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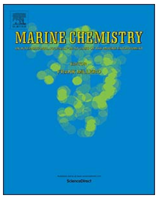

Diurnal variations of dissolved organic matter in the hydrothermal system of Green Island, Taiwan

Hao Zheng[a], Changdong Xu[a], Liyang Yang[b], Jianfang Chen[c], Chen-Tung Arthur Chen[a,d,*], Bing-Jye Wang[d]

[a] Ocean College, Zhejiang University, Zhoushan 316021, PR China
[b] College of Environment and Resources, Fuzhou University, Fuzhou, Fujian, PR China
[c] Key Lab of Marine Ecosystem and Biogeochemistry, Second Institute of Oceanography of SOA, Hangzhou 310012, PR China
[d] Department of Oceanography, National Sun Yat-Sen University, Kaohsiung 804, Taiwan, ROC

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ABSTRACT

Hydrothermal systems are increasingly recognized as important fields for the biogeochemical cycling of dissolved organic matter (DOM) in the ocean. While the quantity and quality of DOM have been characterized for various hydrothermal systems, little is known about the temporal variations of DOM and the underlying processes in these systems. Diurnal variations of DOM were investigated for the hot water area and the warm water area of the hydrothermal system in Green Island off southeast Taiwan at 3 h intervals, using measurement of dissolved organic carbon (DOC), absorption spectroscopy, and fluorescence excitation emission matrix-parallel factor analysis (EEM-PARAFAC). The results of temperature, salinity, pH and DOC indicated that the warm water area had a third freshwater source, besides the seawater and the hot fluids from the hot water area, that was probably groundwater. The DOC concentration showed significant diurnal changes within 84–130 μM at the warm water area. A negative correlation between the DOC concentration and temperature was observed at the warm water area, while the DOC concentration at the hot water area was largely independent of temperature and tide. Four fluorescent components were identified by PARAFAC, including tryptophan-like C1 and C4, and humic-like C2 and C3. The fluorescence intensities of C1 and C4 were higher at the hot and the warm water areas than those in the ambient seawater, suggesting that the former two areas were sources of fluorescent DOM for seawater. The fluorescence intensities of C1 and C2 at the hot water area were higher at night than those in the daytime, implying the effects of potential photobleaching. Overall, the low values of the humification index (HIX) and the high values of the fluorescence index (FI) and the autochthonous index (BIX) at the hot and the warm water areas revealed that the DOM might have a mainly autochthonous biological or bacterial origin.

1. Introduction

Dissolved organic matter (DOM) is one of the most abundant forms of organic matter and a critical component in the global carbon cycle (Jaffé et al., 2014; Ogawa and Tanoue, 2003). It has been extensively investigated in the aquatic environment because this active material has a significant role in many important ecological and environmental processes, which directly affect the carbon cycle (Hedges, 1992). DOM can be used as a light attenuator, a metabolic substrate for heterotrophic bacteria, a strong chelating agent for metals in aquatic systems, and a significant sink of atmospheric carbon dioxide against global warming (Jaffé et al., 2014). The variable structure and composition as well as the source, sink and migration process of DOM are key to understanding the dynamics of various biogeochemical processes in the global carbon cycle (Abdulla and Hatcher, 2014). Hydrothermal systems are extreme environments with unique biogeochemical cycle and ecological processes. DOM from hydrothermal systems may affect the speciation and toxicity of metals and the carbon cycle of the ocean. Although several studies have focused on associated gases and heavy metals (Chen et al., 2005; Chen et al., 2016b; Herzig and Hannington, 2006; Sano et al., 2008; Wu et al., 2009; Zeng et al., 2012), very little is known about DOM in hydrothermal systems (Mccarthy et al., 2010; Chen, 2011; Gomez-Saez et al., 2015; Hawkes et al., 2016; Yang et al., 2012, 2017).

Recently, DOM has been investigated using absorption spectroscopy and fluorescence excitation emission matrices-parallel factor analysis (EEM-PARAFAC) in submarine hydrothermal vents off the Kueishantao islet, NE Taiwan. The results indicated that the white vent was a source

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[*] Corresponding author at: Ocean College, Zhejiang University, Zhoushan 316021, PR China.
E-mail address: ctchen@mail.nsysu.edu.tw (C.-T.A. Chen).

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of chromophoric DOM (CDOM) for seawater and that the characteristics of DOM from the hydrothermal vents were similar to that from terrestrial caves and spring waters (Yang et al., 2012). In the Juan de Fuca Ridge, the DOC in ridge-flank fluids was synthesized by chemosynthetic crustal microbial communities from very old dissolved inorganic carbon, with radiocarbon ages of 11,800–14,400 years, suggesting that ridge-flank circulation may be a source of substantial fixed carbon for the overlying ocean (Mccarthy et al., 2010). The molecular alteration of marine DOM under experimental hydrothermal conditions revealed that the vast majority of DOM cannot survive in hydrothermal circulation at high temperature ($>$ 300 °C),low pH and a high concentration of sulfide, and only some species with a low molecular weight, heteroatom (N, S, P) and low oxygen content may survive at temperatures of 100–200 °C (Hawkes et al., 2016). The DOC concentration (27 μM) in fluids of the shallow hydrothermal vent system in the Caribbean Sea was lower than that in surface seawater (99 μM), owing to the effect of meteoric water with a low DOC concentration or the depletion of DOC during circulation through the sediment (Gomez-Saez et al., 2015).

The fluorescence EEM spectroscopy, which is both simple and fast, has been applied for characterizing the level and chemical composition of DOM, and for tracking the source and transformation of DOM. EEM provides highly detailed fingerprint spectral information from which fluorescent peaks can be identified (Coble, 1996). Since EEM was used to trace the evolution of DOM in the Black Sea (Coble et al., 1990), it has been widely used to identify fluorophores in seas, rivers and lakes (Coble, 1996; McKnight et al., 2001; Patel-Sorrentino et al., 2002). However, not all of the fluorescence peaks can be identified visually with the "peak-picking" method owing to the complexity of DOM in seawater and the overlapping of peaks in EEM (Coble, 1996). PARAFAC modeling is a three-way method that originates in psychometrics and can enhance the characterization of fluorescent DOM (FDOM). Additional information can be gained by decomposing the obtained fluorescence EEMs into several independent fluorescent components (Stedmon et al., 2003; Jaffé et al., 2014). Stedmon et al. (2003) were the first to combine EEM with PARAFAC modeling. EEM-PARAFAC has been widely applied in different aquatic environments to identify various fluorescent components and reveal their dynamic processes (Bai et al., 2013; Yamashita and Jaffé, 2008; Yang et al., 2015b; Zhou et al., 2016a).

Green Island (22.63°–22.70°N, 121.45°–121.55°E) is an extinct volcanic island in the Pacific Ocean that is about 33 km off the east coast of Taitung, Taiwan (Yang et al., 2011). The island has numerous hills with the presence of crater hills, in which the rocks are conglomerate and andesite. A hydrothermal system, called Chao-jih Hot Springs, covers two typical areas that include the hot water area and the warm water area in the intertidal zone (Fig. 1). The distance between the hot water area and the warm water area is approximately 82 m. The temperature at the hot water area is around 90 °C and that at the warm water area is around 50 °C. A recent study revealed low DOC concentration and distinct FDOM composition at the hot water area, which was attributed to potential thermal degradation at high temperature (Yang et al., 2017). However, the quantity and quality of DOM at the warm water area and the diurnal variations of DOM in both areas have not been reported previously.

DOM concentrations in seas and rivers vary annually and interannually (Clark et al., 2016; Leech et al., 2016; Lin et al., 2012; Shi et al., 2016). However, few studies have focused on variations of DOM concentrations over shorter time scales such as the diurnal cycle (Chen et al., 2016a; Pokrovsky and Shirokova, 2013; Spencer et al., 2007). No study has investigated the impact of diurnal variability on DOM dynamics at a hydrothermal system using EEM-PARAFAC. Thus, the specific objectives of this study are as follows: (1) to identify the source and diurnal changes of DOM at the hot and warm water areas in Green Island, and (2) to determine the concentration and composition of CDOM at the hot and warm water areas using absorption spectroscopy and EEM-PARAFAC.

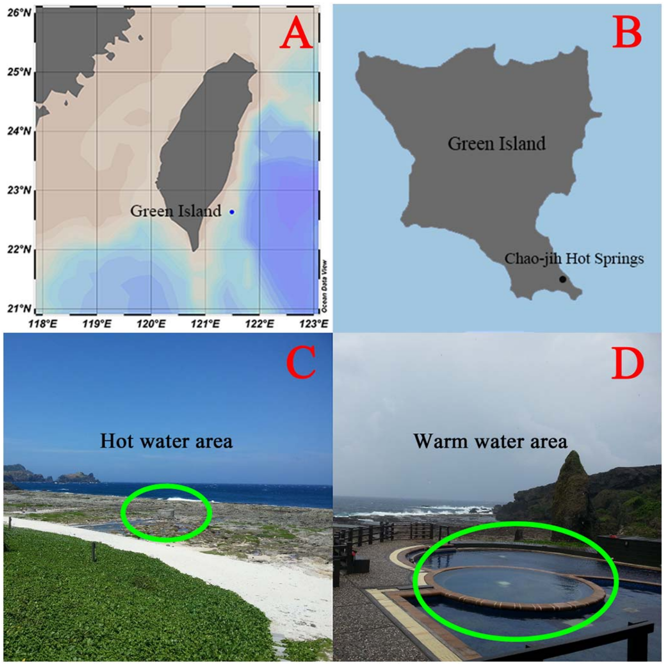

Fig. 1. The hot water area and the warm water area at Chao-jih Hot Springs on Green Island, Taiwan.

2. Materials and methods

2.1. Sample collection and pretreatment

Samples were collected on September 18th–23rd, 2014. Typhoon Fung-wong hit Green Island on September 19th–21st, so samples were not collected on these three days. Most samples were collected after Typhoon Fung-wong on September 22nd–23rd. At both the hot and the warm water areas, samples were taken at 3 h intervals over a diurnal period. The hot water area was sampled at its surface while samples at the warm water area were collected at the underwater outlet. Samples were filtered through acid-rinsed 0.2 μm Millipore polycarbonate filters and then stored in 60 mL brown glass bottles at 4 °C before use. Samples with added saturated mercuric chloride were used to measure DOC and those without were used to measure CDOM. Before they were used, the brown bottles were washed with Milli-Q water and then placed in a muffle furnace at 450 °C for 5 h to remove any organic matter.

The temperature of the hot spring was measured in situ using a thermocouple. Salinity and pH values were determined in the laboratory with a Guildline salinometer (Autosal 8400B) and a Radiometer PHM-85 pH meter respectively.

2.2. DOC measurements

The DOC concentrations were measured using a Total Organic Carbon Analyzer (Shimadzu, TOC-LCSH) and the method of high-temperature catalytic oxidation (~680 °C). Samples were acidified with excessive amounts of hydrochloric acid and purged with nitrogen to remove inorganic carbon before the DOC concentrations were determined. The DOC measurement was made at least twice for each sample, with a coefficient of variation $<$ 2%. The calibration curve was determined using five solutions of potassium hydrogen phthalate as standards.

2.3. Absorption measurements

A Cary 50 UV–vis spectrometer was used to obtain the absorption

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