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Factors affecting the formation of nitrogenous disinfection by-products during chlorination of aspartic acid in drinking water

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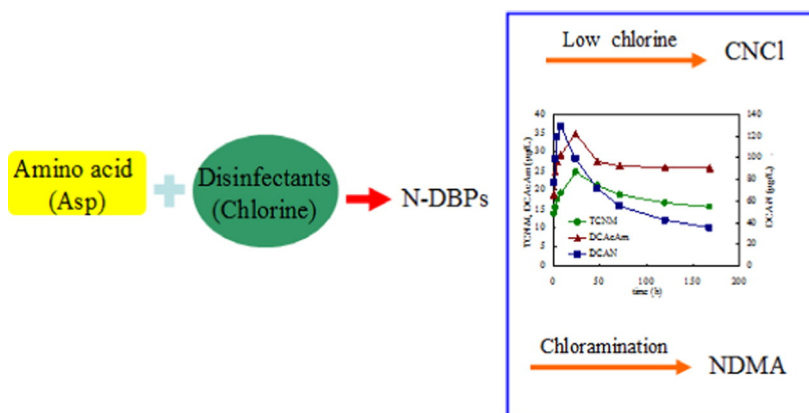
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HIGHLIGHTS

- CNCl was detected when the Cl₂/Asp molar ratio was lower than 5 during the chlorination of Asp.
- Higher temperatures reduced the formation of DCAN and DCACAm, but increased TCNM formation.
- DCAN and DCACAm formation decreased and TCNM formation increased, with increasing free chlorine contact time during chloramination.
- NDMA could be produced during chloramination of Asp.
- The presence of bromide ions shifted N-DBPs to more brominated species.

GRAPHICAL ABSTRACT



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ABSTRACT

The formation of emerging nitrogenous disinfection by-products (N-DBPs) from the chlorination of aspartic acid (Asp) was investigated. The yield of dichloroacetonitrile (DCAN) was higher than other N-DBPs, such as dichloroacetamide (DCACAm) and chloropicrin (TCNM) during the chlorination of Asp. The formation of DCAN, DCACAm, and TCNM all showed a trend of first increasing and then decreasing during the chlorination of Asp with increasing contact time. The dosage of chlorine had an impact on the formation of DCAN, DCACAm, and TCNM. The highest yields of DCAN and DCACAm appeared when the Cl₂/Asp molar ratio was about 20, the yield of TCNM increased with increasing the Cl₂/Asp molar ratio from 5 to 30 and TCNM was not produced when the ratio was less than 5. Cyanogen chloride (CNCl) was detected when the Cl₂/Asp molar ratio was lower than 5. N-DBPs formation was influenced by pH. DCAN formation increased with increasing pH from 5 to 6 and then decreased with increasing pH from 6 to 9, but DCACAm and TCNM increased with increasing pH from 5 to 8 and then decreased. Higher temperatures reduced the formation of DCAN and DCACAm, but increased TCNM formation. DCAN and DCACAm formation decreased, and relatively stable TCNM formation increased, with increasing free chlorine contact time during chloramination. N-nitrosodimethylamine (NDMA) was produced during chloramination of Asp and increased with prolonged chloramination contact time. The presence of bromide ions enhanced the yields of haloacetonitriles and shifted N-DBPs to more brominated species.

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1. Introduction

As its low cost and high disinfection efficiency in inactivating pathogenic microorganisms, chlorine has been widely used to deliver major public health improvements worldwide by preventing the spread of waterborne diseases. However, the formation of hazardous disinfection by-products (DBPs), such as trihalomethanes, haloacetic acids, nitrosamines, cyanogen halides, haloacetonitriles (HANs), haloacetamides, halonitromethanes and so on, through reactions between precursor materials and chlorine has been reported (Ma et al., 2014; Muellner et al., 2007; Stalter et al., 2016; West et al., 2016).

In recent years, the number of reports on nitrogenous disinfection by-products (N-DBPs) has increased rapidly (Bond et al., 2011; Krasner et al., 2013). These species typically occur in drinking water at low concentrations, but have high cytotoxicity and genotoxicity (Frank et al., 1988; Muellner et al., 2007). Collectively, dichloroacetonitrile (DCAN), chloropicrin (TCNM), dichloroacetamide (DCAcAm), *N*-nitrosodimethylamine (NDMA) and cyanogen chloride (CNCl) are the most important DBPs that are often identified in chlorinated water (Chu et al., 2016; Richardson et al., 1999; Shah and Mitch, 2012; Yang et al., 2010).

The presence of dissolved organic nitrogen (DON) in surface water has a negative effect on drinking water security as it is an important precursor of N-DBPs (Krasner et al., 2009; Lee et al., 2007). Amino acids are the most extensively studied precursors of DON. They are widespread in surface water and are difficult to remove by conventional drinking water treatment (Jia et al., 2016). Aspartic acid (Asp) is a naturally present amino acid in DON and is an important component of peptides, proteins and algae organic compounds (Chu et al., 2012; Liu et al., 2015). Studies have indicated that Asp had one of the highest N-DBPs yields among 20 free amino acids tested (Chu et al., 2012; Yang et al., 2012a). The DCAcAm yield from Asp was 0.231 mmol/mol (Asp = 0.05 mM, Cl₂ = 0.2 mM, pH = 7.5) and its source was the Qingcaosha Reservoir in China (Chu et al., 2010). Asp produced TCNM and DCAN during chlorination (Asp = 0.1 mM, Cl₂ = 3 mM, pH = 7.2) (Yang et al., 2012a).

Chloramination is often performed on the laboratory scale, but ammonia and chlorine are added separately in practical processes. The free chlorine contact time may impact N-DBPs formation especially NDMA during post-chloramination (Schreiber and Mitch, 2005). Most brominated DBPs are more cytotoxic and genotoxic than their chlorinated analogues, the distribution can be shifted from DCAN to bromochloroacetonitrile (BCAN) and dibromoacetonitrile (DBAN) by increasing the bromide ion concentrations (Guanghui and David, 2008; Plewa et al., 2002). In addition, the influence of chlorination on the formation of N-DBPs from Asp added into the effluent of filters is currently unknown.

A better understanding of the comprehensive formation mechanism of N-DBPs during chlorination disinfection may improve the accuracy of N-DBP occurrence prediction and disinfection practice processes could be optimized to minimize the formation of N-DBPs. The main objectives of this study were to assess the role of Asp in the formation of the most common N-DBPs, namely, DCAN, TCNM, DCAcAm, NDMA and CNCl, and investigate the influence of operational parameters, such as contact time, chlorine dosage, pH, temperature, chloramination and bromide ion concentration on these N-DBPs formation.

2. Materials and methods

2.1. Materials and chemicals

Table 1 shows the water quality used in this study, collected from the V-type clarifying filtration in a drinking water treatment plant of Ningbo city, Zhejiang Province, China. DCAN, TCNM, DCAcAm, NDMA, CNCl, BCAN and DBAN were not detected in the collected water used for the experiments. Except where noted in the text, all reagents used

Table 1

Water quality collected from filtration.

pH	Temperature (°C)	Turbidity (NTU)	COD _{Mn} (mg/L)	NH ₃ -N (mg/L)	Br ⁻ (μg/L)
6.9	21	0.07	0.72	0.06	5.2

in the experiments were of analytical purity. Asp was obtained from Wako (Osaka, Japan), Four HANs, including DCAN, trichloroacetonitrile (TCAN), BCAN and DBAN, TCNM and NDMA were purchased from Sigma-Aldrich (St. Louis, Missouri, USA), DCAcAm and CNCl were obtained from Alfa Aesar (Karlsruhe, Germany). All solutions were prepared using ultrapure water and all bottles were prewashed at 105 °C for 24 h. A stock of free chlorine solution was prepared from 5% sodium hypochlorite (NaOCl) (from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China), diluted to 1000 mg/L as chlorine and stored in an aluminum foil-covered, glass stoppered flask. Bromide ion solutions were prepared using the dilution of sodium bromide solution (NaBr, GR grade). Buffer solutions at different pH values were prepared by phosphate salts.

2.2. Experimental procedures

For all experiments during chlorination/chloramination, the concentration of Asp solutions was 0.1 mM and the disinfectant to Asp molar ratio was 30:1, providing an excess of disinfectant. All chlorinated/chloraminated samples were stored headspace-free in the dark at room temperature (22 ± 1 °C). After reaction for a certain time, the samples were quenched and extracted immediately with methyl *tert*-butyl ether, based on US EPA Method 551.1. In order to study the influence of operation parameters on N-DBP formation, an orthogonal matrix experimental was designed. In this design, chlorine dosages to Asp molar ratios were 1–30, pH were 5–9, temperatures were 10–30 °C, adding times of post-chloramination were 0–24 h and bromide ion concentrations were 0–5 mg/L.

2.3. Analytical methods

Residual and total chlorine concentrations were detected by a portable spectrophotometer (HACH DR1900) based on HACH method 8021. HANs, and TCNM were analyzed by purge and trap (OI Analytical, Eclipse 4660, College Station, Texas, USA) and gas chromatography/mass spectrometry (GC/MS, Shimadzu-QP-2010 Ultra, Japan), DCAcAm was measured by liquid-liquid extraction and GC/MS (Shimadzu-QP2010) (Chu et al., 2012). The detection limits for them were below 1 μg/L, and recovery of them were 81.5%–118.7%. For the analysis of NDMA, a sample was used for solid phase extraction using procedures described by the US EPA 521 (Munch and Bassett, 2006). NDMA was determined by following the method described by Zhang et al. (Zhang et al., 2014). The method detection limit for NDMA was 4 ng/L. The recovery of NDMA was 82% ± 5%. The analytical technique to monitor CNCl was in-line membrane introduction mass spectrometry (MIMS). A detailed description of the reactor and MIMS detection system can be found in an earlier publication (Na and Olson, 2004). An HPLC pump (Acuflo Series I, Lab Alliance Inc.), a membrane inlet probe, and a quadrupole mass spectrometer with electron impact ionization (Hewlett-Packett 5972) were used in MIMS detection system. The analysis of CNCl concentration was virtually nondestructive and the detection limit was about 4.5 μg/L.

3. Results and discussion

3.1. N-DBPs formation

Fig. 1 shows the formation of N-DBPs in different reaction time during chlorination of Asp. It was found that the concentrations of N-DBPs

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