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The removal process of 2,2-dichloroacetamide (DCAcAm), a new disinfection by-product, in drinking water treatment process and its toxicity on zebrafish

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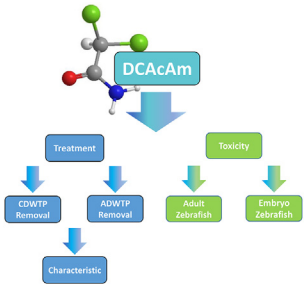
HIGHLIGHTS

- The MFP, which was DCAcAm-specific, was firstly applied.
- DCAcAm could not be effectively removed via CDWTP.
- The hydrophilic NOM and NOM with MW <1 kDa and >10 kDa had a higher MFP of DCAcAm.
- DCAcAm caused delayed development and malformation to zebrafish embryos.
- DCAcAm could cause acute DNA damage to the adult zebrafish.

GRAPHICAL ABSTRACT



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ABSTRACT

The removal process of 2,2-dichloroacetamide (DCAcAm), a new disinfection by-product (DBP) in conventional drinking water treatment plant (C-DWTP) and advanced DWTP (ADWTP) was studied with newly maximum formation potential (MFP) process. It was demonstrated that the advanced treatment displayed greater removal efficiency towards DCAcAm formation potential (MFP) than the conventional treatment. The hydrophilic natural organic matter and natural organic matter with molecular weight <1 kDa or >10 kDa leaded to more DCAcAm formation, and the aromatic protein was inferred as one part of DCAcAm precursor. DCAcAm was found to cause delayed development and malformation to zebrafish embryos at embryonic growth stage. Compared with heart toxicity, it caused a significant neuron toxicity. It also could cause the acute DNA damage to adult zebrafish, which should be extremely cautioned.

1. Introduction

Since the formation of disinfection by-products (DBPs) was confirmed due to the reaction between natural organic matter (NOM) and chlorine in 1970s, the formed DBPs in disinfection process have attracted great attention (Rook, 1974). It is well

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known that most DBPs are carcinogens or suspected carcinogens to human beings, which can cause long term disease. Therefore, DBPs are strictly regulated with water quality standards in many countries (Richardson et al., 2002). The carbonaceous disinfection by-products (C-DBPs) such as trihalomethanes (THMs) have been well studied and are regulated with maximum concentration limits (Richardson et al., 2007). However, nitrogenous DBPs (N-DBPs), especially the emerging haloacetamides (HAcAms) which demonstrated higher cytotoxicity and genotoxicity than C-DBPs and other N-DBPs (e.g., HANs) (Plewa et al., 2008), have been limited studied in terms of their formation, removal and toxicity properties and thus cause a wide concern (Chu et al., 2010b). Among the detected HAcAms, DCAcAm is present at the highest concentration and caused significantly chronic cytotoxicity and acute genotoxicity (Krasner et al., 2006; Plewa et al., 2008). Therefore, it is necessary to fully investigate the DCAcAm, especially its removal performance during the drinking water treatment plant (DWTP) process.

In order to reduce the content of DBPs, removing DBP precursor before disinfection is more effective than altering the disinfection process or directly removing the formed DBPs (Rehan and Manuel, 2004). The DBP precursor is removed according to its physico-chemical properties. Among the properties of organic matter, both molecular weight (MW) and hydrophobicity are widely investigated to promote the control of DBPs (Lin et al., 2014a). However, these two properties of DCAcAm precursor have not been comprehensively studied.

Zebrafish, a tropical fish, is now the valuable vertebrate model to identify the toxicity (Lima et al., 2013; Pavagadhi et al., 2014). Its high genomic homology with humans (over 80%) may enable a significant correlation of the data obtained between the two species (Lin et al., 2014b). Its fast embryonic development (72 h from zygote to larvae) may facilitate toxicity study during development (Oliver et al., 2011). However, there is still no research on the toxic effects of DCAcAm on zebrafish embryos. The hatchability, mortality and malformation have been frequently applied to measure the developmental toxicity induced by contaminants to organism embryos (Lin et al., 2014b). The heart circulatory function plays a key role in the metabolism and development of zebrafish (Rubinstein, 2006) and the heart rate is an important indicator of circulatory function. The spontaneous movement of zebrafish embryos is a reflection of body coordination ability and frequently used to analyze integrative neuronal function (Zou et al., 2009). In many previous reports, the single cell gel electrophoresis (SCGE) assay has been used to study the toxicity of contaminants in aquatic environment (Liu et al., 2014; Yang et al., 2014; Glei et al., 2009). It is a simple and effective method for quantitatively measuring genomic DNA damage. In our previous study, we have found that DCAcAm could cause enzymatic damage to adult zebrafish at concentrations of 10, 50, 100, 500, and 1000 μg/L (Yu et al., 2015). But the genotoxicity of DCAcAm towards adult zebrafish has not been studied.

In our previous research, we have detailed the occurrence, biomarker response and the bio-concentration factor of DCAcAm (Yu et al., 2015). The results showed that the concentration of DCAcAm reached μg/L level in finished water of DWTPs around Yangtze River or Taihu Lake in China. DCAcAm could cause the acute metabolism damage and was easily accumulated in zebrafish. However, in this research, we have studied the formations and removal efficiency of DCAcAm in both conventional and advanced DWTPs (CDWTP and ADWTP). We also comprehensively studied the MW and hydrophobicity of DCAcAm precursor. The toxic effects of DCAcAm on zebrafish embryonic development and adult zebrafish were also detected.

2. Methods and materials

2.1. Chemicals and zebrafish culture

The DCAcAm was obtained from Alfa Aesar (Karlsruhe, Germany). The chemical structure of DCAcAm is shown in Fig. S1. Other chemicals were analytical grade and purchased from Nanjing Chemical Reagent Co. Ltd. (Nanjing, China). Ethyl acetate (ETAC) was of HPLC grade and purchased from Tedia (USA). The ultrapure water was obtained using a Millipore Milli-Q Gradient water purification system (Billerica, USA).

The adult zebrafish was obtained from the Model Animal Research Center of Nanjing University. The breeding method was referred to Westerfield's method (Westerfield, 1995). The detailed breeding method is shown in Text S1. Embryos produced by adult zebrafish were collected and cleaned using MilliQ water (Millipore Corp., USA) to remove impurities. The embryos, which were at least at the four-cell stage, were selected to perform the subsequent embryonic development toxicity test. The culture solution for zebrafish embryos, was conformed with recommendations outlined in the Zebrafish book (Hallare et al., 2006).

2.2. Sample and collection

Water samples were collected in April 2015 from the CDWTP and ADWTP, both receiving raw water from Yangtze River and the water treatment scales of these two waterworks are 450 and 600 thousand tons per day, respectively. Fig. S2 shows the location of the two waterworks. The treatment processes in CDWTP were as follows: coagulation, sedimentation, sand filtration and chlorination. That of ADWTP mainly included such units as coagulation, sedimentation, sand filtration, ozonation, biological activated carbon (BAC) filtration and chlorination. The treatment process and the sampling points in each DWTP were displayed in Fig. S3. The water samples were collected in pre-cleaned 1 L glass bottles. Buffer solution, prepared by 0.2 M sodium acetate and 0.3 M acetic acid, was immediately added to each bottle to hold the pH value at about 5.0, where DCAcAm was stable (Chu et al., 2010a). The bottles were then stored in an ice-bath immediately. The same samples were analyzed in triplicates.

2.3. Characteristics of precursor

In hydrophobicity test, XAD-8 and XAD-4 resins (Sigma, USA) were used to fractionate the NOM. Those two resins were activated as described by Leenheer, 1981). The XAD-8 and XAD-4 resin columns should be rinsed with 0.1 N NaOH, 0.1 N HC1, and distilled water just before application of the sample. Cleaned resins should be stored in methanol. The effluent, firstly through XAD-8 and then XAD-4, was defined as the hydrophilic fraction. The fraction adsorbed by XAD-8 was defined as hydrophobic NOM and that retained by XAD-4 as transphilic fraction. The detailed steps were described in Text S2 and shown in Fig. S4.

In MW detection, the filtered water was fractionated via Millipore ultrafiltration membranes (Amicon, Beverly, MA) with molecular weight cut-offs (MWCO) of 10 kDa, 5 kDa, 3 KDa and 1 kDa respectively. Therefore, five fractions were obtained, namely MW of >10, 5–10, 3–5, 1–3 and <1 kDa. The ultrafiltration process was referred to Hua et al., 2015. The detailed steps are described in Text S3 and shown in Fig. S5.

The dissolved organic carbon (DOC) concentration of each fraction was measured using a TOC analyzer (Multi N/C 2100, German). Each sample was analyzed in triplicates.