

## **1. Introduction**

Global warming resulting from the increase in greenhouse gases, such as carbon dioxide, is of great concern to the world community. In the last few decades, the carbon cycle in the ocean has been studied to clarify the balance of carbon dioxide between the atmosphere and the ocean. One important issue is the quantification of the role played by the biological pump: how much atmospheric CO<sub>2</sub> is assimilated in the sunlit layer (euphotic zone) and how much carbon is exported to the deep ocean?

The short-lived radionuclide <sup>234</sup>Th (half-life, 24.1 days) serves as a valuable tracer for studying the rates of particle-associated scavenging and the subsequent particle export from the euphotic zone (Coale and Bruland, 1985; Buesseler, 1998). This tracer is produced in the water column as a dissolved species by radioactive decay of the conservative <sup>238</sup>U in seawater and is redistributed between the dissolved and particulate phases depending on particle reactivity and the availability of particle surfaces.

Particulate organic carbon (POC) fluxes, calculated using the <sup>234</sup>Th method, have been reported from oceans worldwide: North Atlantic (Buesseler *et al.*, 1992), Equatorial Pacific (Buesseler *et al.*, 1995; Bacon *et al.*, 1996; Murray *et al.*, 1996), Middle Atlantic (Michaels *et al.*, 1994; Buesseler, 1998), Arabian Sea (Lee *et al.*, 1998; Buesseler *et al.*, 1998), Northeast Polynya off Greenland (Cochran *et al.*, 1995), Antarctic Ocean (Rutgers van der Loeff *et al.*, 1997), the northeastern North Pacific (Charette *et al.*, 1999), North Pacific Subtropical Gyre (Benitez-Nelson *et al.*, 2001), and Southern Ocean (Buesseler *et al.*, 2001a; Coppola *et al.*, 2005). Buesseler (1998) proposed that the export ratio (*e*-ratio), which is the ratio of POC flux to primary production, would be less than 5–10% in much of the oceanic area of the world, but would be significantly higher at high latitudes and during episodic export pulses such as spring blooms. Charette *et al.* (1999) and Amiel *et al.* (2002) reported the importance of diatoms in elevating POC export.

The northern North Pacific Ocean, especially its western part, experiences intense winter cooling and receives large supplies of nutrients through upwelling, resulting in high productivity in spring and summer. It is well documented that spring blooms, consisting mainly of diatoms, occur only in the western part of the subarctic Pacific

(Saito *et al.*, 2002; Yamaguchi *et al.*, 2002). It has been suspected that diatoms in this area play a key role in transporting POC to the deep ocean, because of their relatively large size and resultant high settling velocity (*e.g.*, Tsunogai and Noriki, 1991; Kemp *et al.*, 2000; Smetacek, 2000; Honda *et al.*, 2002). The large decrease of nutrients and surface  $p\text{CO}_2$  from spring to autumn also support the high activity of the biological pump in the northwestern North Pacific (Louanchi and Najjar, 2000; Wong *et al.*, 2002; Takahashi *et al.*, 2002).

Between 1997 and 2000, we carried out studies of the carbon cycle in the northwestern North Pacific. High intensities of biological pump have been estimated from sediment traps (Honda *et al.*, 2002). Large drawdown of total  $\text{CO}_2$  (ca.  $109 \mu\text{mol kg}^{-1}$ ) was found in the mixed layer in spring (Murata *et al.*, 2002). Kawakami *et al.* (2004) reported that seasonal and horizontal variability in POC fluxes estimated from  $^{234}\text{Th}$  was large in this region. In this dataset, we present vertical profiles of  $^{234}\text{Th}$ , POC, and PON in the northwestern North Pacific. These data will help further understanding of particle dynamics at the euphotic layer.

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### 3. Water sampling and sample analysis

Samples were collected during 5 cruises of R/V *Mirai* between 1997 and 2000 in the northwestern North Pacific and Okhotsk Sea. The cruises were MR97-02 (Nov. 1997; doi: 10.17596/0001891), MR98-K01 (Nov.–Dec. 1998; doi: 10.17596/0001901), MR99-K02 (May 1999; doi: 10.17596/0001904), MR00-K01 (Jan. 2000; doi: 10.17596/0001761), and MR00-K03 (May–Jun. 2000; doi: 10.17596/0001762).

Seawater samples (30 L) were collected from the upper 300 m (at some stations, 200 m) of the water column by Niskin bottle samplers attached to CTD sensors (SBE 911plus Sea-Bird Electronics Inc.) that recorded salinity, temperature, and depth. The seawater samples were immediately filtered through pre-combusted (4 h at 450 °C) Whatman GF/F glass fiber filters (0.7  $\mu\text{m}$  nominal pore size) of 47-mm (for  $^{234}\text{Th}$ ) or 25-mm diameter (for POC, PON, and chlorophyll *a*). Filtering was done on board the research vessel immediately after collection. After filtration, water samples were acidified to pH 1 by the addition of concentrated HCl solution, and the  $^{230}\text{Th}$  yield tracer (2.5 dpm) and 100 mg of Fe (as  $\text{FeCl}_3$ ) were added. After the samples were shaken and allowed to stand for about half a day, ammonia was added to the water samples to precipitate the iron.

$^{234}\text{Th}$  beta background for glass-fiber filters was reported to be likely higher than that for quartz fiber filters (Benitez-Nelson *et al.*, 2001; Buesseler *et al.*, 2001b); however, because in this study we conducted beta-counting for particulate  $^{234}\text{Th}$  without the filter after the purification, this background was negligible.

To measure particulate  $^{234}\text{Th}$ , the filter sample was digested with a mixture of concentrated HCl and  $\text{HNO}_3$  (3:1, v/v) in the presence of the  $^{230}\text{Th}$  yield tracer (2.5 dpm). Radiochemical separation and purification of these nuclides in the dissolved and particulate samples were achieved by procedures similar to those of Anderson and Fleer (1982) using the anion exchange technique. A column separation was carried out on board to remove  $^{238}\text{U}$  from the dissolved samples within two days after the iron precipitation. The rest of the sample purification for determination of dissolved and particulate  $^{234}\text{Th}$  was performed in the land-based laboratory.

In the laboratory,  $^{234}\text{Th}$  of each phase was prepared by using TTA extraction and plating onto stainless steel planchets. The planchets were covered with aluminum foil (6.6 mg  $\text{cm}^{-2}$ ) and first beta-counted using a low-background (0.1–0.3 cpm)

anticoincidence gas-flow beta detector (LBC-470, Aloka Co. Ltd.) to determine their  $^{234}\text{Th}$  activity, and then alpha-counted with silicon surface barrier detectors (Octéte, Seiko EG&G Co. Ltd.) to determine the yield from the  $^{230}\text{Th}$  activity. The concentration of  $^{234}\text{Th}$  was calibrated against deep-water samples equilibrated with  $^{238}\text{U}$ . The analytical precisions ( $1\sigma$ ) for dissolved and particulate  $^{234}\text{Th}$  determinations were about 7.6% and 11.9%, respectively.

Samples for POC and PON analysis were stored in a freezer before analysis. POC and PON were measured with an elemental analyzer (Model 2400II, PerkinElmer Inc.). Before measurement, the samples were treated with concentrated HCl vapor for 24 h to remove calcium carbonate and dried at 50 °C for 3 h. The repeatability ( $1\sigma$ ) of POC and PON measurements was usually less than 3%.

The concentration of chlorophyll *a* in seawater samples was measured by fluorometric determination (Suzuki and Ishimaru, 1990). The precision ( $1\sigma$ ) based on replicated measurements was usually better than 3%.

### 3. Dataset

Data obtained from cruises were electrically compiled as an excel file “234Th\_data\_1997-2000.xls”, and were depth, potential temperature (Theta), salinity, potential density (Sigma-theta), POC, PON, particulate  $^{234}\text{Th}$  (P-234Th), dissolved  $^{234}\text{Th}$  (D-234Th), and chlorophyll *a*. The data of depth, potential temperature, salinity, and potential density were measured by CTD sensors. The error of P-234Th and D-234Th was estimated from counting error.

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