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Hydrogen peroxide in tropical shelf Waters: The Northern South China Sea Shelf



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ABSTRACT

The distribution of hydrogen peroxide was determined at 28 stations in four transects across the Northern South China Sea Shelf-sea (NoSoCS) in June 2010. The average concentration in the top 10 m ranged between 0.063 and 0.231 μM . Among the hydrographic regimes: the inner shelf outside the influence of upwelling, the inner shelf under the influence of upwelling, the middle shelf, the outer shelf and the open South China Sea, the highest concentration was found in the day-time in the inner shelf outside of the influence of upwelling and the lowest concentration was found in the night-time in the inner shelf under the influence of upwelling. The average concentration at the day-time stations in each hydrographic regime was invariably higher than the corresponding concentration at the night-time stations. Across the shelf, the concentration was the highest in the inner shelf. The variations in the concentrations of hydrogen peroxide and total organic carbon (TOC) in the different hydrographic regimes followed a similar pattern. All these behaviors are consistent with the photochemical production of hydrogen peroxide, using dissolved organic matter as the chromophore, as a primary control on its occurrence in the surface waters. Moreover, at day-time stations at different locations in the NoSoCS and at a single location sampled through the day, the relationship between the concentration of hydrogen peroxide and the product of the concentration of TOC and the time-integrated irradiance at the time of sample collection fell on two distinct linear lines with different slopes and intercepts: one for mostly the inner and middle shelf waters and the other for mostly the water in the open northern SCS. These relationships suggest that the light history, the pre-existing concentration of hydrogen peroxide, and the concentration and the photochemical efficiency of the dissolved organic matter in the production of hydrogen peroxide play major roles in determining the temporal and spatial variations in the concentration of hydrogen peroxide in the surface waters in the day-time. In the sub-surface, a notably large fraction, about one-third, of the profiles of hydrogen peroxide did not follow the typical distribution of a quasi-exponential decrease in concentration with depth. Almost all the profiles at the stations in the slope where the internal waves were most active were atypical. At these stations, the concentration of hydrogen peroxide was generally elevated down to about 100 m, well beyond the depths where solar irradiance was plentiful. Superimposed on these elevated concentrations was a persistent sub-surface maximum, with concentrations reaching about 0.5 μM , at 80 m. While the origin of these atypical distributions is not yet clear, dark biological production and the action of internal waves conceivably could have contributed to their occurrence.

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1. Introduction

Hydrogen peroxide is a reactive transient that is present rather ubiquitously in the surface oceans at concentrations of 10^{-2} –

10^{-1} μM (Zika et al., 1985a, 1985b; Johnson et al., 1989; Moore et al., 1993; Miller and Kester, 1994; Sarthou et al., 1997; Avery et al., 2005; Yuan and Shiller, 2001, 2005; Steigenberger and Croot, 2008). It can act both as an oxidizing and a reducing agent (Zafriou, 1983; Zafriou et al., 1984; Moffett and Zafriou, 1990) and can react with a fairly large suite of biologically important and redox-sensitive trace elements in the oceans such as iron (González-Davila et al., 2005; Millero and Sotolongo, 1989; Moffett and Zika, 1987; Santana-Casiano et al., 2006), copper (Millero et al., 1991; Sharma and Millero, 1989), chromium

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(Pettine and Millero, 1990; Pettine et al., 1991), arsenic (Pettine et al., 1999; Pettine and Millero, 2000), manganese (Sunda and Huntsman, 1994), and iodine (Wong and Zhang, 2008). The concentration of hydrogen peroxide is high enough that it may then play a role in controlling the redox speciation of these trace elements and thus affect their bioavailability or toxicity, which may in turn affect the functioning of the ecosystem as a whole (Wong et al., 2003; Zhang and Wong, 1999).

Hydrogen peroxide is formed primarily through indirect photochemical reactions in which dissolved organic matter acts as the chromophore. As these organic molecules capture the light energy, they become excited. Subsequently, the excited molecules react with oxygen to form superoxide which disproportionates to form hydrogen peroxide (Cooper and Zika, 1983; Cooper et al., 1994; Scully et al., 1996). The efficiency of the reaction depends on the composition of the organic matter, and, the intensity and the spectral composition of the incident light since ultraviolet light is more effective than visible light in inducing the formation of hydrogen peroxide (Scully et al., 1996; Wong and Wong, 2001; O'Sullivan et al., 2005). This photochemical production of hydrogen peroxide is rapid and can be readily detected in minutes to hours when samples of natural water are exposed to solar irradiation (Cooper and Zika, 1983; Cooper et al., 1988; Yocis et al., 2000; Avery et al., 2005; Yuan and Shiller, 2005; Clark et al., 2009). Traditionally, biological production (Palenik et al., 1987; Palenik and Morel, 1988; Yuan and Shiller, 2004) and atmospheric deposition (Zika et al., 1982; Cooper et al., 1987; Hanson et al., 2001; Kieber et al., 2001) have been considered minor sources of hydrogen peroxide to the oceans. However, recent studies suggest that the biological production of hydrogen peroxide may be more widespread and its contribution to the total production of hydrogen peroxide in the aquatic environment may be more significant than previously thought (Kustka et al., 2005; Rose et al., 2005, 2008, 2010; Vermilyea et al., 2010; Dixon et al., 2013; Diaz et al., 2013; Shaked and Rose, 2013). On the other hand, the primary sink of hydrogen peroxide in the oceans is its biologically mediated decomposition (Moffett and Zafiriou, 1990; Petasne and Zika, 1997). The efficiency in the decomposition is species-dependent. Thus, among the marine phytoplankton studied, *Synechococcus* sp. and *Synechococcus costatum* were found to be the most efficient and *Pleurochrysis carterae* and *Dunaliella tertiolecta* were the least efficient decomposers (Wong et al., 2003). Other minor sinks include chemical auto-decomposition (Szymczak and Waite, 1988; Cooper et al., 1994) and photochemical decomposition (Moffett and Zafiriou, 1990). The life-times of hydrogen peroxide in seawater in the dark range between hours to

days (Cooper et al., 1994; Petasne and Zika, 1997; Yuan and Shiller, 2001). Given its relatively short response times in its photochemical production and biological decomposition, the occurrence of hydrogen peroxide in the oceans represents primarily a kinetic balance between them. As a result of the separation of these two processes with depth, the concentration of hydrogen peroxide typically decreases quasi-exponentially with depth (Zika et al., 1985a, 1985b; Johnson et al., 1989). Yuan and Shiller (2004) recently reported that low concentrations, in 10^{-3} μM , of hydrogen peroxide may be found in deep water probably as a result of dark biological production.

The primary environmental conditions that affect the photochemical production and biological decomposition of hydrogen peroxide are: the concentration of dissolved organic matter, the solar irradiance and solar spectrum in its production, and, the level of biological activity in its decomposition. Relative to the oligotrophic ocean, the biological activities and the concentration of dissolved organic matter are both elevated in the shelf-seas. Moreover, total solar irradiance and the contribution of ultraviolet to the total irradiance tend to increase with decreasing latitude. These environmental characteristics intersect in the tropical shelf-seas which may provide a unique sub-environment for detecting how the interactions among these environmental conditions may affect the occurrence of hydrogen peroxide in the oceans. Past studies on the occurrence of hydrogen peroxide in the oceans tended to focus on the oligotrophic open oceans (Johnson et al., 1989; Miller and Kester, 1994; Sarthou et al., 1997; Yuan and Shiller, 2001, 2004, 2005; Avery et al., 2005) and inshore surface marine waters and freshwater (Cooper et al., 1989; Kieber and Helz, 1995). Systematic studies on the shelf-seas, and especially the tropical shelf-seas, are few. Here, we report the distribution of hydrogen peroxide in a sub-tropical shelf-sea, the Northern South China Sea Shelf-sea (NoSoCS).

2. Experimental

2.1. The study area

The Northern South China Sea Shelf-sea (NoSoCS) (Fig. 1) stretches southwestward from a ridge system, at about 23°N and 119°E , that delineates the southern end of the Taiwan Strait to the northeastern coasts of the Leizhou Peninsula and Hainan Island at about 20°N and 111°E , and, from the southeastern coast of China to the shelf-break at about the 120-m isobath. At the landward side, the highly urbanized Pearl River discharges into the middle section of the NoSoCS with an annual discharge of $330 \text{ km}^3 \text{ yr}^{-1}$, and it is the dominant source of freshwater to the NoSoCS. At the seaward side, a surface boundary current flows along the shelf edge and slope southwestward in the fall through the spring and north-eastward in the summer in response to the forcing of the monsoonal wind (Gan et al., 2006). Furthermore, extensive activities of internal waves can be found along the entire outer-shelf and slope. These waves are formed at the Luzon Strait. As they propagate westward and reach the shallower waters at the outer shelf and slope of the NoSoCS, they undergo extensive refraction, transformation and dissipation (Lien et al., 2005; Li et al., 2008; Farmer et al., 2011; Guo et al., 2012). The largest of these waves, with amplitudes in excess of 150 m, are found around the Dongsha Atoll (Klymak et al., 2006; Farmer et al., 2011). Although the NoSoCS is connected to the Taiwan Strait to the north, free exchange is limited by a ridge which extends from Dongshan to the Taiwan Bank. The water depths along this ridge do not exceed about 40 m. The NoSoCS was connected to the Beibu Gulf through the narrow Qiongzhou Strait which is situated between the Leizhou Peninsula and the Hainan Island. Coastal

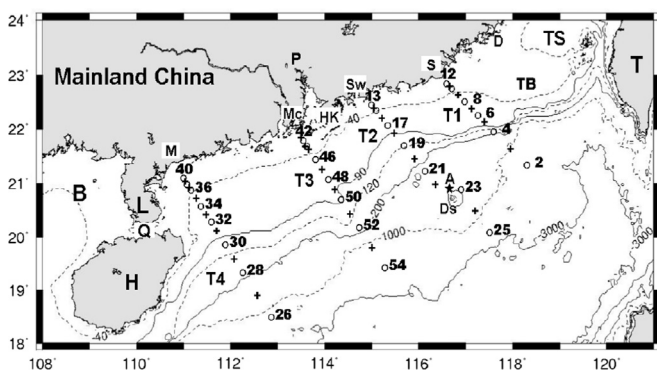


Fig. 1. The study area and the sampling locations in the Northern South China Sea Shelf-sea (NoSoCS). + – CTD stations; o – CTD and discrete sample stations. A – Time series station; B – Beibu Gulf; D – Dongshan; DS – Dongsha Atoll; H – Hainan Island; HK – Hong Kong; L – Leizhou Peninsula; M – Maoming; Mc – Macao; P – Pearl River; Q – Qiongzhou Strait; S – Shantou; Sw – Shanwei; T – Taiwan; TB – Taiwan Bank; and TS – Taiwan Strait.

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