



# The shadow of dichloroacetoneitrile (DCAN), a typical nitrogenous disinfection by-product (N-DBP), in the waterworks and its backwash water reuse



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## HIGHLIGHTS

- The DCAN FP's remove by conventional and advanced treatment process was investigated.
- DCAN FP in effluent of old GAC filter increased due to the decreased adsorption.
- The recycle ratio of sand filters' backwash water >5% may cause DCAN's accumulation.

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## ABSTRACT

Dichloroacetoneitrile (DCAN) is one of nitrogenous disinfection by-products (N-DBPs) with strong cytotoxicity and genotoxicity. In this study, the formation potential (FP) of DCAN was investigated in the samples of six important water sources located in the Yangtze River Delta. The highest formation concentration of DCAN was 9.05 µg/L in the water sample taken from Taihu Lake with the lowest SUVA value. After the NOM fractionation, the conversion rate of hydrophilic fraction to DCAN was found the highest. Subsequently, a waterworks using Taihu Lake as water source was chosen to research the FP variations of DCAN in the treatment process and backwash water. The results showed that, compared to the conventional treatment process, O/biological activated carbon (BAC) process increased the removal efficiency of DCAN from 21.89% to 50.58% by removing aromatic protein and soluble biological by-products as main precursors of DCAN. The DCAN FP in the effluent of BAC filters using old granular activated carbon was higher than that in the influent and the DCAN FP of its backwash water was lower than that in raw water. In the backwash water of sand filters, the DCAN FP higher than raw water required the recycle ratio less than 5% to avoid the accumulation of DCAN.

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## 1. Introduction

The disinfection by-products have been concerned in the drinking water treatment field since they were found in the early 1970s (Bellar and Kroner, 1974; Rook, 1974). Disinfectants are used to inactivate microorganisms such as bacterial and virus before the drinking water was delivered to people. However, disinfectants can react with NOM in water to form DBPs, which are carcinogens or

suspected carcinogens to human beings. Among more than 600 identified DBPs, the N-DBPs are substantially more toxicity than the C-DBPs (Plewa, 2008).

HANs is one of the main N-DBPs identified with regular frequency in water (Richardson et al., 2007; Bond et al., 2011; Shah et al., 2011; Barceló, 2012). HANs typically occur at higher concentration in the treated water using chlorine or chloramines disinfection compared to many other N-DBPs, e.g., nitrosamines, halonitromethanes and haloacetamides (Krasner et al., 2006; Chu et al., 2009). HANs have significantly higher cytotoxicity and genotoxicity than regulated THMs and HAAs (Mark et al., 2007). DCAN accounts for a considerable proportion in the sum of detected HANs (Krasner et al., 2006). According to the Canadian survey (Williams

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## List of symbols

### Symbols & abbreviations

DCAN	dichloroacetonitrile
N-DBP	nitrogenous disinfection by-product
FP	formation potential
SUVA	specific ultraviolet absorbance
DOC	dissolved organic carbon
DON	dissolved organic nitrogen
BAC	biological activated carbon
DBP	disinfection by-product
NOM	natural organic materials
C-DBP	carbonaceous disinfection by-product
HAN	haloacetonitrile

THM	trihalomethane
HAA	haloacetic acid
DHAN	dihaloacetonitrile
RfD	referenced dose
DCAcAm	dichloroacetamide
COD	chemical oxygen demand
MTBE	methyl <i>tert</i> -butyl ether
HPLC	high performance liquid chromatography
LLE	liquid-liquid extraction
ECD	electron capture detector
EEM	excitation-emission matrix
GC	gas chromatography
MW	molecular weight
GAC	granular activated carbon
TOC	total organic carbon

et al., 1997), DCAN was found in 97% of all samples. Median level of DHAN concentration in the effluent of 11 US drinking water treatment plants with raw water influenced by algal or wastewater was somewhat higher than 4 µg/L (Krasner et al., 2006). In the water-treatment plants of South Korea, DCAN has been frequently reported to be higher than 40 µg/L since 2009 (Shin et al., 2013), while the provisional guideline value of DCAN is 20 µg/L according to the World Health Organization (2004). The oral reference dose is 8 µg per kilogram per day (µg/kg·d), and non-cancer dose for lifetime is 6 µg/kg·d from health advisories of US EPA. However, the stipulated concentration of DCAN has not been required in Chinese standards for drinking water quality.

In order to reduce DBPs, removing precursors of DBPs before disinfection is more effective and practical than changing the disinfection conditions or implementing other technologies to remove the formed DBPs (Sadiq and Rodriguez, 2004; Bond et al., 2012). In previous study (Lin et al., 2016), the removal of DCACAm precursors in drinking water treatment process was investigated to evaluate the removal efficiency of precursors and the formation potentials of seven C-DBPs and five N-DBPs, in the effluent of each conventional process, were also examined. Although the advanced oxidation technologies have not been used widely in water treatment plants, they showed the good removal performance for DCAN precursors (Chu et al., 2014, 2015). The previous results (Krasner et al., 2012) have indicated that the lime softening and ozonation were the main contributors to the precursors removal of dihalogenated HAN in some waterworks. However, the removal performance of DCAN precursors in water treatment process has not been investigated in detail.

Plenty of waterworks have reused the backwash water from sand filter or BAC filter in order to conserve water resources. The usual reuse method of backwash water is directly mixing it with raw water prior to coagulation process. The backwash water recycling has been researched in many issues, from biosafety on giardia and cryptosporidium (Karanis et al., 1998; Edzwald et al., 2003; Bourgeois et al., 2004) to water quality variations such as zeta potential, turbidity, COD and so on (Park et al., 2014; Chen et al., 2015). DBP formation in drinking water mixed with 10% chlorinated UF-filter backwash water was investigated and the high initial concentrations of THM and HAA species brought challenges to the finished water quality (Walsh et al., 2008). With higher THM FP and HAA FP found in backwash water in comparison to those measured in the raw water, the recycling of wastewater (by blending 10% untreated backwash water with raw water) would contribute to additional DBP formation above the baseline of 20 µg/L in some waterworks (McCormick et al., 2010). But whether the wastewater

recycling could cause the increase of DCAN FP has not been studied.

In this paper, the DCAN FP in raw water from different water sources of Yangtze River Delta was investigated to explore the relationship between water quality and DCAN FP. The removal efficiency of DCAN in both conventional and advanced water treatment process was also studied. In addition, the issue of backwash water recycling was considered according to the variation of DCAN FP at different recycling ratio.

## 2. Methods and materials

### 2.1. Materials and chemicals

DCAN and resins were obtained from Sigma-Aldrich (United States). The molecular formula of DCAN is C<sub>2</sub>HCl<sub>2</sub>N (109.94 g/mol). MTBE was HPLC grade and purchased from AccuStandard. Other chemicals were analytical grade and purchased from Sinopharm Chemical Reagent Co. Ltd., China. The ultrapure water was obtained using a Millipore Milli-Q Gradient water purification system.

### 2.2. Water samples

Samples of water source were collected from Yangtze River, Taihu Lake and Yangcheng Lake in September, 2016 (Table 1). Among them, although Y2 is near Taihu Lake, it is a tributary of the Yangtze River. The water samples, reflecting water quality of the treatment process effluent or backwash water, were collected from a waterworks near Taihu Lake, using raw water taken from water sources T1. All water samples were collected in pre-cleaned 1 L glass bottles. Once collected, the waters were immediately filtered through a microporous filter membrane (0.45 µm) and stored in the dark at 4 °C until use.

### 2.3. Chlorination of water samples

In this paper, DCAN FP in chlorination rather than chloramination was investigated due to the wide use of chlorine as a disinfectant in the waterworks using Taihu Lake as water source. The DCAN FP tests were performed according to the methods developed by Krasner et al., (Krasner et al., 2006; Compton et al., 2011). A sodium hypochlorite solution (active chlorine = 20000 mg/L) was used as free chlorine stock solution. The disinfectant dosages for FP tests were calculated by Eq. (1) for free chlorine.

$$\text{Cl}_2 \text{ dosage (mg/L)} = 3 \times \text{DOC (mg-C/L)} + 7.6 \times \text{NH}_3 \text{ (mg-N/L)} + 10 \text{ (mg/L)} \quad (1)$$

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