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Genotoxicity of drinking water treated with different disinfectants and effects of disinfection conditions detected by *umu*-test

Xuebiao Nie, Wenjun Liu, Liping Zhang, Qing Liu*

Division of Drinking Water Safety, School of Environment, Tsinghua University, Beijing 100084, China. E-mail: 363120244@qq.com

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ABSTRACT

The genotoxicity of drinking water treated with 6 disinfection methods and the effects of disinfection conditions were investigated using the *umu*-test. The pretreatment procedure of samples for the *umu*-test was optimized for drinking water analysis. The results of the *umu*-test were in good correlation with those of the *Ames*-test. The genotoxicity and production of haloacetic acids (HAAs) were the highest for chlorinated samples. UV + chloramination is the safest disinfection method from the aspects of genotoxicity, HAA production and inactivation effects. For chloramination, the effects of the mass ratio of Cl_2 to N of chloramine on genotoxicity were also studied. The changes of genotoxicity were different from those of HAA production, which implied that HAA production cannot represent the genotoxic potential of water. The genotoxicity per chlorine decay of chlorination and chloramination had similar trends, indicating that the reaction of organic matters and chlorine made a great contribution to the genotoxicity. The results of this study are of engineering significance for optimizing the operation of waterworks.

Introduction

Disinfection is an important final step in the treatment process of drinking water in China and elsewhere. For chlorination, the most common disinfection process for drinking water, many researchers have focused on the reactions of chlorine with natural organic matters (NOMs) that generate typical disinfection byproducts (DBPs), such as trihalomethanes (THMs) and HAAs. The constituents of NOMs in surface water are complex, including humic substances, soluble microbial products and detritus of plants and animals. The dissolved and colloidal forms, referred to as dissolved organic matters (DOMs), are the most challenging and detrimental fractions of NOMs for water treatment and supply. Many researchers have reported that chemical disinfectants such as chlorine react with DOMs to produce numerous DBPs with genotoxic, mutagenic and/or carcinogenic activity (Glaze et al., 1993; Koivusalo and Vartiainen, 1997; King and Marrett, 1996; Koivusalo et al., 1997; Betts, 1998). At the same time, some of the DOMs are toxic. Therefore, only measuring the formation of typical DBPs is not sufficient to investigate the effects of disinfection on the chemical safety of drinking water, and it is imperative to detect comprehensive toxicity indexes such as genotoxicity.

Genotoxicity of drinking water has been investigated by a large number of researchers in many countries. Shen et al. (2003) detected the mutagenic potential in chlorinated tap water in Shanghai, and found that boiled water had stronger mutagenic potential. Li et al. (2006) investigated samples from four locations along the Yangtze River in Nanjing and found that the organic concentrates were genotoxic. Bolognesi et al.

* *Corresponding author*. E-mail: wjliu@tsinghua.edu.cn (Wenjun Liu).

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(2004) found that surface water in Italy treated by chlorine or chlorine dioxide indicated DNA/by-product interaction, while peracetic acid was a safe disinfectant. The genotoxicity of water in swimming pools has attracted more attention due to the long and continuous exposure to disinfectants. Disinfected recreational pool water samples induced more genomic DNA damage than the source tap water (Liviac et al., 2010). There was higher mutagenicity and higher DBP content in freshwater pools than seawater pools, and HAAs were found to be the most prevalent chemical class (Manasfi et al., 2016).

For chlorination, chlorinated DBPs play the most important role in the genotoxicity of drinking water. In order to reduce DBP production, modified chlorination has been investigated, such as sequential chlorination and chlorination plus chloramination (Zhang et al., 2006; Liu et al., 2009). Chloramination, which is considered to be safer than chlorination in drinking water treatment with less production of DBPs (Richardson et al., 2007; Lu et al., 2009), has attracted more attention. However, Wang et al. (2005, 2007a, 2007b) found that ammonia nitrogen could increase the genotoxicity of wastewater during chlorination, which means chloramine disinfection may have great potential to increase the genotoxicity of wastewater. If the same conclusion can be drawn for drinking water treatment, the advantages of chloramination will be challenged.

In recent years, ultraviolet (UV) disinfection, as an alternative disinfection method, is becoming more widespread. The benefits of UV disinfection include reduced risk of microbial pathogens such as *Cryptosporidium* and minimal production of regulated DBPs (EPA, 2006). Studies have shown that, at practical UV disinfection doses applied to drinking water (40 mJ/cm^2), the chlorine demand and resulting DBPs do not appreciably change (Malley et al., 1996; Kashinkunti et al., 2004). However, UV has no persistent impact in the pipelines, so application of UV in water supply works is usually combined with chlorine or chloramine. Recent research found that medium-pressure (MP) UV irradiation of nitrate-containing drinking waters followed by chlorination enhanced the levels of chloropicrin and halonitromethane, especially when followed by chloramination (Reckhow et al., 2010; Shah et al., 2011). The genotoxicity of drinking water treated by medium-pressure ultraviolet (MP-UV) and chlorine is lower than that treated by MP-UV and chloramine (Plewa et al., 2008). For reclaimed water disinfection, Wang et al. (2011) found that sequential disinfection of low-pressure ultraviolet (LP-UV) (8 mJ/cm^2) and chlorine (1.5 mg/L) led to less genotoxicity than chlorination alone. From the aspect of chemical safety, low-pressure (LP) UV has advantages over MP-UV.

Most newly built large waterworks in China have applied the approach of UV combined with the traditional chlorination/chloramination, while many existing waterworks have ameliorated the disinfection process by adding UV treatment. The addition of UV brings significant advantages into the disinfection process. If the chlorination needs to be changed to chemically safer chloramination, UV treatment can ensure the inactivation efficiency for pathogenic microorganisms; while for chlorination, with the addition of UV treatment, it is possible to reduce DBP production by lowering the chlorine dose.

As mentioned above, HAAs and THMs are the two most common DBPs in chlorinated drinking water. In addition, there are many other regulated DBPs that have been found to have higher genotoxic risk, such as haloacetamides (Plewa et al., 2008), halonitromethane (Liviac et al., 2009) and N-nitrosamines (Liviac et al., 2011; Wagner et al., 2012). In this study, as the source water contained little ammonia, there were almost no N-DBPs formed during the treatment. Between HAAs and THMs, HAAs were reported by some researchers to be the most abundant DBPs and the major source of carcinogenic risk in chlorinated drinking water (Nieminski et al., 1993; Label et al., 1997; Zhang and Li, 2000). Therefore, this study focused on the investigation of HAAs, and a sample concentration method that focuses on nonvolatile substances was used (HAAs are nonvolatile substance while THMs are volatile).

In this study, the genotoxicity of drinking water samples from a waterworks in North China was assessed with the *umu*-test. Testing results of water samples treated by chlorination alone, chloramination alone, LP-UV alone, and their combinations were compared and evaluated. HAA production was also analyzed. For chloramination cases, the influence of the mass ratio of Cl_2 to N was investigated. The results of this study are of significance for optimizing the operation of waterworks in China, especially waterworks applying UV combined with chlorination/chloramination as the disinfection process.

1. Materials and methods

1.1. *Sample collection and preparation*

The water samples investigated in this study were collected from a surface water source in Beijing and the effluents of rapid sand filtration (RSF) and ozone-biological activated carbon (O_3-BAC) processes from a waterworks that used this source water. The water samples were immediately delivered to the laboratory after filtering through glass-fiber membranes (0.45 μm, Millipore, USA) to eliminate suspended solids before other water quality parameters were analyzed. The characteristics of the water samples are shown in Table 1.

Chlorine solution was prepared from sodium hypochlorite solution (about 30% (*w*/*w*) as Cl_2) and de-ionized water. Chloramine solution was prepared by mixing sodium hypochlorite and ammonium sulfate solutions (the mass ratio of Cl_2:N was 4:1) with continuous stirring for 10 min in an ice bath. The initial pH of the solution was 8.0. The concentration of available chlorine was measured by a Pocket Chlorine Colorimeter (PCII, Hach, USA). All of the chemical reagents used were of analytical grade. The concentrations of available

Table 1 – Characteristics of the water samples.

Water type	DOC (mg/L)	UV_{254} (cm^{-1})	NH_4^+-N (mg/L)	Total HAAs (μg/L)	pH
Surface source water	3.45	0.051	0.04	0.44	8.1–8.3
Effluent of RSF	0.83	0.024	<0.01	6.32	7.7–8.1
Effluent of O_3-BAC	0.62	0.015	<0.01	3.31	7.5–8.0

DOC: dissolved organic carbon; UV: ultraviolet; HAAs: haloacetic acids; RSF: rapid sand filtration.