

Vertical and horizontal distribution of fluorescent dissolved organic matter in the Southern Ocean

Youhei Yamashita*, Ayumi Tsukasaki, Tamihito Nishida, Eiichiro Tanoue

*Department of Earth and Environmental Sciences, Graduate School of Environmental Studies,
Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8601, Japan*

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Abstract

The vertical and horizontal distribution of fluorescent dissolved organic matter (FDOM), determined by fluorescence intensity at 320 nm excitation and 420 nm emission, were clarified in nine stations on two transects at the Southern Ocean, including a subtropical, subantarctic, polar frontal and Antarctic zone. All vertical profiles of fluorescence intensity showed that levels were lowest in the surface waters, increased with increasing the depth in mid-depth waters (~2000 m), and then stayed within a relatively narrow range from there to the bottom. Such vertical profiles of FDOM were similar to those of nutrients, but were adverse to dissolved oxygen. In water columns below the temperature-minimum subsurface water (dichothermal waters) in the Antarctic zone and below the winter mixed layer in the other zones, we determined the relationships of fluorescence intensity to concentrations of nutrients and apparent oxygen utilization (AOU) over the entire area of the present study, and found significant linear correlations between the levels of fluorescence intensity and nutrient concentrations ($r=0.70$ and 0.71 for phosphate and nitrate+nitrite, respectively) and AOU ($r=0.91$). From the strong correlation coefficient between fluorescence intensity and AOU, we concluded that FDOM in the Southern Ocean is formed *in situ* via the biological oxidation of organic matter. The regeneration of the nutrients/consumption of the oxygen/formation of FDOM was active in mid-depth waters. However, the correlations between fluorescence intensities and nutrients and AOU were different in the mid-depth water masses, Subantarctic Mode Water (SAMW), and Antarctic Intermediate Water (AAIW), indicating that the sources of organic matter responsible for FDOM formation were different. A considerable amount of FDOM in the SAMW is thought to be produced by the remineralization of DOM in addition to sinking particulate organic matter, while DOM is less responsible for FDOM formation in the AAIW.

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1. Introduction

Dissolved organic matter (DOM) in the ocean constitutes one of the biggest organic carbon pools on

the earth's surface (Hedges and Keil, 1995). Amino acids (Yamashita and Tanoue, 2003a) and monosaccharides (Skoog and Benner, 1997) liberated after the hydrolysis of oceanic DOM contributed less than 5% of the bulk dissolved organic carbon (DOC). Lipids are also a minor constituent of DOC (Kennicutt and Jefferey, 1981; Kattner et al., 1983). Thus, a major component of DOM is comprised of chromatographically uncharacterized molecules, a so-called molecularly-uncharacterized component (MUC, Hedges et al.,

* Corresponding author. Present address: Southeast Environmental Research Center, and Department of Chemistry and Biochemistry, Florida International University, Miami, FL 33199, USA. Tel.: +1 305 348 6085; fax: +1 305 348 4096.

E-mail address: yamashiy@fiu.edu (Y. Yamashita).

2000). Such chemical characteristics of DOM are in striking contrast to those of marine organisms in which more than 80% of organic constituents is comprised of chromatographically measurable amino acids, carbohydrates and lipids (Wakeham et al., 2000).

Marine humic substances have been considered typical of MUC. Such substances are generally investigated by two different technical approaches. In one approach, the humic fraction is extracted from seawater using a resin such as XAD-8 (Stuermer and Harvey, 1977; Malcolm, 1990; Hedges et al., 1992). The chemical and isotopic characteristics of marine humic fractions have been shown to differ from those of terrestrial humic fractions (Malcolm, 1990; Hedges et al., 1992), in that they showed similar $\delta^{13}\text{C}$, but depleted $\delta^{14}\text{C}$ values compared with those of bulk DOC (Bauer et al., 1992; Druffel et al., 1992), suggesting that they were marine in origin and had recalcitrant properties. The other approach is to spectroscopically characterize bulk DOM without extraction, where humic-like light-absorbing and fluorescence properties can be clearly observed (Coble et al., 1990; Mopper and Schultz, 1993; Coble, 1996; Blough and Del Vecchio, 2002). The humic-like optical properties of oceanic DOM are similar but not identical to those of humic fractions, especially the fulvic acid fraction present in both soil and freshwater (Green and Blough, 1994; Coble, 1996; Yamashita and Tanoue, 2003b). The humic-like optical properties of bulk DOM were similar to those of the humic fractions extracted from seawater (Green and Blough, 1994; Coble, 1996).

An optically active component in DOM has recently been called chromophoric DOM (CDOM) or fluorescent DOM (FDOM), the dynamics of which have been well studied in coastal environments *via* measurements of both absorbance and fluorescence. Since the major sources of CDOM (FDOM) in coastal environments are riverine humic substances, negative correlations have been observed between levels of CDOM (FDOM) and salinity (Dorsch and Bidleman, 1982; Hayase et al., 1987; de Souza Sierra et al., 1997; Del Castillo et al., 1999; Jaffé et al., 2004).

In oceanic environments, CDOM (FDOM) has usually been investigated by fluorescence intensity because of its high sensitivity. The levels of FDOM were lowest in the surface waters, and increased with increasing depth (Hayase et al., 1988; Chen and Bada, 1992; Hayase and Shinozuka, 1995; Determann et al., 1996). The depth profiles are mirror images of those of the biomolecules in DOM as well as bulk DOC, and are similar to those of nutrients. Such a distinctive profile is interpreted as indicative of the photobleaching of FDOM by sunlight in surface water (Mopper et al.,

1991; Chen and Bada, 1992; Bertilsson et al., 2004) and the *in situ* production of FDOM accompanied by a remineralization of sinking organic particles in mid-depth waters (Hayase et al., 1988; Chen and Bada, 1992; Hayase and Shinozuka, 1995). However, at present, the relationships among *in situ* production of FDOM, apparent oxygen utilization (AOU), and nutrient regenerations are still at a preliminary stage of qualitative interpretation, since dynamics and relationships among FDOM, nutrients and AOU could not be determined solely from the snapshot vertical observations.

The Southern Ocean has a unique system of water mass distribution, whereby the transition of warm subtropical waters in the north to cold polar waters in the south occurs in a series of fronts, and the relatively uniform and narrow circumpolar water zones divided by those fronts are distinguished by step-like horizontal changes as well as by the characteristic vertical structures of salinity, temperature, and nutrients (*e.g.*, Belkin and Gordon, 1996; Rintoul and Trull, 2001; Sokolov and Rintoul, 2002).

In the present study, we determined 9 vertical profiles of FDOM on two transects that covered various water masses in the Southern Ocean within a relatively small spatial scale. Transect observations carried out in the present study first make it possible to compare the dynamics of FDOM, nutrients and AOU not only vertically but also laterally. Special attention has been paid to clarify the quantitative relationships between fluorescence intensity and water mass characteristics, *i.e.*, AOU and nutrient regeneration and their variability among water masses.

2. Materials and methods

Samples were collected from the South Pacific and Southern Ocean (Fig. 1) during the KH04-5 cruise of the R/V Hakuho-Maru (December 18, 2004–January 14, 2005). Two or three casts with the CTD/Carousel sampling system equipped with 12 L Niskin-X bottles were used to obtain a vertical profile throughout the water column at each station. Seawater samples (*ca.* 14 ml) collected from the spigot of a Niskin-X bottle were poured into Pyrex test tubes (14 ml volume) with Teflon-lined caps after rinsing each one three times with seawater. The outsides of test tubes were coated by brown color. The test tubes were pre-combusted before the cruise and, after being used once, were washed with Milli-Q water for the next use onboard ship. After sampling, the seawater samples were kept at a low temperature ($\sim 5\text{ }^{\circ}\text{C}$). Fluorescence measurements were usually conducted within 3 days after sampling,

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