Zurich, Berlin, Dusseldorf, Monaco, Vienna etc. In the United States it is used in Ohio, Texas, Indiana etc. (Masschelein, 1992). Disinfection using chlorine dioxide has several advantages. It is a strong bactericide and viricide over a wide pH range and forms a residual capable of persisting in the distribution system. Chlorine dioxide does not react with humic acids to form THMs, however the greatest potential disadvantage is that it produces chlorate and chlorite ions which are toxic (Viessman and Hammer, 1992; Hurst and Knocke, 1997).

Monochloramine

One of the least expensive methods of limiting chlorination by-products including THMs formation is to use monochloramine instead of free chlorine to maintain a distribution system residual. Monochloramine limits the free chlorine contact time which is the most important variable affecting on THM formation (Croll, 1994; Carlson and Hardy, 1998). However, chloramines are weak disinfectants and, except for establishing a residual to control bacteria, are of little benefit in inactivating viruses and protozoal cysts (Viessman and Hammer, 1992).

3.2 Removal of bromate

Since bromate was classified as a human carcinogen by WHO in 1990, many researches have been carried out to obtain the techniques that can remove or control bromate production in drinking water. Westerhoff et al., (1998) found that the presence of (NOM) in water reduced the bromate formation with two reasons: (1) NOM reacts directly with ozone and hydroxyl radicals to reduce the amount of oxidant available for bromide and bromine oxidation respectively and (2) NOM reacts with bromine, the main intermediate in the formation of DBPs. The methods for controlling the bromate formation are as follows:

3.2.1 Control of pH

A study carried out by Song et al., (1997) showed that acid addition was a very effective technique to minimise bromate formation since bromate formation is favoured at high pH. This finding is in agreement with the study done by Holmes et al., (1994) which showed that reducing the pH would both reduce bromate formation and improve disinfection due to decrease in ozone decay rate. This method is useful for water with low alkalinity and water subjected to acid addition prior to enhanced coagulation treatment.

3.2.2 Ultraviolet- irradiation

UV-irradiation can be used to reduce bromate in drinking water. Siddiqui and Amy (1994) found that a medium

pressure mercury lamp with intensity of 1 watt/cm2 is very effective in destroying bromate with a concentration of 50 μ g/L when the contact time is about 30 seconds. The similar finding was obtained in the study done by Symons and Zheng (1997) which showed that when hydrogen peroxide and UV-visible irradiation was used, the bromate formation did not occur. It was suggested that reduction of any HOBr formed by the excess of hydrogen peroxide may have prevented the formation.

However, controlling bromate formation may, depending on the water quality and water treatment variables, increase the formation of total organic bromine, of which some compounds may pose health risks (Song *et al.*, 1997). Therefore, case studies are necessary before employing any control method.

RECOMMENDATIONS

Although many techniques to get rid of potential problems of by-products are possible to be implemented, other factors should be taken into consideration such as technical and financial aspects. In Thailand, people are concerned about the health risks of THMs and think of ozone as a better alternative. From this report it is shown that ozone can be a good alternative to chlorine for disinfection considering THM formation. However due to very high cost of installation and operation, use of ozone

noticeably increases the cost of drinking water which may not be affordable to the public. In addition, ozonation produces bromate which is also carcinogenic. Therefore, the removal of THM precursors is likely to be the most suitable technique. This technique can be implemented with low cost. By making use of the current treatment processes such as coagulation, filtration and softening etc, with some adjustment, THMs can be significantly reduced.

SUMMARY

Use of chlorine or ozone for disinfection in drinking water treatment usually results in the formation of chemical by-Particularly, the by-products products. trihalomethanes (THMs) from chlorination and bromate from ozonation are possibly human carcinogens. In this paper, studies that have been conducted to reduce these by-products and to seek alternative disinfectant were reviewed. Several approaches are found that can be applied in practice. Enhanced coagulation, use of activated carbon and enhanced softening help reduce the precursors of THMs. Using ozone with controlled pH substantially reduces the formation of bromate and ultraviolet irradiation can also be used to reduce bromate. However, it is found that each technique has both advantages and disadvantages. In conclusion, disinfection by-products in

drinking water can be reduced to some extent but cannot be totally avoided. The long-term effects of these products should not be compromised with the destruction of pathogenic microorganisms that have immediate adverse health effects.

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