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## Photo-production of dissolved inorganic carbon from dissolved organic matter in contrasting coastal waters in the southwestern Taiwan Strait, China

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### Abstract

Photo-production of dissolved inorganic carbon (DIC) from chromophoric dissolved organic matter (CDOM) is an important transformation process in marine carbon cycle, but little is known about this process in Chinese coastal systems. This study investigated an estuarine water sample and a coastal seawater sample from the subtropical waters in southeast of China. Water samples were exposed to natural sunlight and the absorption and fluorescence of CDOM as well as the DIC concentration were measured in the summer of 2009. The estuarine water had higher CDOM level, molecular weight and proportion of humic-like fluorescent components than the seawater that exhibited abundant tryptophan-like fluorescent component. After a 3-day irradiation, the CDOM level decreased by 45% in the estuarine water and 20% in the seawater, accompanied with a decrease in the molecular weight and aromaticity of DOM which was inferred from an increase in the absorption spectral slope parameter. The photo-degradation rates of all the five fluorescent components were also notable, in particular two humic-like components (C4 and C5) were removed by 78% and 69% in the estuarine water and by 69% and 56% in the seawater. The estuarine water had a higher photo-production rate of DIC than the seawater (4.4 vs. 2.5  $\mu\text{mol}/(\text{L}\cdot\text{day})$ ), in part due to its higher CDOM abundance. The differences in CDOM compositions between the two types of waters might be responsible for the higher susceptibility of the estuarine water to photo-degradation and hence could also affect the photo-production process of DIC.

**Key words:** dissolved inorganic carbon; photo-production and -degradation; chromophoric dissolved organic matter; estuary; seawater

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### Introduction

Photochemical degradation is an important removal process for aquatic chromophoric dissolved organic matter (CDOM) and provides a source of some biologically important compounds such as inorganic nutrients, low molecular weight organic compounds and carbon monoxide (CO) (Bushaw et al., 1996; Moran and Zepp, 1997; Stedmon et al., 2007; Stubbins et al., 2008; Spencer et al., 2009). Among all the CDOM photo-degradation products, dissolved inorganic carbon (DIC) is produced most efficiently (Moran and Zepp, 1997). The photo-production rate of DIC may be as high as 24  $\mu\text{mol}/(\text{L}\cdot\text{hr})$  in humic-rich freshwaters (e.g., Granéli et al., 1998). Furthermore, the global photo-production rate of DIC ( $10^{14}$ – $10^{15}$  mol/yr) in the ocean is comparable to other  $\text{CO}_2$  cycle terms such as photosynthesis, respiration and anthropogenic input (Johannessen et al., 2007; Wang et al., 2009a). Therefore, the photo-degradation of CDOM and the photo-production of DIC play important roles in both the global and regional carbon cycles and should be studied in more aquatic ecosystems.

The rate of photochemical mineralization is dependent on a number of factors such as CDOM level and composition, radiation intensity and wavelength, previous light exposure,  $\text{O}_2$  content and metal ions (Granéli et al., 1998; Gao and Zepp, 1998; Stubbins et al., 2008; Wang et al., 2009b). For example, large variations in the photo-production rate of DIC are found in both lakes and rivers in part due to changes in the CDOM level (Granéli et al., 1998; Remington et al., 2011). In addition, UV radiation is more effective than longer-wavelength radiation for CDOM photo-degradation (Kieber et al., 2007). Gao and Zepp (1998) and Wang et al. (2009b) reveal the facts that the DIC photo-production is accelerated by increased  $\text{O}_2$  and decreased iron concentrations.

In particular, although recent studies have suggested that the photo-reactivity of CDOM is related strongly to its initial aromaticity (Stubbins et al., 2008; Gonsior et al., 2009), how the composition of CDOM and the possible dissimilar changes for different CDOM components during the irradiation course affect the photo-production of DIC is still not clear. This can be addressed by comparing the photo-production rate of DIC in different types of water with variable CDOM composition, which can be well characterized by the CDOM absorption spectral slope and

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the combination of fluorescence excitation emission matrix spectra and parallel factor analysis (EEMs-PARAFAC). The CDOM absorption spectral slope is correlated negatively with the molecular weight and aromaticity of dissolved organic matter (DOM), while EEMs-PARAFAC is a method for identifying different fluorescent components (e.g., humic-like and protein-like components) of DOM and to study the variation of DOM source and dynamics (e.g., Blough and Del Vecchio, 2002; Stedmon and Markager, 2005; Yamashita et al., 2008; Kowalczyk et al., 2009; Zhuo et al., 2010; Guo et al., 2010, 2011; Yang et al., 2012).

This study investigated the photo-degradation of CDOM and the photo-production of DIC in two typical Chinese coastal waters using absorption spectroscopy, EEMs-PARAFAC and DIC measurement. One sample was collected from the Jiulong River Estuary, which was mainly influenced by allochthonous terrestrial DOM inputs. Another was a coastal seawater sample from the Taiwan Strait, which was more derived from autochthonous DOM source by *in-situ* production. In particular, both samples in our study were collected in the subtropical area and photo-degraded during the summer time. The strong natural solar radiation of the area in this period facilitates the photo-production of DIC from CDOM. To the best of our knowledge, there is no previous report about the photo-production of DIC in Chinese coastal waters. Considering that there are many large estuaries and shelf waters in China (such as the Changjiang River Estuary, the East China Sea and the much area of the South China Sea), our study would help to understand the role of photochemical processes in the carbon transformation in Chinese coastal waters, where marine carbon cycles have become a research focus in recent years (e.g., Dai et al., 2004; Song, 2010).

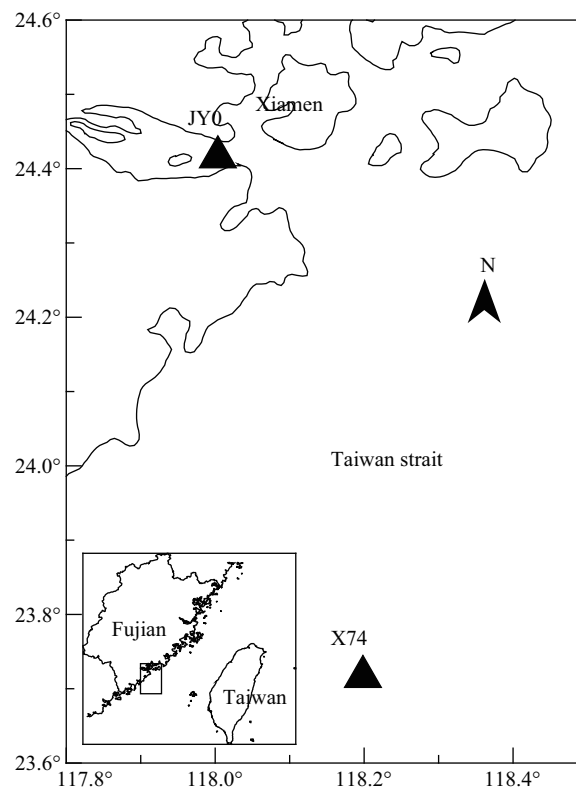
## 1 Materials and methods

### 1.1 Study area

The Jiulong River Estuary is a subtropical estuary in Fujian Province, southeastern China, with a length of 60 km and an average width of 6.5 km (Guo et al., 2011). CDOM in this estuary is derived mainly from the freshwater discharge of Jiulong River which has a drainage area of 14,741 km<sup>2</sup> and an annual runoff of  $1.23 \times 10^{10}$  m<sup>3</sup> (Guo et al., 2007). The Taiwan Strait, located between Taiwan Island and southeast Mainland China, is 180 km wide and 350 km long with an average depth of 60 m.

### 1.2 Sample collection and pretreatment

Surface water samples were collected in 2009 from the lower Jiulong River Estuary on June 29th and from the Taiwan Strait on July 1st (Fig. 1). Subsamples were taken using a set of cleaned vessel and immediately filtered sequentially through GF/F filters (pre-combusted at 500°C for 5 hr) and acid-rinsed 0.2 μm Millipore polycarbonate filters, and then stored in cold (4°C) and in the dark before the irradiation experiments. Salinity was



**Fig. 1** Sampling locations, with surface waters collected from stations JY0 in the Jiulong Estuary and X74 in the Taiwan Strait; the water depth was 13 m at JY0 and 43 m at X74. The insert shows a larger map of the study area.

measured *in-situ* with a SBE917 Plus self-contained CTD (conductivity-temperature-depth) profiling system (Sea-Bird Electronics Inc., USA).

### 1.3 Irradiation experiments

Samples were warmed up to room temperature before irradiation experiments within 4 days after filtration. For JY0 estuarine sample, filtrates were acidified to pH < 3, sparged with air for 1 hr to remove DIC and adjusted to the initial pH value by the addition of Na<sub>2</sub>B<sub>4</sub>O<sub>7</sub>·10H<sub>2</sub>O (ACS level, Sigma Corporation, USA). Then, the sample was transferred pneumatically to 15 cleaned quartz tubes with glass stoppers (125 mL, precombusted at 500°C for 5 hr). Three tubes of the sample were measured at the beginning of the experiment. Six of the remaining 12 tubes were wrapped in aluminum foils as the dark group for comparing with the irradiated group. Thereafter, the 12 tubes were placed in a shallow water tank on board under natural sunlight. After 1-day and 3-day irradiations, samples were taken for optical and DIC measurements in triplicate.

The irradiation experiment procedure for X74 seawater sample was similar to that for JY0 sample, except that the air-sparging time was increased to 2 hr to remove DIC and that the water tank was filled with surface seawater and placed on the rooftop of Ocean Building, Xiamen University, China. Since the irradiation and the seawater filling the tank were similar in the two irradiation experiments for both samples, the results for the two types of water were comparable.

## 1.4 Absorption, fluorescence and DIC measurements

CDOM absorption spectra were scanned using a UV-Vis spectrophotometer (UV2300, Techcomp, China) over wavelengths of 240–800 nm (every 1 nm) with Milli-Q water as the blank (Guo et al., 2007). Absorbance at each wavelength ( $A$ ) was subtracted by the mean absorbance from 700–800 nm for baseline correction.  $A$  was converted to absorption coefficient ( $a$ ) as  $a_\lambda = 2.303 A_\lambda/l$ , where  $\lambda$  is the wavelength and the  $l$  is the path length (0.1 m). The absorption coefficient at 280 nm ( $a_{280}$ ) is presented in this study as an indicator for CDOM abundance. The absorption spectral slope over 270–350 nm ( $S_{270-350}$ ) calculated by non-linear regression method is used to indicate the relative molecular weight of CDOM (Wang and Guo, 2010). The mean analytical precision was 3.5% for  $a_{280}$  and 2.2% for  $S_{270-350}$ .

Fluorescence excitation emission matrix spectra (EEMs) were measured using a fluorescence spectrophotometer (Cary Eclipse, Varian, Australia) with excitation and emission wavelengths of 250–450 nm (every 5 nm) and 300–600 nm (every 2 nm) (Guo et al., 2011; Yang et al., 2012). Sample EEMs were Raman calibrated and subtracted by Milli-Q water blank scanned on the same day. Samples were not corrected for the inner filter effect because of low CDOM absorptions. Then, EEMs were modeled using PARAFAC in MATLAB 7.5 with “the N-way toolbox for MATLAB” (Stedmon and Bro, 2008). PARAFAC decomposed the EEM spectra into individual fluorescent components and the number of fluorescent components was determined with split-half validation. The fluorescence intensity of each component in each sample was represented by its maximum fluorescence  $F_{\max}$  (RU, i.e. Raman units) (Stedmon and Bro, 2008). The mean analytical precision was 2.8% for the fluorescence intensity of all components.

For DIC measurements, samples were acidified with 10%  $H_3PO_4$  to convert DIC to  $CO_2$ , the latter being carried by  $N_2$  to a  $CO_2/H_2O$  analyzer (LI-7000, LI-COR, USA, Cai et al., 2004; Zhai et al., 2005). Seawater DIC standards provided by A. Dickson from the Scripps Institution of Oceanography, USA and an 100  $\mu\text{mol/L}$   $NaHCO_3$  solution with various injection volumes were used for calibration. The mean standard deviation of the DIC measurement was  $\pm 1.1 \mu\text{mol/L}$ .

## 2 Results and discussion

### 2.1 Initial optical characteristics

Salinity increased from 29 for the JY0 estuarine sample to 33 for the X74 seawater sample, while the CDOM absorption coefficient ( $a_{280}$ ) decreased from 4.2 to  $1.3 \text{ m}^{-1}$ . This agreed with a negative relationship between CDOM abundance and salinity in the Jiulong River Estuary and its adjacent coastal waters (Guo et al., 2007, 2011). The  $S_{270-350}$ , which correlates negatively with the molecular weight and aromaticity of CDOM, increased from  $0.0163 \text{ nm}^{-1}$  for the JY0 estuarine sample to  $0.0294 \text{ nm}^{-1}$  for the X74 seawater sample. This suggested a decrease in the molecular weight and aromaticity of CDOM with increasing salinity, similar to the results of previous studies (e.g., Blough and Del Vecchio, 2002; Guo et al., 2007).

Five fluorescent components (C1–C5) were identified using EEMs-PARAFAC, including four humic-like components and one protein-like component (i.e., the tryptophan-like C3) (Fig. 2). C1 had excitation/emission (Ex/Em) maxima at  $\leq 250, 295/378 \text{ nm}$ , similar to the humic-like C5 (Ex/Em maxima: 250, 300/374 nm) in Lapiere and Frenette (2009). The fluorescence intensity of C1 correlated more closely with other humic-like components ( $r = 0.983-0.995$ ) than with the tryptophan-like component ( $r = 0.948$ ) in our study. C2, with Ex/Em maxima at  $\leq 250, 370/458 \text{ nm}$ , resembled the humic-like C4 (Ex/Em maxima:  $< 250, 360/440 \text{ nm}$ ) introduced by Stedmon and Markager (2005). C3 had Ex/Em maxima at 275/332 nm, identical to the tryptophan-like C4 described by Guo et al. (2011) which has Ex/Em maxima at 275/328 nm. C4, with Ex/Em maxima at 335/420 nm, was similar to the humic-like C2 (Ex/Em maxima: 345/433 nm) in the study of Yamashita et al. (2008). C5 had Ex/Em maxima at 270, 400/492 nm, similar to the terrestrial humic-like C4 (Ex/Em maxima: 270, 390/508 nm) reported by Kowalczyk et al. (2009).

The JY0 estuarine sample had higher fluorescence intensity for all fluorescent components than the X74 seawater sample (Fig. 3), similar to previous studies (e.g., Guo et al., 2011). Furthermore, fluorescence intensities of the four humic-like components for the JY0 sample were 4.5–6.9 times those for the X74 sample, while that of the tryptophan-like C3 for the JY0 sample was only 1.7 times that for the X74 seawater sample. In addition, the humic-like C1 and C2 were the two components which

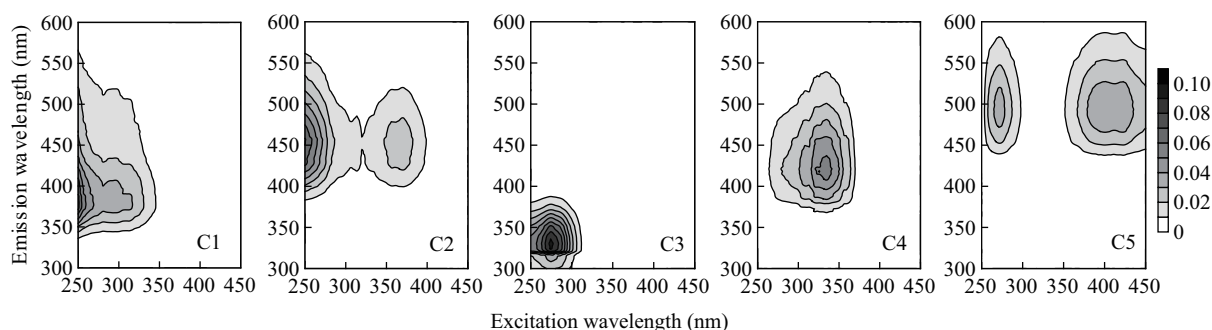
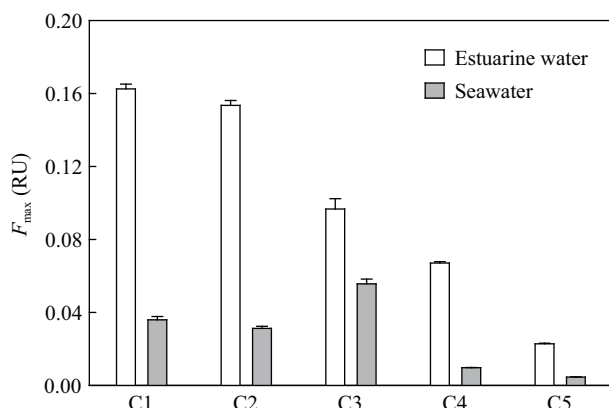


Fig. 2 EEM contour plots of the five fluorescent components identified using PARAFAC.



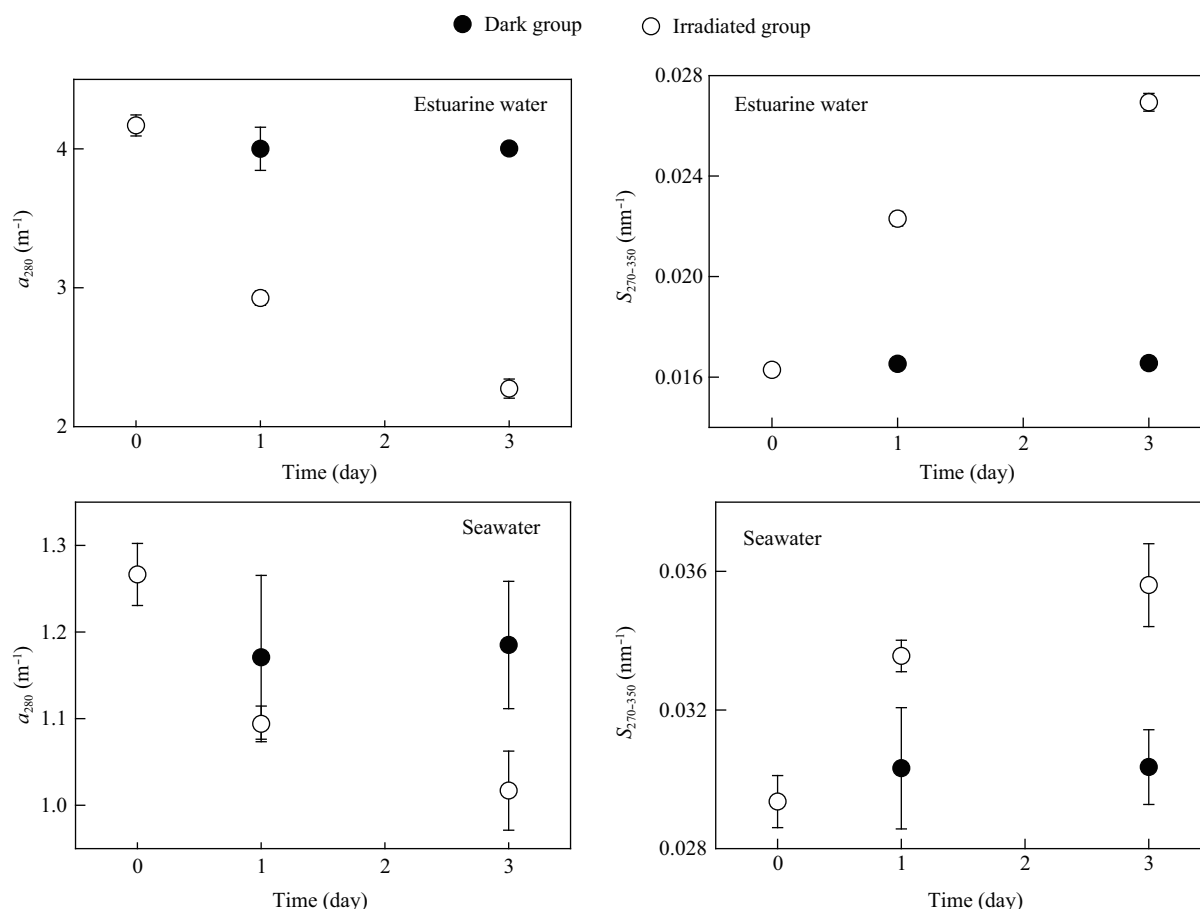
**Fig. 3** Initial fluorescence intensities ( $F_{\max}$ ) of the five components in the JY0 estuarine and the X74 seawater samples.

had highest fluorescent intensities in the JY0 estuarine sample, while the tryptophan-like C3 was the most abundant component in the X74 seawater sample. This was probably because DOM in the estuarine water was mainly from the river discharge which contained abundant humic-like materials, while DOM in the coastal ocean received additions from autochthonous production. This also agreed with previous studies showing an increasing proportion of protein-like components with increasing salinity in estuarine and coastal waters (Stedmon and Markager, 2005; Kowalczyk et al., 2009; Guo et al., 2011).

## 2.2 Photo-degradation of CDOM

For the JY0 estuarine sample,  $a_{280}$  and  $S_{270-350}$  changed little in the dark group (Fig. 4), and both varied by < 4.1% throughout the experiment course. This suggests that the aluminum foils covering the tubes prevented the filtrates from irradiation and there was no significant addition or removal process for CDOM in the dark group. In contrast,  $a_{280}$  and  $S_{270-350}$  both changed notably in the irradiated group. After three day's irradiation,  $a_{280}$  decreased by 45% from 4.2 to 2.3  $\text{m}^{-1}$ , indicating that photo-degradation removed CDOM effectively. The  $S_{270-350}$  value increased by 65% from 0.0163 to 0.0269  $\text{nm}^{-1}$  after the same irradiation, suggesting a decrease in the molecular weight and aromaticity of DOM by photo-degradation due to bond cleavage and/or disaggregation (Blough and Del Vecchio, 2002; Helms et al., 2008; Wang and Guo, 2010).

In the X74 seawater dark group,  $S_{270-350}$  changed little (< 3.4%), similar to that in the estuarine dark group, while  $a_{280}$  decreased by 7.6% from 1.3 to 1.2  $\text{m}^{-1}$  after 1-day irradiation and then remained constant. The variance of  $a_{280}$  in the seawater dark group might be mainly due to the analytical deviation for these CDOM-poor samples which reached as high as 6.2%–8.1%, much higher than those for other samples (1.0%–3.9%, Fig. 4). Removal processes including photo-degradation, adsorption to particles and microbial degradation were probably insignificant for CDOM in the seawater dark group, since aluminum



**Fig. 4** Temporal changes in the CDOM absorption coefficient ( $a_{280}$ ) and spectral slope ( $S_{270-350}$ ) in the JY0 estuarine and the X74 seawater samples in irradiation experiments.

foils covering the tubes prevented CDOM from photo-degradation while filtration with 0.2  $\mu\text{m}$  filters before the experiment removed most particles and bacteria from the sample. In contrast, both the abundance and the composition of CDOM changed notably in the X74 seawater irradiated group. There were 20% loss in  $a_{280}$  (from 1.3 to 1.0  $\text{m}^{-1}$ ) and a 21% increase in  $S_{270-350}$  (from 0.0293 to 0.0356  $\text{nm}^{-1}$ ) after the 3-day irradiation (Fig. 4). These changes in the seawater sample had characteristics similar to the estuarine water, but to smaller extents. Possible explanations for the weaker photo-degradation of CDOM in seawater were that the seawater had a weaker light absorption and that the CDOM in seawater had a lower molecular weight and aromaticity with more constituents less susceptible to photo-degradation. For example, Stubbins et al. (2008) find a strong dependence of CDOM photo-degradation upon the aromaticity of humic substance, suggesting that aromatic moieties are the principal chromophores and photo-reactants. In addition, Gonsior et al. (2009) find that the aromaticity and the photo-reactivity of CDOM are higher in a Cape Fear River sample than in a Cape Fear Estuary sample. Combining the results from previous studies and our study, the CDOM photo-degradation is dependent on its initial concentration and composition from freshwater to the seawater.

### 2.3 Photo-degradation of fluorescent components

For the JY0 estuarine sample, fluorescence intensities of C1, C2 and C4 changed little (0.20%–2.2%) after 3 days in the dark group, while that of C5 increased a little by

8.2% from 0.023 to 0.025 RU and that of tryptophan-like C3 decreased by 16% from 0.097 to 0.081 RU. In contrast, fluorescence intensities of the four humic-like components decreased largely by 56%–78% in the irradiated group, and that of protein-like C3 decreased by 32% (Fig. 5). These results indicated an effective removal of fluorescent components in estuarine waters by photo-degradation, similar to those in rainwater (Kieber et al., 2007) and the Baltic Sea (Stedmon et al., 2007). Our results also suggested that some DOM constituents such as the humic-like components in our study could be preferentially photo-degraded, which agreed with the changes in the absorption spectral slope as mentioned above. In particular, the humic-like C4 and C5 showed highest photo-degradation rates (78% and 69%, respectively) among the five components, suggesting that they were probably the most sensitive components to photochemical degradation. In contrast, the photochemical removal of the tryptophan-like C3 was much less effective (only by 16% if the decrease in the dark group was subtracted from that in the irradiated group). Similarly, humic-like fluorophores are removed more rapidly than protein-like fluorophores and the bulk CDOM in rainwater (Kieber et al., 2007). For comparison, previous studies find that microbial degradation remove protein-like components much more effectively than humic-like components (Lønborg et al., 2010). Therefore, the two removal processes of DOM, photo-degradation and microbial degradation, can be differentiated by their preferential removal of different DOM components in aquatic environments.

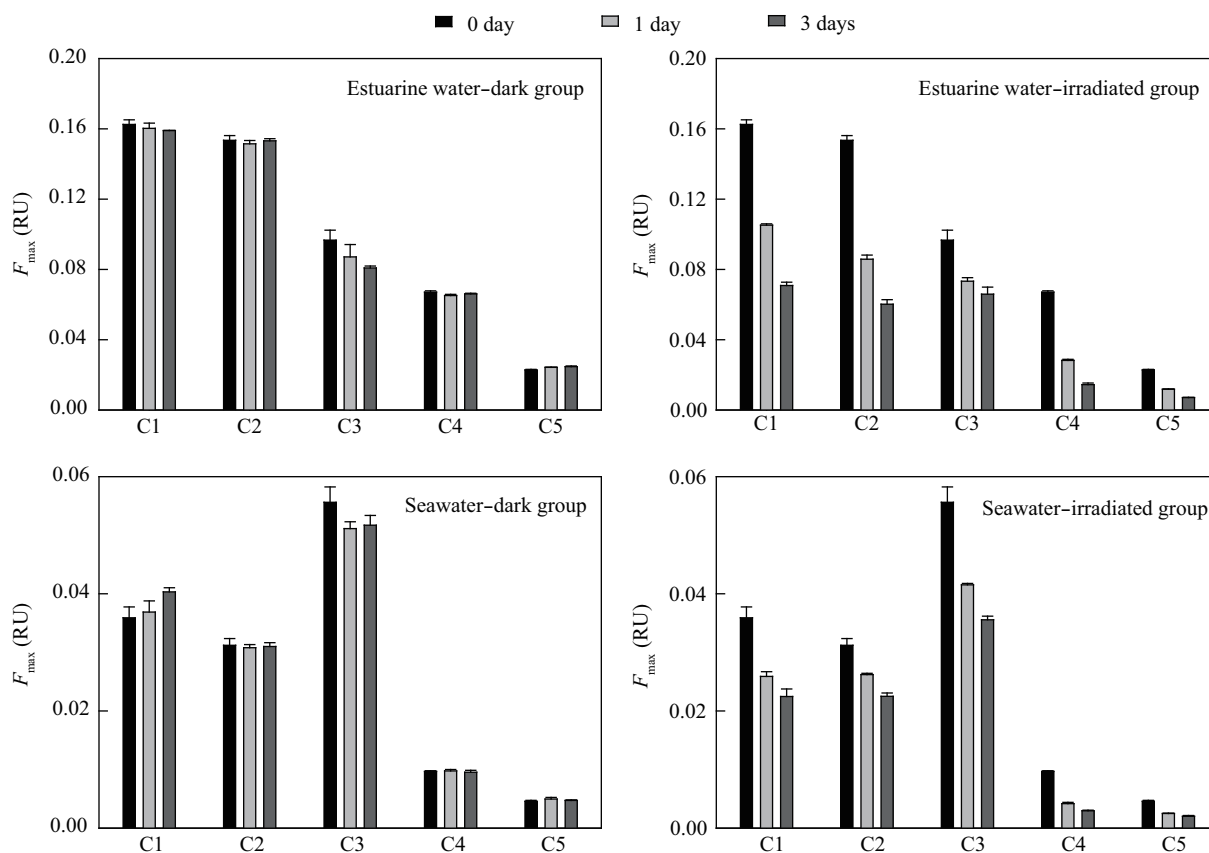
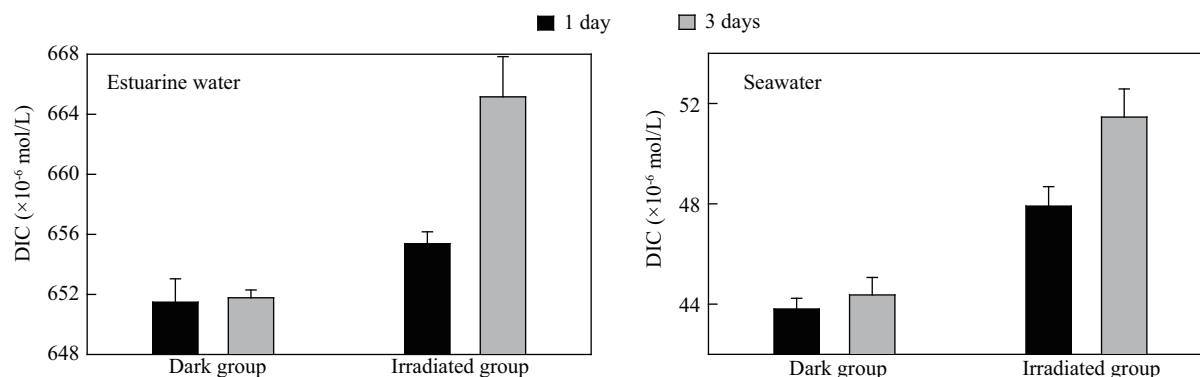


Fig. 5 Changes in fluorescence intensities ( $F_{\text{max}}$ ) of the five components with irradiation time in the JY0 estuarine and the X74 seawater samples.





**Fig. 6** Concentration of DIC after 1-day and 3-day irradiations in the JY0 estuarine and the X74 seawater samples.

For the X74 seawater sample, fluorescence intensities of C2, C4 and C5 showed insignificant changes (by < 3.7%) in the dark group, while that of C1 increased by 12% and that of C3 decreased by 7.1% after 3 days. In contrast, all the fluorescent components were removed effectively (by 28%–69%) in the irradiated group. C4 and C5, whose fluorescence intensities decreased by 69% and 56%, were the two components most susceptible to photo-degradation in the X74 sample, similar to the case in the JY0 estuarine sample (Fig. 5). Furthermore, all the four humic-like components were photo-chemically removed less in the X74 sample than in the JY0 estuarine sample, in agreement with the results of the loss in  $a_{280}$ . In addition, the removal of C3 in the irradiated seawater (36%) was 5 times that in the dark, indicating the ability of photo-degradation to remove the tryptophan-like DOM (although to a smaller extent than such humic-like components as C4 and C5).

#### 2.4 Photo-production of DIC and its relationship to the photo-degradation of CDOM

For the JY0 estuarine sample, the DIC concentration changed little in the dark group, with values of 651.5  $\mu\text{mol/L}$  after 1 day and 651.8  $\mu\text{mol/L}$  after 3 days. In contrast, it increased by 9.8  $\mu\text{mol/L}$  from 655.4  $\mu\text{mol/L}$  after 1 day to 665.2  $\mu\text{mol/L}$  after 3 days in the irradiated group (Fig. 6). Similarly, the DIC concentration in the X74 seawater sample showed a limited increase in the dark group (43.8  $\mu\text{mol/L}$  after 1 day vs. 44.4  $\mu\text{mol/L}$  after 3 days), but increased significantly in the irradiated group from 47.9  $\mu\text{mol/L}$  after 1 day to 51.5  $\mu\text{mol/L}$  after 3 days. In addition, DIC in both types of water showed higher

concentrations in the irradiated group than in the dark group after irradiations. These results demonstrated that DIC was photo-chemically produced.

The photo-production rate of DIC was estimated as the slope of the calibration curve of DIC photo-production amount vs. irradiation time. The DIC photo-production amount at the start was set to be zero and those after 1-day and 3-day irradiations were calculated by subtracting DIC concentrations in the dark group from those in the irradiated group. The DIC-yielding photo-production rate was 4.4  $\mu\text{mol}/(\text{L}\cdot\text{day})$  for the JY0 estuarine sample and 2.5  $\mu\text{mol}/(\text{L}\cdot\text{day})$  for the X74 seawater sample. These values were comparable to those in previous studies (Table 1). They were lower than those in many lakes and rivers (e.g., Granéli et al., 1998), comparable to those in the Delaware Estuary (White et al., 2010) but higher than that in the open ocean (e.g., Gulf Stream, White et al., 2008). In fact, the photo-production rate of DIC varies largely over 3 orders of magnitude higher than that of natural aquatic environments (Table 1). In comparison, the typical euphotic zone primary productivity of the northern South China Sea adjacent to our seawater station has been reported as 0–35  $\text{mg C}/(\text{m}^3\cdot\text{day})$ , i.e., < 2.9  $\mu\text{mol}/(\text{L}\cdot\text{day})$  (Chen and Chen, 2006). Therefore, the DIC photo-production rate in surface seawater sample revealed in this study was comparable to the primary production. This further demonstrates the importance of DIC photo-production in the marine carbon cycle in Chinese coastal waters, especially the subtropical and tropic area (e.g. the Taiwan Strait and the northern South China Sea). The effective photo-production of DIC from CDOM in the Taiwan Strait may be in part due

**Table 1** Photo-production of DIC from bulk DOM in natural waters

Study site	Salinity	Irradiation source	DIC photo-production rate ( $\mu\text{mol}/(\text{L}\cdot\text{day})$ )	Reference
Sweden and Brazil Lakes		Natural sunlight	7.3–141.2 <sup>a</sup>	Granéli et al., 1998
Green Lake		Solar simulator	0.3 <sup>b</sup>	White et al., 2008
Batata Lake (Amazon)		Natural sunlight	0.4–12.4 <sup>a</sup>	Amado et al., 2006
Caraná stream (Amazon)		Natural sunlight	3.0–11.2 <sup>a</sup>	Amado et al., 2006
Amazon River		Natural sunlight	1–16	Remington et al., 2011
Delaware Estuary	0.1–21	Natural sunlight	2–4	White et al., 2010
Coastal water (Rhode Island)	32	Solar simulator	0.2 <sup>b</sup>	White et al., 2008
Gulf Stream	36	Solar simulator	0.05 <sup>b</sup>	White et al., 2008
Jiulong Estuary	29	Natural sunlight	4.4	This study
Taiwan Strait	33	Natural sunlight	2.5	This study

<sup>a</sup> Sunlight exposure is limited to  $\pm 3$  hr around true noon (Granéli et al., 1998; Amado et al., 2006); <sup>b</sup> 8-hr solar simulator light used is roughly equivalent to 1-day summer sunlight at 40°N, in terms of  $\text{CO}_2$  photo-production (White et al., 2008)

to the strong irradiation in subtropical area, the effective light penetration in the oligotrophic seawater of this region and the input of CDOM from both allochthonous and autochthonous sources (although autochthonous production might be dominant).

The irradiation experiments of estuarine water sample clearly showed higher photo-production rate of DIC than that of the seawater sample (i.e. 4.4 vs. 2.5  $\mu\text{mol}/(\text{L}\cdot\text{day})$ ). Possible explanations included differences in the irradiation intensity, the CDOM abundance and its composition. However, the irradiation intensity was similar during the experiment courses for the two types of water. For example, the intensity of UV radiation, which was the most efficient radiation for photo-degradation of DOM, was 4.86 and 4.84  $\text{MJ}/\text{m}^2$  during the experiment for the JY0 estuarine and the X74 seawater samples, respectively. In contrast, the CDOM abundance was much higher in the JY0 sample than in the X74 sample as mentioned above, implying a stronger ability of light absorption and hence photo-degradation of CDOM and photo-production of DIC (Granéli et al., 1998; Remington et al., 2011). This also agreed with the DIC photo-production rate exceeding 100  $\mu\text{mol}/(\text{L}\cdot\text{day})$  for humic-rich lake and river waters (Granéli et al., 1998; Gao and Zepp, 1998) but being as low as 0.3  $\mu\text{mol}/(\text{L}\cdot\text{day})$  in the pristine Green Lake (White et al., 2008). In addition, the CDOM composition was different between the two types of water, as reflected by the lower absorption spectral slope and the higher contribution of humic-like fluorescent components in the JY0 estuarine sample. This would affect the susceptibility of CDOM to photo-degradation (Stubbins et al., 2008; Gonsior et al., 2009). In fact, the loss percentage of CDOM in the JY0 estuarine water sample was 2.3 times that in the X74 seawater sample (45% vs. 20%). If this was considered, the photo-production efficiency of DIC would be lower in JY0 estuarine sample than in X74 seawater sample (i.e., 0.98  $\mu\text{mol}/(\text{L}\cdot\text{day})$  vs. 1.27  $\mu\text{mol}/(\text{L}\cdot\text{day})$  every 10% decrease of  $a_{280}$ ), which could be explained by that the seawater sample contained more algal derived CDOM which has a higher photo-production efficiency of DIC than terrestrial CDOM (Johannessen et al., 2007). The photochemical removal fraction was also higher in the humic-rich JY0 estuarine water sample than in the X74 seawater sample for all the four humic-like fluorescent components but not for the tryptophan-like component. Furthermore, the differences in the CDOM composition might also affect the photo-degradation pathway of CDOM (i.e., the pathway in which the carbon bound in CDOM is degraded to DIC or that in which CDOM is transformed to non-chromophoric organic matter and the carbon is not released). However, the importance of this effect for the photo-production of DIC needs further study.

### 3 Conclusions

This study revealed notable differences between estuarine water and coastal seawater types in China coastal areas, in terms of the CDOM concentration, composition and photo-degradation dynamics, as well as the photo-production of

DIC. The estuarine water had a higher CDOM absorption coefficient but a lower absorption spectral slope (and hence probably a higher molecular weight and aromaticity) than the seawater. In addition, CDOM in the estuarine water was dominated by humic-like fluorescent components, while that in the seawater by a tryptophan-like component. The photo-degradation of CDOM was notable for both types of water, with an increase in the absorption spectral slope and a preferential removal of two humic-like components (C4 and C5). Furthermore, the photo-degradation of CDOM was stronger in the estuarine water than in the seawater after a 3-day irradiation, probably due to their different CDOM abundance and composition. This was also likely the reason for a higher photo-production rate of DIC in the estuarine water than in the seawater (4.4 vs. 2.5  $\mu\text{mol}/(\text{L}\cdot\text{day})$ ). In conclusion, photochemical reactions played an important role in removing and transforming CDOM and in the DIC production in Chinese coastal waters and hence will significantly influence the carbon cycle processes in this important marginal sea area.

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