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A comparison of genotoxicity change in reclaimed wastewater from different disinfection processes

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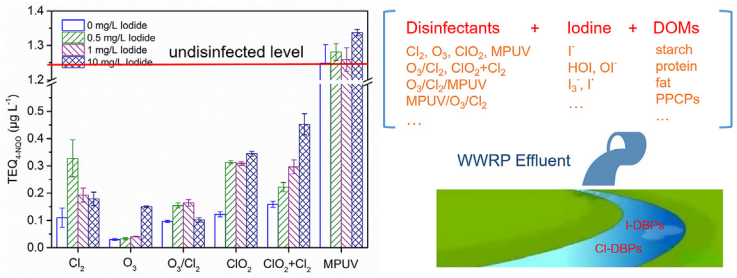
HIGHLIGHTS

- Chemical disinfection is effective for genotoxicity removal of reclaimed wastewater.
- MPUV increased genotoxicity, other disinfectants reduced it in varying degrees.
- Combination of MPUV, ozone and chlorine reduced genotoxicity to the greatest extent.
- Ozonation effectively controlled genotoxicity by limiting I-DBP formation.

GRAPHICAL ABSTRACT



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ABSTRACT

Effluents before disinfection from four wastewater reclamation plants were treated with chlorine (Cl_2), ozone (O_3), chlorine dioxide (ClO_2), medium-pressure ultraviolet (MPUV) and four different combinations of the above, to evaluate the effect of disinfection processes on the genotoxicity removal by the SOS/umu test. Results showed that the genotoxicity increased after MPUV irradiation (10–100 mJ/cm^2), but declined when adopting other disinfection processes. The effectiveness of genotoxicity reduction by five chemical disinfectants was identified as: O_3 > pre-ozonation with $Cl_2 \approx ClO_2$ > combination of ClO_2 and Cl_2 > Cl_2. The sequential combination of MPUV, Cl_2 and O_3 reduced the genotoxicity to a level similar to the source water. The influence of differential disinfection process varied on iodinated wastewater, which is closely related to the competitive reactions between disinfectants, iodine and dissolved organic matters. The removal of genotoxic pollutants and the formation of genotoxic disinfection by-products are the two major factors that lead to the change in genotoxicity during disinfection.

1. Introduction

Water scarcity is becoming one of the most pressing issues in China. Reclamation and reuse of wastewater is necessary to reduce water pollution, replenish streams and extend the lifespan of water sources, helping water-scarce regions to meet their long-term water needs (Huertas et al., 2008). In Beijing, for example, reclaimed water has been utilized in industrial processes, for supplementing rivers and lakes, and for irrigation and residential purposes (Tang and Miao, 2014). However, reclaimed wastewater which contains harmful substances along with transformation byproducts via biological and chemical treatment processes poses

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risks to the aquatic ecosystem and human health (Plumlee et al., 2008; Duong and Saphores, 2015).

Toxicity is highly recommended as a general indicator to investigate potential ecological risks (Farre and Barcelo, 2003). In the past several decades, a series of methods have been developed based on bioassays to evaluate various classes of toxicity in environmental matrices, including acute toxicity, cytotoxicity, genotoxicity, estrogenic activity, etc. (Farre and Barcelo, 2003). Among them, the SOS/umu test is a genotoxicity assessment based on genetically engineered Salmonella typhimurium TA1535/pSK1002 strain (ISO, 2000). β-galactosidase induced by DNA damaging substances is used for the SOS/umu test to quantitate genotoxicity. It is a short-term and high-throughput detection method and has been extensively employed in evaluating the toxicity of environment samples (Oda, 2016).

Reclaimed water must be disinfected to eliminate pathogens before being recycled or discharged. Commonly applied disinfectants are chlorine, ozone, chlorine dioxide, ultraviolet and a combination of the above. Chlorine is the most utilized disinfectant, which is prone to react with dissolved organic matters (DOMs) to form halogenated disinfection by-products (DBPs), typically trihalomethanes (THMs) and haloacetic acids (HAAs) (Rebhun et al., 1997; Sun et al., 2009; Xie, 2016). However, regulated DBPs account for a fraction of total organic halogen (TOX) and the mole ratio of TOX to total organic carbon (TOC) is only approximately 0.4%–1.2% in chlorinated wastewater (Jekel and Roberts, 1980; Koch et al., 1991; Richardson et al., 2008b). Many emerging contaminants which are ubiquitous in wastewater (e.g. antibiotics, UV filters and dyes) have been found to have higher genotoxicity after chlorination (Bedoux et al., 2012; Vacchi et al., 2013; Chai et al., 2017). When considering the overall adverse effect, previous researches have already reported that chlorination could decrease the genotoxicity of secondary treated wastewater but the presence of a high ammonium concentration would inhibit the decline (Wang et al., 2007; Wu et al., 2010).

Ozone has a strong oxidation ability, allowing it to be used as a disinfectant to react with DOMs selectively by ozone molecule, and non-selectively through hydroxyl radicals generated from ozone decomposition (von Gunten, 2003). Ozone is often applied with chlorine due to the lack of continuous disinfection ability. Ozonated DBPs are typically small molecules of aldehydes, ketoacids and carboxylic acids (Xie, 2016). Among them, acetaldehyde and formaldehyde were found to cause metabolic gene mutation (Richardson et al., 2007). Studies have showed that the genotoxicity of wastewater effluents could be reduced to a negligible level after ozonation (Nakamuro et al., 1989; Cao et al., 2009). However, few have discussed whether subsequent chlorination would alter the genotoxicity.

Chlorine dioxide is another effective alternative, with about 68% and 9% of it decomposing into chlorite and chlorate, respectively (Korn et al., 2002). It has been shown that chlorine dioxide can produce a considerable amount of unknown TOX during drinking water treatment (Werdehoff and Singer, 1987; Zhang et al., 1999; Hua and Reckhow, 2007). Research on the genotoxicity change of model compounds or actual waters by chlorine dioxide disinfection is limited. The Ames test showed that wastewater treated with 1.5 mg/L chlorine dioxide exhibited mutagenicity (Monarca et al., 2000). Genotoxicity tests using human HepG2 cells revealed that chlorate led to an increase of DNA damage while chlorite did not, and no increase in micronuclei frequency was caused by chlorate and chlorite (Feretti et al., 2008).

UV irradiation alone does not produce halogenated DBPs, but can cause photodegradation or photorearrangement of some DOMs (Magnuson et al., 2002). A range of contaminants have been observed with increasing genotoxicity under UV irradiation by the umu test (Watanabe-Akanuma et al., 2005; Takamura-Enya et al., 2011). The toxicity change is also associated with implementing polychromatic (between 200 nm and 300 nm) medium pressure UV (MPUV) and monochromatic (254 nm) low pressure UV (LPUV). Plewa et al. reported that with a combination of UV and chlorine, the mammalian cell genotoxicity of drinking water was observed lower with MPUV than with LPUV (Plewa et al., 2012). However, it is proved that MPUV presents greater potential to photodegrade DOMs, whose maximum absorption occurs within 200 nm–300 nm (Lekkerkerker-Teunissen et al., 2012). In addition, MPUV has a wider application in large-scale wastewater reclamation plants, due to its smaller footprint (Lekkerkerker-Teunissen et al., 2013).

Iodine is an essential element for mammals and the WHO recommends a daily intake level of 150 μg for adults (WHO, 2007). As a result, iodine is in various physiological fluids and ultimately enters into domestic sewage. Iodine also exists in iodinated X-ray contrast media and the productions of chemicals, iodophor, herbicides, pharmaceuticals and nylon etc., which end up in different wastewaters (Moreda-Pineiro et al., 2011). In addition, seawater intrusion is another cause of higher iodine content in coastal waters (Wong, 1991). Iodide (I^-), iodate (IO_3^-) and organo-iodine are the three main forms of iodine in water (Hansen et al., 2011). They may undergo transformation into one another in the ambient environment and during wastewater treatments (Gong and Zhang, 2013). Iodide, as an important DBP precursor, can participate in oxidation, substitution and other reactions during disinfection (Hua et al., 2006). In the US and Canada, up to 104 μg/L iodide has been reported in some source waters (Richardson et al., 2008a). Hong Kong's saline wastewater effluents contained iodide ranging 30–60 μg/L (Gong and Zhang, 2013).

Iodide can be oxidized by chemical disinfectants into hypoiodous acid (HOI), which then reacts with DOMs to form iodoorganic by-products or is further oxidized to iodate by ozone or chlorine (Bichsel and von Gunten, 1999). Generally, chlorine dioxide can only oxidize iodide into iodine radicals ($I^•$). $I^•$ has a longer half-life than HOI, leading to more iodoorganic by-products by reacting with DOMs (Fabian and Gordon, 1997; Bichsel and von Gunten, 1999). Iodoorganic compounds exhibit higher toxicity than their chloro/bromo analogues (Richardson et al., 2007; Duirk et al., 2011). For example, among the identified DBPs, iodoacetic acid (IA) is considered to be the most genotoxic, which is more easily generated by chlorination in iodide-containing water (Plewa et al., 2004).

In this research, chlorine, ozone, chlorine dioxide, ultraviolet and their combinations were adopted to treat effluents before disinfection from four wastewater reclamation plants in northern China. By applying the SOS/umu test, removal efficiency of the genotoxicity under different disinfection processes was comprehensively compared. The effect of UV dose and iodide concentration on genotoxicity change during disinfection was also investigated. Three-dimensional fluorescence-excitation emission matrix (F-EEM) spectroscopy was employed to examine the relationship between genotoxicity change and contaminant removal.

2. Material and methods

2.1. Water sampling

Four large scale wastewater reclamation plants (WWRPs) accepting sewage from different regions in northern China were selected for this study, named WWRP1, WWRP2, WWRP3 and WWRP4. Samples were collected from influents, effluents before disinfection, and effluents after disinfection of the four WWRPs, and the recipient stream near the WWRP2 outlet was also sampled.