



Photochemical behavior of dissolved and colloidal organic matter in estuarine and oceanic waters



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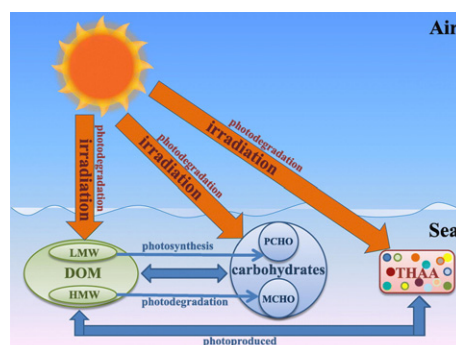
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HIGHLIGHTS

- terrestrially derived DOM was more susceptible to irradiation than autochthonous DOM.
- Photo-induced synthetic reaction transformed the LMW matters to polysaccharides.
- Photo-induced degradation reaction decomposed the HMW DOM to monosaccharides.
- Stable compositions of THAA significantly accumulated during irradiation.
- Labile compositions of THAA were prone to photodegradation.

GRAPHICAL ABSTRACT



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ABSTRACT

Chromophoric dissolved organic matter (CDOM), carbohydrates, and amino acids were analyzed to investigate the photochemistry of total dissolved (<0.22 μm) organic matter (DOM), high-molecular-weight (HMW, 1 kDa–0.22 μm) DOM and low-molecular-weight (LMW, <1 kDa) DOM at stations in the Yangtze River and its coastal area, and in the Western Pacific Ocean. Results revealed that the humic-like and tryptophan-like CDOM fluorescent components in riverine, coastal, and oceanic surface waters were photodegraded during irradiation. However, the photochemical behavior of tyrosine-like component was obscured by the excessive fluorescence intensities of humic- and tryptophan-like fluorescent components. Light sensitivity varied depending on the source material; terrestrially derived DOM was more susceptible to irradiation than autochthonous DOM. In contrast to the expected photodegradation of CDOM, photo-induced synthetic reaction transformed the LMW matters to polysaccharides (PCHO) and degradation reaction decomposed the HMW DOM to Monosaccharides. Colloidal DOM preferentially underwent photodegradation, whereas permeate DOM mainly photosynthesized PCHO. The total hydrolysable amino acid (THAA) pool changed because of the additional input by the photodegradation of DOM or THAA itself. The compositions of THAA changed during the irradiation experiments, indicating that the different photochemical behavior of individual amino acids were related to their different original photoreactivities; the relatively stable amino acids (e.g., Ser and Gly) significantly accumulated during irradiation, whereas photo-active aromatic amino acids (e.g. Tyr and His) were prone to photodegradation.

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The data presented here demonstrated that irradiation significantly influence the conversion between dissolved and colloid organic matter. These results can promote the understanding of irradiation effect on the carbon and nitrogen cycle in riverine, estuarine and oceanic ecosystems.

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1. Introduction

Dissolved organic matter (DOM) is a major form of organic matter in aquatic environments. Photochemical reactions and their interaction with biological processes are important pathways for mediating the degradation and cycling of DOM in aquatic food webs; hence, these process act in conjunction with each other to affect ecosystem structure and function (Bushaw et al., 1996; Jørgensen et al., 1998). Photodegradation plays a pivotal role in subsequent biodegradation by yielding a variety of biologically labile or resistant substrates (Mopper and Kieber, 2002). Biodegradation is, in turn, mediated primarily by bacteria that use the biologically labile substrates for growth and respiration. DOM largely consists of chromophoric dissolved organic matter (CDOM), which effectively absorbs solar radiation in the ultraviolet (UV) and visible ranges of the light spectrum (Dillon and Molot, 1997; Nelson et al., 1998; Kragh et al., 2008). Photochemical degradation decreases the ultraviolet and visible light absorption and fluorescence of CDOM in aquatic environments, thereby controlling the penetration of UV in the deep water layers and thus limiting the UVB-protective effect of CDOM on organisms and the extent of further photochemical reactions (Miller and Zepp, 1995; Del Vecchio and Blough, 2002). Photo-induced degradation of high-molecular weight (HMW) CDOM into low-molecular weight (LMW) organic substances might enhance the biological utilization and fate of DOM (Mopper, 1989; Obernosterer et al., 1999; Moran et al., 2000; Obernosterer and Benner, 2004). In some cases, the photodegradation process causes the partial or complete mineralization of DOM (Mopper et al., 2015). Photomineralization can convert DOM into various inorganic forms, such as dissolved inorganic carbon, CO, and CO₂, which can potentially influence carbon cycling (Valentine and Zepp, 1993; Miller and Zepp, 1995; Granéli et al., 1998; Zhang et al., 2006; Johannessen et al., 2007). The chemical composition and origin of CDOM significantly influence its photoreactivity and subsequent bioreactivity (Obernosterer et al., 1999; Benner and Ziegler, 2000; Moran et al., 2000; Vähätalo and Wetzel, 2004; Stubbins et al., 2012; Reader and Miller, 2014). The fluorescence of extracted fulvic and humic acids generally decrease by photochemical degradation under sunlight (Mostofa et al., 2011). Terrestrially produced CDOM, with higher concentrations of aromatic groups than algal produced CDOM in surface waters, is highly susceptible to photodegradation (Helms et al., 2008). By contrast, microbially produced CDOM is more influenced by a complicated photochemical–biological coupling process (Miller, 1997; Moran et al., 2000; Miller et al., 2009). Cherrier et al. (1999) indicated that photo-oxidation may contribute up to 20% of old, refractory DOM to bacteria in the Pacific Ocean and may contribute substantial carbon for bacteria consumption in the Atlantic Ocean (Mopper, 1989). Helms et al. (2013) estimated that the concentration of photochemical refractory dissolved organic carbon (DOC) in the surface water of the Pacific Ocean is 27 μmol of carbon per liter.

Carbohydrates play an integral part in aquatic ecosystems because they serve as structural components and energy storage products in marine organisms. Carbohydrates constitute a large fraction of the marine dissolved and particulate DOM (Benner and Kaiser, 2003; Engel and Händel, 2011), ranging from 1% to 2% of LMW DOC (<1 kDa; Engel and Händel, 2011) and 50%–70% of HMW DOC (>1 kDa; Benner et al., 1992; Aluwihare et al., 1997). Carbohydrates in sea water include neutral sugars, amino sugars, acidic sugars (mainly uronic acids), and phosphorylated and sulfated sugars. In general, carbohydrates are mainly composed of monosaccharides (MCHO, free monomers) and polysaccharides (PCHO, neutral sugars released after acid hydrolysis of

HCl). Large amounts of carbohydrates are released into seawater by phyto- and bacterioplankton cells and herbivorous grazing, where they represent the most abundant and reactive components of the dynamic carbon source in the ocean (Engel and Händel, 2011). In fact, carbohydrates are the prime products of photosynthesis which can supply nutrients for plankton productivity and increase the bioavailable carbon substrates for microorganisms, such as proteins, lipids, and nucleic acids (Mykkestad and Børsheim, 2007). However, the photodegradation (or production) and phototransformation of carbohydrates in dissolved and colloidal organic matter remain poorly understood to date. Jørgensen et al. (1998) observed an increase in the content of carbohydrates after exposure of lake waters to sunlight. A typical polysaccharide (sodium alginate) is photodegraded to about 40% of its average molecular weight, and complex heterogeneous polysaccharides (pectin) are photochemically depolymerized from 400 kDa to 200 kDa by ultraviolet light irradiation (Burana-osot et al., 2009, 2010). Sarasidis et al. (2011) also found that sodium alginate is effectively photodegraded through a hybrid photocatalysis–ultrafiltration system. In addition, Wasikiewicz et al. (2005) applied ultraviolet and gamma irradiation to photodegrade sodium alginate and chitosan, and observed obvious decrease in their aqueous solutions.

Photo-induced formation of N-containing (NH₄⁺ or NO₂⁻) inorganic compounds may be typically produced by the degradation of dissolved organic nitrogen (DON) (Bronk, 2002; Kim et al., 2006; Vähätalo and Järvinen, 2007; Li et al., 2008). Several studies estimated the importance of photochemically produced ammonium as a source of nitrogen to the food web and observed that ammonium photoproduction highly correlates with CDOM loss (Bushaw et al., 1996; Bronk, 2002; Stedmon et al., 2007b; Aarnos et al., 2012; Xie et al., 2012). Amino acids represent an important component of the labile fraction of DON. Total hydrolysable amino acids (THAA), the major compound classes of DON, are rapidly transformed from DON pool to higher trophic levels, or, alternatively, are mineralized to dissolved inorganic nitrogen (DIN) via microbial utilization (Fuhrman, 1990; Jørgensen and Tranvik, 1999). However, photochemical accumulation (or loss) of THAA is not as well documented as photo-ammonification, and all published results to date have been obtained with dissolved free amino acids (DFAA), which are minor components of THAA (Linares, 2006; Yang et al., 2009). Mopper et al. (2015) proposed that photochemically produced DFAA. However, Jørgensen and Tranvik (1999) observed a net decrease (or no change) in DFAA concentrations in some irradiated samples but found no specific compositional changes in the DFAA pool. Boreen et al. (2008) demonstrated that histidine (His), methionine (Met), tyrosine (Tyr) and tryptophan (Trp) are subject to photodegradation in natural waters.

In the present study, we estimated the net effect of solar radiation on the degradation (or productions) and transformation of dissolved and colloid organic matter in the surface waters of the Yangtze River and its coastal area, and in the Western Pacific Ocean. Previous studies have examined photochemical behavior of DOM from various sources and different environments. However, the photochemical processes of different molecular weight DOM have not yet been fully understood. The aim of this study was to investigate the photochemical variations of dissolved (<0.22 μm), HMW (1 kDa–0.22 μm) and LMW (<1 kDa) DOM by using the cross-flow ultrafiltration (CFF) system. Individual fluorescent components of CDOM, 14 L-amino acids, MCHO, and PCHO were measured, thus providing novel and comprehensive insight into understanding the DOM dynamics in estuarine and oceanic waters.

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