



Effects of UV/PS and UV/H₂O₂ pre-oxidations on the formation of trihalomethanes and haloacetonitriles during chlorination and chloramination of free amino acids and short oligopeptides



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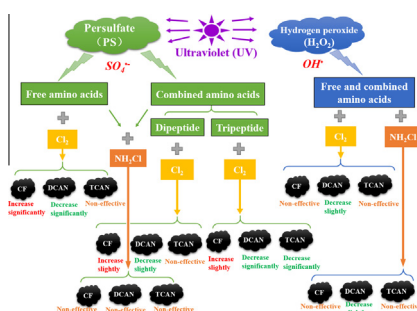
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HIGHLIGHTS

- UV/PS presented better performance on controlling HAN formation than UV/H₂O₂.
- UV/PS somewhat increased THM formation from AA chlorination, but UV/H₂O₂ did not.
- UV/PS and UV/H₂O₂ did not obviously alter the DBP formation prior to chloramination.
- More bromine was incorporated into DHANs than into THMs from combined AAs.
- UV/PS decreased the formation of brominated DBPs, but UV/H₂O₂ did not.

GRAPHICAL ABSTRACT



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ABSTRACT

Free amino acids (AAs) can form trihalomethanes (THMs) and haloacetonitriles (HANs) during chlorination in drinking water treatment plants, but they account for a small fraction of dissolved organic nitrogen (DON) pool in natural waters. In contrast, combined AAs contribute to a greater identifiable DON portion, but the information on the formation and control of THMs and HANs from them is limited. Ultraviolet/hydrogen peroxide (UV/H₂O₂) and ultraviolet/persulfate (UV/PS), two typical UV-based advanced oxidation processes (UV-AOPs), are both promising drinking water technologies for the reduction of trace organic contaminants. The objective of this study is to examine the effects of the two UV-AOP pre-treatments on the formation of carbonaceous and nitrogenous disinfection by-products (C- and N-DBPs), which were indicated by THMs and HANs, respectively, during post-chlorination and chloramination of tyrosine (Tyr) in free and combined forms. Results showed that direct UV photolysis, PS pre-oxidation alone or H₂O₂ pre-oxidation alone did not significantly impact the THM and HAN formation during subsequent chlorination. However, the two UV-AOP pre-treatments could somewhat alter the DBP formation. In general, under the identical doses (UV irradiation dose and molar oxidant dose), UV/PS pre-treatment better controlled the HANs than UV/H₂O₂ during AA

Abbreviations: AA, amino acid; Ala, alanine; AOP, advanced oxidation process; BCAN, bromochloroacetonitrile; BDCM, bromodichloromethane; BF, bromoform; BIF, bromine incorporation factor; BUF, bromine utilization factor; C-DBPs, carbonaceous disinfection by-products; CF, chloroform; DBAN, dibromoacetonitrile; DCAN, dichloroacetonitrile; DBPs, disinfection by-products; DBCM, dibromochloromethane; DOC, dissolved organic carbon; DON, dissolved organic nitrogen; DOM, dissolved organic matter; DWTPs, drinking water treatment plants; HANs, haloacetonitriles; H₂O₂, hydrogen peroxide; MW, molecular weight; N-DBPs, nitrogen-containing disinfection by-products; PS, persulfate; P&T, purge and trap; TCAN, trichloroacetonitrile; THMs, trihalomethanes; Tyr, tyrosine; UV, ultraviolet.

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post-chlorination, though UV/PS might slightly promote the formation of chloroform. In contrast, the two UV-AOP pre-treatments did not obviously affect the THM and HAN formation when post-chloramination was applied. In addition, a marked difference was observed in the speciation of DBPs formed from free AAs and short oligopeptides in the presence of bromide. For free Tyr, more brominated THMs were formed than brominated HANs; but for short oligopeptides, more bromine was incorporated into HANs than THMs.

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1. Introduction

Although disinfection as an outstanding public health achievement in the 20th century is a key drinking water treatment step, it is challenged by the formation of unwanted disinfection by-products (DBPs). DBPs are primarily formed from the reactions between disinfectants (e.g., chlorine) and certain organic matters or inorganic species (e.g., bromide and iodide) [1]. Over the past four decades, a few hundreds of DBPs have been identified [2,3]. Among them, nitrogen-containing DBPs (N-DBPs) have gradually become a new focus, because they are generally more toxic than carbonaceous DBPs (C-DBPs) such as trihalomethanes (THMs), which are well studied and regulated in many countries [4–6]. Haloacetonitriles (HANs), a commonly occurring N-DBP class, typically occur at $\mu\text{g/L}$ levels in chlorinated and/or chloraminated drinking water [1,7]. Moreover, they are generally more cytotoxic and genotoxic than the regulated THMs [8]. Therefore, it is essential to simultaneously control the formation of unregulated N-DBPs (e.g., HANs) and regulated C-DBPs (e.g., THMs) during chlorination for drinking water treatment.

Generally, less N-DBPs are formed than several common C-DBPs in drinking water. One of the major reasons is that dissolved organic nitrogen (DON), believed to be the main precursors of N-DBPs (e.g., HANs), is usually less than dissolved organic carbon (DOC) that are the principal precursors of C-DBPs (e.g., THMs) in source water. Nevertheless, water resource shortages and growing water demands have recently spurred water treatment utilities to search new or additional water sources, including the water bodies impaired by municipal wastewater effluents and/or algal blooms. Microorganism-associated (e.g. algae, bacteria) products, wastewater effluent organic matters (EfOMs) and proteinaceous compounds are typically found in such water sources, leading to a much higher level of DON in which amino acids (AAs) constitute an important class of the DON pool [6,9–12]. Free AAs were selected as the precursors in many previous studies on the halogenated N-DBP formation [6,11]. However, free AAs merely accounts for a minor fraction (<6%) of the DON pool [13–15]. In contrast, combined AAs (e.g. oligopeptides and proteins) represent the majority of the identifiable parts [15–19]. Typically, free AAs range from ng/L to $\mu\text{g/L}$, while combined AAs (e.g., polypeptides) are observed at mg/L level in surface waters [13,15,20]. Conventional treatment processes in drinking water treatment plants (DWTPs) preferentially remove the high molecular weight (MW) organics (e.g., proteins) prior to chlorination disinfection, and part of the low-MW free AAs and combined AA short oligopeptides (e.g., $\text{MW} < 1000 \text{ Da}$) likely pass through to react with chlorine to form DBPs [15,18,21]. Therefore, there is a demand to investigate the formation and control of DBPs produced from the chlorination of both free AAs and combined AAs. As known, tyrosine (Tyr) can react with chlorine to form a range of C-DBPs and N-DBPs, including THMs and HANs, thereby being a representative model AA precursor of C-DBPs and N-DBPs [6,11,22]. In the study, free Tyr and the combined Tyr in some short oligopeptides, including tyrosine-alanine (Tyr-Ala), alanine-tyrosine (Ala-Tyr), and tyro-

sine-tyrosine-tyrosine (Tyr-Tyr-Tyr), were selected as model precursors. These model precursors possess similar molecular structures with or without peptide bonds (Fig. 1) [23,24].

A sound strategy to control the DBP formation is to effectively remove their precursors before chlorination. However, conventional water treatment processes (e.g., coagulation-sedimentation-filtration) poorly reduce low-MW DON precursors [15,18,21]. Therefore, pre-treatment processes are needed to improve the removal. Ultraviolet/hydrogen peroxide (UV/ H_2O_2) and ultraviolet/persulfate (UV/PS), two typical UV-based advanced oxidation processes (UV-AOP), are both promising drinking water technologies for the reduction of a broad spectrum of synthetic organic contaminants and undesirable natural organic constituents [25–30]. However, it is unknown whether the two UV-AOP pre-treatments can control the formation of HANs and THMs from the selected free and combined AA precursors (Tyr, Tyr-Ala, Ala-Tyr, and Tyr-Tyr-Tyr) during subsequent chlorination. Thereby, the objective of this study is to compare the effects of two UV-AOP pre-treatments (UV/ H_2O_2 versus UV/PS) on the formation of HANs (the representative of N-DBPs) and THMs (the representative of C-DBPs) during subsequent chlorination of free and combined Tyr.

2. Materials and methods

2.1. Materials

All the chemicals used were at least reagent grade except otherwise noted. The HANs mixture standard solution (Supelco 48,046), containing dichloroacetonitrile (DCAN), trichloroacetonitrile (TCAN), bromochloroacetonitrile (BCAN), and dibromoacetonitrile (DBAN), and the THMs mixture standard solution (Supelco 47,904) containing chloroform (CF), bromodichloromethane (BDCM), dibromochloromethane (DBCM), and bromoform (BF) were purchased from Sigma-Aldrich (St Louis, Missouri, USA). Tyr (98.5%), Tyr-Ala (98%), Ala-Tyr (98%), and Tyr-Tyr-Tyr (98%) were obtained from Aladdin Industrial Inc. (Shanghai, China). These AA solutions were freshly prepared prior to use. Guaranteed reagent grade reagents including sodium hypochlorite (NaOCl), sodium hydroxide (NaOH), hydrochloric acid (HCl), sodium bromide (NaBr), ammonium chloride (NH_4Cl), ascorbic acid and glacial acetic acid were purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). H_2O_2 solution (35% v/v, stab.), standardized by colorimetric method using DPD [31], was obtained from Alfa Aesar, USA. Sodium persulfate ($\text{Na}_2\text{S}_2\text{O}_8 \geq 99.5\%$) was purchased from Sinopharm Chemical Reagent Co., Ltd (Shanghai, China). All the solutions were prepared using ultrapure water produced with a Millipore Milli-Q Gradient water purification system (Billerica, Massachusetts, USA). All the bottles were prewashed with phosphate-free detergent, rinsed with ultrapure water, and dried in an oven at 105°C for 24 h. Chlorine solutions were prepared by diluting a 6% NaOCl solution with ultrapure water standardized daily prior to use. Monochloramine (NH_2Cl) solution was freshly prepared as follows. NaOCl solution was gently added into a

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