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Particulate organic carbon fluxes estimated from ^{234}Th deficiency in winters and springs in the northwestern North Pacific

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Activities of particulate and dissolved ^{234}Th and concentrations of particulate organic carbon (POC) were measured in the northwestern North Pacific in winters and springs. The export fluxes of POC from surface waters were estimated by using ^{234}Th as a tracer. The POC fluxes in winter showed a wide variation (20–190 mg-C $m^{-2}d^{-1}$) and were higher in the western region than in the eastern region. The influence of the Oyashio Current and the supply of substances from the continent to the study area also appeared to be higher in the western region. Therefore, the horizontal distribution of POC fluxes in winter was influenced by continental materials. In the spring bloom, the fluxes of POC were much higher (up to 520 mg-C $m^{-2}d^{-1}$), as were the e-ratios (up to 70%). In the northwestern North Pacific, the efficiencies of export of POC from the euphotic zone were higher than in other areas of the world ocean. The increase in POC fluxes in spring was considered to depend not only on the increased abundance of phytoplankton, but also on the transition of phytoplankton species, as evidenced by the relationship between ^{234}Th adsorption rates and the concentrations of POC.

Keywords: ^{234}Th, POC, PON, export production, northwestern North Pacific

INTRODUCTION

Global warming due to the increase of greenhouse gases, such as carbon dioxide, in the atmosphere is of great concern to the world community. Over the past few decades, the carbon cycle in the ocean has been studied to clarify the balance between carbon dioxide in the atmosphere and that in the ocean. In order to better understand the processes of carbon transport to the deep ocean, in particular, it is essential to estimate the efficiency of the biological CO_2 pump in the ocean surface.

The northern North Pacific Ocean, especially its western part, has been attracting the attention of biogeochemists because of its importance in global biogeochemical cycles of carbon and related substances. Characterized by intense winter cooling and supply of nutrients by upwelling, the northwestern North Pacific Ocean is thought to make a significant contribution to the global CO_2 sink in winter. It is well documented that the spring blooms, consisting mainly of diatoms, occur only in the western part of the subarctic Pacific (Saito *et al*., 2002; Yamaguchi *et al*., 2002). Owing to their relatively large size and thus high settling velocity, diatoms may play a key role in the area in transporting particulate organic carbon (POC) to the interior of the ocean (e.g., Tsunogai and Noriki, 1991; Kemp *et al*., 2000; Smetacek, 2000).

Previous methods for measuring the export flux relied on sediment traps deployed at relatively shallow depths (*ca*. 200 m) or floating sediment traps. However, such shallow traps have many problems associated with hydrodynamics and the invasion of predatory zooplankton ("swimmers"). Buesseler and co-workers have reported the biases in the quantity and quality of material caught in floating traps (Buesseler, 1991, 1998; Buesseler *et al*., 1994), and proved that the ^{234}Th method works best for estimating particle export from the euphotic zone (Buesseler *et al*., 1998). ^{234}Th is a short-lived (half-life = 24.1 d) radionuclide that can serve as a valuable tracer for studying the rates of particle-associated scavenging processes and the subsequent export of particles from the euphotic zone (Bhat *et al*., 1969; Matsumoto, 1975; Coale and Bruland, 1985, 1987; Bacon *et al*., 1996; Murray *et*

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al., 1996; Buesseler *et al*., 1998). This tracer is introduced into the water column as a dissolved species arising from the radioactive decay of the conservative ^{238}U in seawater, and it is redistributed between the dissolved and particulate phases depending on the reactivity of the particles and the availability of particle surfaces.

Global estimates of POC fluxes derived from ^{234}Th measurements have been carried out in the North Atlantic Bloom Experiment (NABE) (Buesseler *et al*., 1992), the Equatorial Pacific Process Study (EqPac) (Buesseler *et al*., 1995; Bacon *et al*., 1996; Murray *et al*., 1996), the Bermuda Atlantic Time-series Study (BATS) (Michaels *et al*., 1994; Buesseler, 1998), the Arabian Sea Process Study (Buesseler *et al*., 1998; Lee *et al*., 1998), the Northeast Water Polynya off Greenland (Cochran *et al*., 1995), the Antarctic Ocean (Rutgers van der Loeff *et al*., 1997), and the northeastern North Pacific (Station Papa) (Charette *et al*., 1999). The results of those studies indicate that the ratios of POC export to primary production (e-ratios) fall in the range of <5–10% in much of the ocean (Buesseler, 1998); these ratios are significantly higher at high latitudes and during episodic export pulses such as spring blooms. Several studies have revealed the importance of diatoms in association with POC export (Buesseler, 1998; Charette *et al*., 1999; Amiel *et al*., 2002). However, the estimates of export production in the northwestern North Pacific are poorly constrained. Information on the magnitude and rate of export production in bloom and non-bloom periods is important because it can reveal general information about the structure of the phytoplanktonic community and provide estimates of this region's contribution to global export production.

The purpose of this study was to evaluate the spatial variation of POC fluxes estimated by the ^{234}Th method in winter and in a spring bloom in the northwestern North Pacific. In this paper, we present profiles of ^{234}Th (in both dissolved and particulate form), POC, and particulate organic nitrogen (PON) in the northwestern North Pacific and the Okhotsk Sea. Sampling was conducted during the winter seasons of 1997, 1998, and 2000, and during a spring bloom in 1999 and 2000. We estimate the export fluxes of POC and PON out of the euphotic zone and discuss the effects of changes in planktonic species on the behavior of ^{234}Th in the euphotic zone during the spring bloom.

SAMPLING AND ANALYSIS

Water sampling

Samples were collected during the R/V *Mirai* cruises MR97-02 (Nov. 1997), MR98-K01 (Nov.–Dec. 1998), MR99-K02 (May 1999), MR00-K01 (Jan. 2000), and MR00-K03 (May–Jun. 2000) in the northwestern North Pacific and Okhotsk Sea. Sampling stations for the cruises

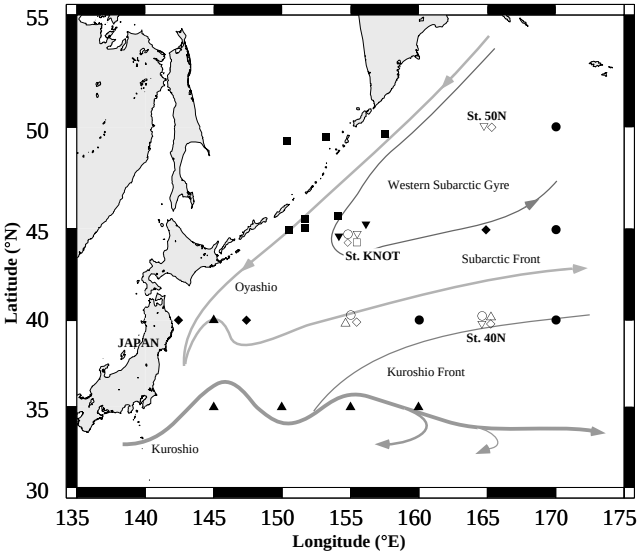

Fig. 1. Sampling locations in the northwestern North Pacific. d *Nov. 1997 (MR97-02);* m *Nov.–Dec. 1998 (MR98-K01);* . *May 1999 (MR99-K02);* r *Jan. 2000 (MR00-K01);* j *May–Jun. 2000 (MR00-K03). The open symbols clustered together are at the same position. The lines with arrows show ocean currents.*

are shown in Table 1 and Fig. 1. Hydrographic parameters were simultaneously measured at each station. During the MR97-02, MR98-K01, and MR00-K01 cruises, surveys were carried out to clarify the spatial variation of ^{234}Th in the northwestern North Pacific in winter. In the spring of 1999, we carried out an intensive survey of a spring bloom (MR99-K02). In 2000, we had the opportunity to survey the area along the Kuril Islands inside the Okhotsk Sea and the Pacific.

Sample analysis

Seawater samples, taken by 30-L Niskin bottle samplers from the upper 300 m (at some stations, 200 m) of the water column, were immediately filtered through precombusted (4 h at 450°C) Whatman GF/F glass fiber filters (0.7 μm nominal pore size) of 47-mm (for ^{234}Th) or 25-mm diameter (for POC, PON, and chlorophyll *a*). After filtration, water samples were acidified with HCl to pH 1, and ^{230}Th yield tracer (2–4 dpm) and 100 mg Fe (as $FeCl_3$) were added. Then ammonia was added to the water samples to induce precipitation. The particulate samples collected on the filters for the measurement of ^{234}Th were digested with a mixture of concentrated HCl and HNO_3 in the presence of ^{230}Th yield tracer. Subsequently, radiochemical separation and purification of the nuclides were achieved by procedures similar to those of Anderson and Fleer (1982) using an anion ion exchange technique. Column separation was carried out on board to remove

Table 1. Sampling locations and POC export fluxes from the euphotic zone in the northwestern North Pacific

Station	Location	Date	Euphotic zone# (m)	^{234}Th fluxes (dpm m^{-2}day^{-1})	POC/^{234}Th (μg-C dpm^{-1})	POC fluxes (mg-C m^{-2}day^{-1})	Remarks
(MR97-02)							
Stn. 1	40°N, 155°E	12 Nov., 97	100	2100	71	150	
Stn. KNOT	44°N, 155°E	16 Nov., 97	100	1200	100	120	
Stn. 3	40°N, 160°E	18 Nov., 97	100	1200	65	81	
Stn. 4	40°N, 165°E	19 Nov., 97	100	760	62	47	
Stn. 7	50°N, 170°E	24 Nov., 97	75	540	90	49	
Stn. 8	45°N, 170°E	26 Nov., 97	100	1100	72	81	
Stn. 9	40°N, 170°E	28 Nov., 97	100	730	67	49	
(MR98-K01)							
Stn. 8	40°N, 145°E	20 Nov., 98	100	2600	74	190	
Stn. 10	35°N, 145°E	25 Nov., 98	100	2100	80	170	
Stn. 11	35°N, 150°E	26 Nov., 98	100	1200	47	54	
Stn. 14	40°N, 155°E	30 Nov., 98	100	1300	51	66	
Stn. 16	35°N, 155°E	2 Dec., 98	100	1300	74	96	
Stn. 17	35°N, 160°E	3 Dec., 98	150	660	34	23	
Stn. 20	40°N, 165°E	8 Dec., 98	150	690	51	35	e-ratio = 15%
(MR99-K02)							
Stn. KNOTL	44°N, 154.5°E	12 May, 99	100	2200*	150	330	
Stn. 50N	50°N, 165°E	17 May, 99	100	460*	83	38	
Stn. 40N	40°N, 165°E	20 May, 99	100	1600*	87	140	
Stn. KNOT2	44°N, 155°E	25 May, 99	100	1700*	210	350	e-ratio = 70%
Stn. HP2	44.3°N, 156.2°E	27 May, 99	100	1600*	320	520	e-ratio = 70%
(MR00-K01)							
Stn. 1	40°N, 142.5°E	6 Jan., 00	150	2100	82	170	
Stn. 2	40°N, 147.5°E	13 Jan., 00	100	790	100	82	
Stn. KNOT	44°N, 155°E	17 Jan., 00	75	260	90	23	e-ratio = 45%
Stn. 14	40°N, 155°E	23 Jan., 00	150	750	89	67	
Stn. 6	50°N, 165°E	26 Jan., 00	100	390	78	30	
Stn. 7	45°N, 165°E	28 Jan., 00	100	370	87	32	
Stn. 8	40°N, 165°E	30 Jan., 00	150	1300	72	95	
(MR00-K03)							
Stn. KNOT	44°N, 155°E	11 May, 00	100	580	100	61	
Stn. 6	44.8°N, 150.8°E	15 May, 00	150	870	140	120	
Stn. 8	48.5°N, 150.5°E	17 May, 00	100	220	89	19	Okhotsk Sea
Stn. 11	45.5°N, 152.2°E	19 May, 00	75	95	120	12	an eddy
Stn. 12	45.2°N, 152.8°E	20 May, 00	100	140	100	15	an eddy
Stn. 17	49°N, 153.2°E	31 May, 00	60	280	100	30	Okhotsk Sea
Stn. 22	49.1°N, 157.9°E	1 Jun., 00	100	2100*	270	560	
Stn. 15	46°N, 154°E	4 Jun., 00	100	2600*	170	430	

**Non-steady-state model used to calculate ^{234}Th flux.*
#Thickness of euphotic zone used for flux calculations.

^{238}U from the dissolved samples. The rest of the sample purification for dissolved and particulate ^{234}Th was performed in the land-based laboratory at the Japan Marine Science and Technology Center (now the Japan Agency for Marine-Earth Science and Technology). Finally, alpha sources were prepared by using TTA extraction and plating onto stainless steel planchets.

The planchets were covered with aluminum foil (6.6 mg cm^{-2}) and first beta-counted using a low background (0.1–0.3 cpm), anticoincidence, gas-flow beta detector to determine their ^{234}Th activity, and then alpha-counted with silicon surface barrier detectors to determine the yield from the ^{230}Th activity. To check the accuracy and precision of the ^{234}Th determination, we analyzed two equilibrated deep-water samples with the techniques described above. The results were also used to determine the beta/alpha ratios of the detectors. Calibrations for instrument counting efficiencies (β/α ratio), ^{234}Th decay correction,

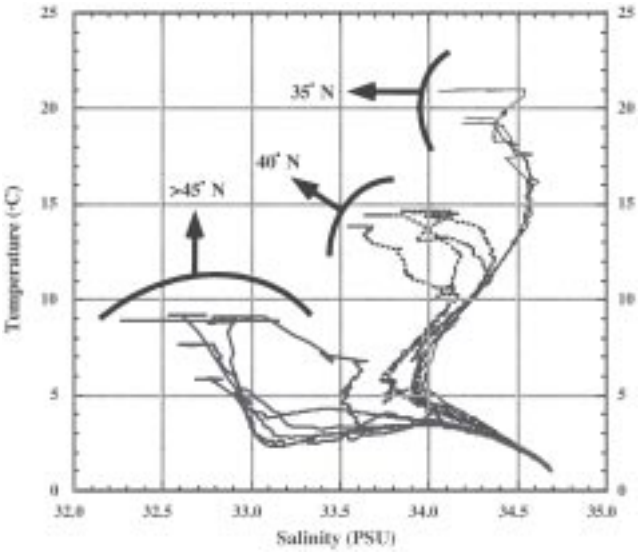

Fig. 2. T-S diagram for water masses in the western North Pacific in Nov. 1997.

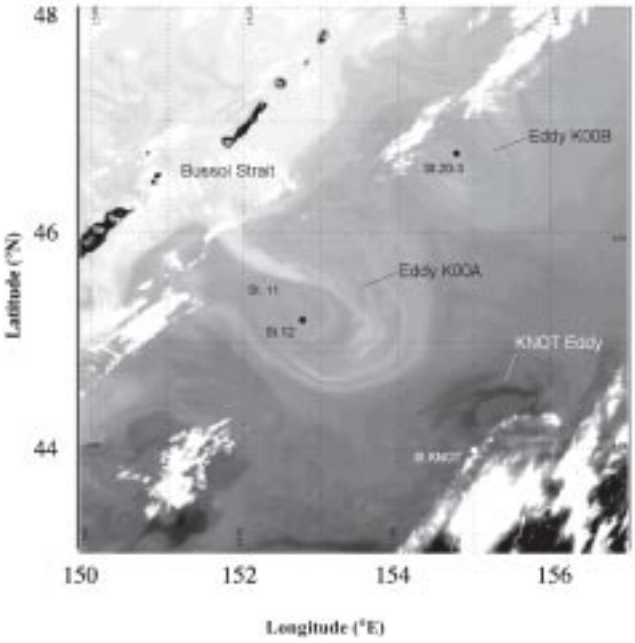

Fig. 3. Mesoscale eddies off the Kuril Islands as shown in an NOAA AVHRR infrared satellite image for 29 May 2000 (UTC). Light shades of gray correspond to cold water. The clouds are white and warm water is dark. Locations of some stations for the MR00-K03 cruise are shown.

and ^{234}Th ingrowth correction were also made. All reported errors are within 1 standard deviation based on the propagation of alpha- and beta-counting errors and the corresponding uncertainties in background, detector blanks, and efficiency as well as errors in spike calibrations.

The samples for POC and PON analyses were stored frozen until analysis. POC and PON were measured with a CHN analyzer (Grasshoff *et al.*, 1999). Before the measurements, the particle samples were treated with the vapor of concentrated HCl for 24 h to remove calcium carbonate, and dried under vacuum at 40°C for 3 h.

Oceanographic Setting of the Study Area

General oceanographic setting

In the study area, two western boundary currents, the Kuroshio and the Oyashio, are associated with subtropical anticyclonic and subarctic cyclonic gyres, respectively (Fig. 1). These currents interact with each other, resulting in a system of a complex frontal structure and mixing of water masses, known as the Kuroshio–Oyashio confluence region (Kawai, 1972). To the north of this region, the Oyashio carries waters originating in the Western Subarctic Gyre and the Okhotsk Sea to the southeast coast of Hokkaido. This Oyashio Water is cold and nutrient-rich, leading to high abundance of phytoplankton and high trophic levels, and is one of the most productive waters of the world ocean (Saito *et al.*, 2002; Yamaguchi *et al.*, 2002). In contrast, the region that comprises the Subtropical Water, south of the Kuroshio Current, is made up of warm, highly saline water. In general, the upper water column of the Subtropical Water is well stratified, severely constraining the vertical mixing of nutrient-rich water essential for phytoplankton growth.

Water masses in the surface can be classified into three groups (Fig. 2). The first group is characterized by high temperature and high salinity influenced by the Kuroshio water and is located around 35°N. The second group has low temperature and low salinity, indicative of the Subarctic Gyre Water, and is located in the regions of >45°N. The third group has characteristics of a mixture of the above two water masses, and is located in the Subarctic Front (40°N).

In addition, mesoscale anticyclonic eddies off the Kuril Islands make the water structure in the area even more complex (Fig. 3). Kusakabe *et al.* (2002) have shown that the boundaries of the eddies are composed of highly productive coastal Oyashio water, and encompass low-temperature Okhotsk Sea water.

Hydrographic properties during the spring bloom

The waters in the region have significant seasonal variations. The biogeochemical cycle of carbon in this region reflects these variations in physical parameters and biological activity. Surveys of a spring phytoplankton bloom were carried out over two separate periods during the MR99-K02 cruise. A variety of measurements were

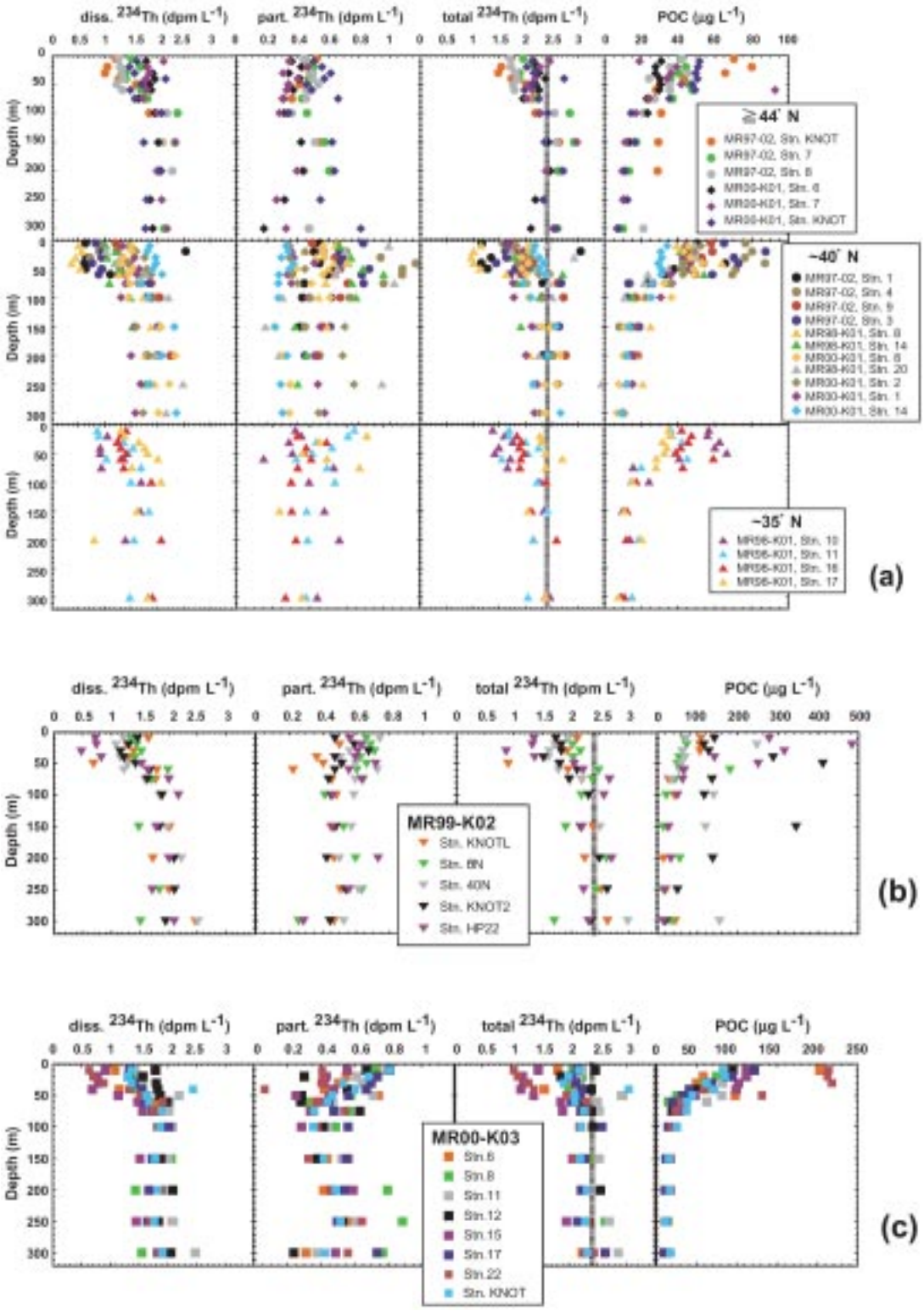

Fig. 4. Vertical distributions of dissolved ^{234}Th, particulate ^{234}Th, total ^{234}Th, and POC. (a) Data obtained during cruises MR97-02, MR98-K01, and MR00-K01; (b) data obtained during cruise MR99-K02; and (c) data obtained during cruise MR00-K03. Shapes of marks used in the plots are the same as those in Fig. 1. Vertical lines in the plots of total ^{234}Th indicate ^{238}U concentration equal to the radioactive equilibrium activity of ^{234}Th (ca. 2.4 dpm L^{-1}). The upper panel of (a) shows the data at or north of 44°N, the middle panel shows data at and around 40°N, and the bottom panel shows data at and around 35°N. Note that the horizontal axes of POC for each plot are different.

made, including measurements of carbonate species. At Stn. 50N, fluorescence was high (*ca*. 10.0 mg m^{-3}) in the upper 20 m, indicating that a spring phytoplankton bloom was occurring, while at Stn. 40N, it was low (1.0 mg m^{-3}) in the upper ocean (Sasaoka, personal communication). Thus Stns. 50N and 40N can be judged to reflect bloom and pre-bloom conditions, respectively, at that time.

During the first survey period, the study area had patches acting as a net sink for atmospheric CO_2 surrounded by areas acting as a net source for atmospheric CO_2 (Murata *et al*., 2002). In the area under bloom conditions, the CO_2 influx to the ocean reached a maximum of approximately 12 mmol $m^{-2}d^{-1}$. The downward CO_2 flux was still dominant 14 days later. In contrast, the area under pre-bloom conditions was a net source of atmospheric CO_2, with an efflux of approximately 3 mmol $m^{-2}d^{-1}$ (Murata *et al*., 2002).

Murata *et al*. (2002) reported that diatoms play an important role in the total CO_2 drawdown in the northwestern North Pacific, because the ratios of SiO_2 to total CO_2 and nitrates + nitrites consumption ($\Delta Si/\Delta C$ and $\Delta Si/\Delta N$) were higher during the spring bloom period.

RESULTS

Vertical profiles of ^{234}Th and POC in winter

Vertical profiles of ^{234}Th (dissolved, particulate, and total) and POC are plotted in Fig. 4(a). For simplicity errors are not shown; errors in the data are, on average, 7.6% for dissolved ^{234}Th, 11.9% for particulate ^{234}Th, and 6.6% for total ^{234}Th. In general, the total ^{234}Th results for all the cruises show the commonly observed depletion relative to ^{238}U throughout the upper approximately 100-m layer. This deficiency is attributed to ^{234}Th scavenging and subsequent downward export *via* settling particles. Below that, down to approximately 300 m, there was occasionally a zone in which ^{234}Th exceeded the radioactive equilibrium value (*ca*. 2.4 dpm L^{-1}; Chen *et al*., 1986), presumably by release from particles sinking through the upper zone. The concentrations of particulate ^{234}Th generally did not show a vertical change as marked as those of dissolved and total ^{234}Th, except at some stations where the particulate ^{234}Th concentrations tended to mirror the dissolved ^{234}Th, showing a maximum in the surface layer and a minimum in the subsurface layer. Enrichment of particulate ^{234}Th in the surface seems most marked in waters of higher POC content. Approximately 15%–35% of the total ^{234}Th was in particulate form.

Spatial variability of the vertical profiles, especially those of particulate ^{234}Th, was largest along the 40°N line (Fig. 4(a)), reflecting the variable features of the water structures such as the coastal water, Oyashio Water, and Kuroshio Front, and their respective biogeochemistries.

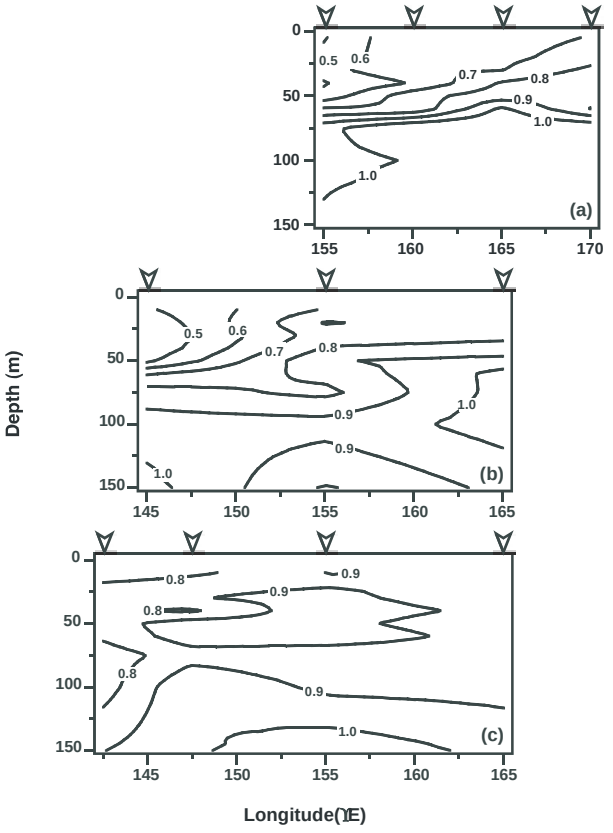

Fig. 5. Vertical distributions of ^{234}Th/^{238}U ratios along the 40°N line in Nov. 1997 (a), Nov.–Dec. 1998 (b), and Jan. 2000 (c). Arrows indicate sampling stations.

The east-west horizontal gradient is more obvious from the ratios of total ^{234}Th to ^{238}U (Fig. 5). The ^{234}Th/^{238}U ratios in the western part of the study area are smaller than those in the eastern part; therefore, because the concentration of ^{238}U is almost constant, the ^{234}Th concentrations are lower in the west.

^{234}Th distributions in the spring bloom

The data on ^{234}Th and POC obtained in spring are plotted in Figs. 4(b) and (c). General vertical profiles in spring are the same as those in winter, although the geographical area covered for sampling was different. Noteworthy differences between the spring and winter data are POC concentrations, especially in the spring of 1999; POC concentrations in the blooming area are approximately five times greater than POC concentrations in winter samples (Fig. 4(b)). The concentration of total ^{234}Th in the high production area (Stn. HP2) was the lowest in this study, possibly because of the vigorous removal of ^{234}Th. In the spring of 2000, despite the relatively small sampling area, there was a wide variation in the distribution of ^{234}Th and POC data (Fig. 4(c)). The Okhotsk Water

and the Pacific Subarctic Water influenced by the Okhotsk Water (Stns. 8, 11, 12, and 17) had a higher total ^{234}Th activity, probably because the spring bloom was yet to occur in those waters.

DISCUSSION

^{234}Th export fluxes

The ^{234}Th method for estimating the export flux is based on the disequilibrium between ^{234}Th and ^{238}U that exists in the euphotic zone. The actual measurement used here is the activity of total ^{234}Th. The activity balance of total ^{234}Th in seawater can be described by the following equation:

$$\partial^{234}\mathrm{Th}_T/\partial t = (^{238}\mathrm{U} - ^{234}\mathrm{Th}_T)\lambda - P + V \qquad (1)$$

where ^{238}U is the uranium activity calculated from salinity, $^{234}\mathrm{Th}_T$ is the measured activity of total ^{234}Th, λ is the decay constant for ^{234}Th (0.0288 d^{-1}), P is the rate of removal of ^{234}Th on settling particles, and V is the sum of the advective and diffusive ^{234}Th fluxes.

We assume that the horizontal gradient of ^{234}Th in this region is negligible. Although a meridional gradient most likely exists, the mean flow of the currents is zonal, from west to east, making V in the meridional direction small. Advection terms generally have very little effect on the value, with the exception of near the equator and upwelling areas (Dunne *et al*., 2000). V can be considered negligible because it is much lower than the removal rate (P) (Buesseler, 1998). Calculations from the North Atlantic Bloom Experiment and the Southern Ocean show that vertical advection comprised only 1%–10% of the ^{234}Th flux (Buesseler *et al*., 1992, 2001). Based on evidence from sediment trap experiments (Honda *et al*., 2002), temporal variation of particulate fluxes in winter in the northern North Pacific is relatively small. Therefore, we assume a steady-state condition when calculating the winter results. For the ^{234}Th balance calculation during the spring blooms, a non-steady-state condition is assumed (see below for details).

The export fluxes of ^{234}Th can be determined from the ^{234}Th deficit relative to ^{238}U. The total ^{234}Th activity profile is integrated from the sea surface down to the euphotic depth. Here we use chlorophyll *a* distribution as a measure to set the bottom of the euphotic layer; i.e., the chlorophyll *a* concentration reaches a minimum at the bottom of the layer. The euphotic depth ranged mostly from 75 m to 150 m (Table 1).

POC/^{234}Th ratios

POC/^{234}Th ratios show wide variations in various regions and with different filtration systems (Buesseler, 1998). In order to use POC/^{234}Th and PON/^{234}Th ratios to convert the rate of removal of particulate ^{234}Th to POC and PON export fluxes out of the euphotic zone, it is important to understand the extent to which POC/^{234}Th ratios vary between various sampling techniques.

In this study, the concentrations of POC and particulate ^{234}Th were determined by filtering approximately 40 L of seawater through a 0.7-μm GF/F filter. The ratios of integrated POC to ^{234}Th in the euphotic layer did not vary very much, with an average and standard deviation of 73 ± 17 μg-C dpm^{-1} in winter (MR97-02, MR98-K01, and MR00-K01) (Table 1). Harada *et al*. (2001) deployed floating sediment traps at Stn. KNOT in Nov. and Dec. of 1998. The POC/^{234}Th ratios obtained at 100 m depth by the traps were 73 and 62 μg-C dpm^{-1}. Although our depths of interest were not strictly the same as Harada *et al*.'s and the sampling locations were not exactly the same, these numbers are comparable to ours, implying that measuring POC/^{234}Th in particles collected on a 0.7-μm GF/F filter can be regarded as representative of POC/^{234}Th ratios in winter. However, in the spring of 1999, POC/^{234}Th ratios showed larger values and wide variation (80–320 μg-C dpm^{-1}) (Table 1). The POC/^{234}Th ratios in this study fell within the range of 10–560 μg-C dpm^{-1} compiled by Buesseler *et al*. (1998) for spring. However, the POC/^{234}Th ratios of material caught in floating sediment traps at Stn. KNOT in spring were lower (74 μg-C dpm^{-1}) than the ratios in the particles we collected on 0.7-μm GF/F filters (Harada *et al*., 2001), though it is the only data available for comparison.

On the basis on evidence from a compilation of POC/^{234}Th ratios in particles of different types, Moran *et al*. (1997) suggested that as suspended particles are packaged into large, rapidly sinking aggregates, carbon is preferentially remineralized over ^{234}Th. The sinking particles are thus depleted in organic carbon relative to ^{234}Th, leading to lower POC/^{234}Th ratios. Buesseler *et al*. (1995) also pointed out that the POC to ^{234}Th ratio is higher in suspended particles than in sinking particles; that is, the suspended particles were fresher than sinking particles. Therefore, we would expect that our estimated POC fluxes should give upper limits. However, as discussed below, during the spring bloom period when a transition of phytoplankton species occurred, the larger diatoms would have a higher POC/^{234}Th ratio than the smaller phytoplankton, as suggested by Charette and Moran (1999).

POC and PON fluxes in winter

The vertical flux of POC (F_{POC}) was calculated from ^{234}Th by using the ratios of POC to particulate ^{234}Th as follows:

$$F_{\mathrm{POC}} = \left(\mathrm{POC}/^{234}\mathrm{Th}_p\right) \times \lambda \int_0^{z_0} \left(^{238}\mathrm{U} - ^{234}\mathrm{Th}_T\right) dz \qquad (2)$$

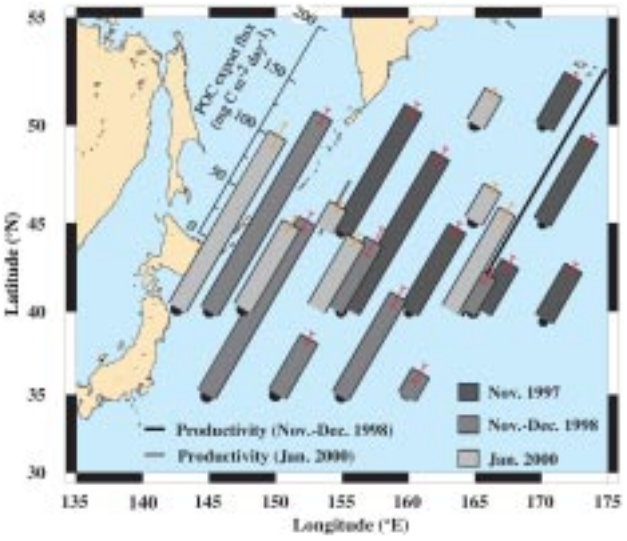

Fig. 6. Horizontal distributions of POC fluxes and productivity in Nov. 1997, Nov.–Dec. 1998, and Jan. 2000. Productivity data are quoted from Imai et al. (2002).

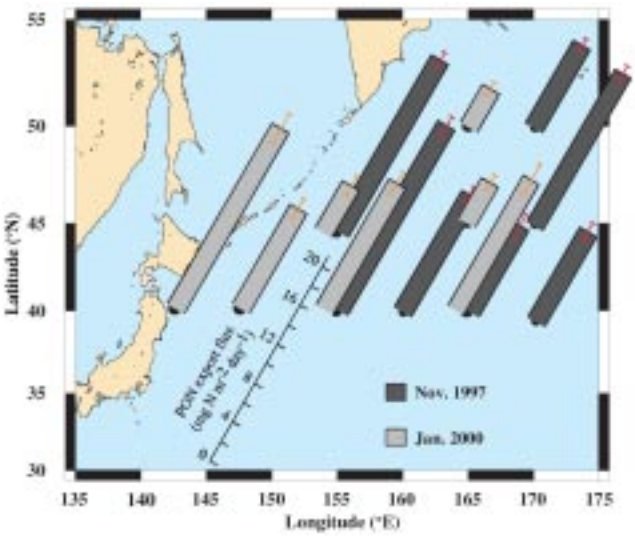

Fig. 7. Horizontal distributions of PON fluxes in Nov. 1997 and Jan. 2000.

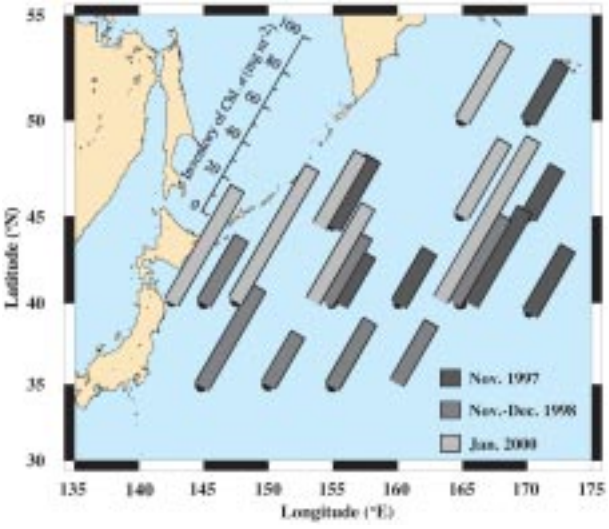

Fig. 8. Horizontal distributions of chlorophyll a in Nov. 1997, Nov.–Dec. 1998, and Jan. 2000. (Data courtesy of Dr. Sasaoka.)

where z_0 is the depth at the base of euphotic zone and $^{234}Th_p$ is the measured activity of particulate ^{234}Th. The ratios POC/$^{234}Th_p$ were determined from inventories of POC and particulate ^{234}Th above depth z_0. In this calculation, POC/$^{234}Th_p$ ratios of particles on the 0.7-μm GF/F filter were presumed to be equal to the ratios of settling particles, as discussed in the section above. Similar terminology is appropriate to the nitrogen cycle, and the fluxes of PON (F_{PON}) were calculated similarly to POC fluxes.

The horizontal distributions of POC fluxes in winter are shown in Fig. 6. The variations of POC fluxes were wide (20–190 mg-C $m^{-2}d^{-1}$). Andreev *et al*. (2002) calculated the average annual POC flux at about 100 m depth in the western Subarctic Gyre (Fig. 1) to be approximately 100 mg-C $m^{-2}d^{-1}$. In addition, Midorikawa *et al*. (2002) estimated the annual average of new production (which should be equivalent to the export flux at a steady state) at 48°N 165°E to be 95 ± 7 mg-C $m^{-2}d^{-1}$. Considering the temporal and spatial variations of the fluxes, these values agree reasonably well with each other. The fluxes of POC appeared higher in the western part of the study area than in the eastern part. The distribution of PON fluxes in winter (Fig. 7) also shows, to a lesser extent, a trend similar to that of POC. The nutrient-rich Oyashio Current, which leads to a high abundance of phytoplankton in the western region, is possibly responsible for the difference. Sediment trap experiments also showed fluxes of aluminum to be higher in the western region (44°N 155°E) than in the eastern region (40°N 165°E and 50°N 165°E) (Honda *et al*., 2002). The continental materials were thought to also affect the profiles of ^{234}Th and POC fluxes. However, these factors alone cannot explain the east-west difference of POC fluxes, because the inventories of chlorophyll *a* do not show a clear difference between the western and the eastern regions (Fig. 8) and, therefore, the biomasses of phytoplankton in the euphotic zone are apparently equivalent between the western and the eastern regions. Sediment trap experiments also showed fluxes of opal to be higher in the western region (44°N 155°E) than in the eastern region (40°N 165°E and 50°N 165°E) (Honda *et al*., 2002). The efficiency of materials sinking out of the

Table 2. PON export fluxes and rates of N (NO_2 + NO_3) decrease in spring

Date	Site	PON/^{234}Th (μg-N/dpm)	PON fluxes (mg-N m^{-2}day^{-1})	N decrease rates (mg-N m^{-2}day^{-1})	E ratio (%)
(MR99-K02)					
12 May, 99	44°N, 154.5°E	32	69	150	46
25 May, 99	44°N, 155°E (KNOT)	36	60	150	40
27 May, 99	44.3°N, 156.2°E	55	91	200	46
(MR00-K03)					
1 Jun., 00	49.1°N, 157.9°E	55	120	180	64
4 Jun., 00	46°N, 154°E	42	110	110	100

euphotic zone is reportedly related to the presence of diatoms (Smetacek, 1985; Sancetta *et al*., 1991; Buesseler, 1998). The difference in standing stocks of diatoms is likely associated with the difference in POC flux between the west and the east.

POC fluxes in the bloom period

During the spring bloom, the fluxes of POC changed suddenly because the standing stocks of phytoplankton increased dramatically. The concentrations of ^{234}Th also decreased, possibly because of enhanced scavenging at that time. Therefore, a steady-state model cannot be used to calculate ^{234}Th flux in the bloom. Using a linear approximation, Eq. (1) gives a rate of removal (P) of ^{234}Th from the euphotic zone as follows:

$$P = [(^{238}U_1 + {}^{238}U_2)/2 - (^{234}Th_1 + {}^{234}Th_2)/2]\lambda - [(^{234}Th_2 - {}^{234}Th_1)/(t_2 - t_1)], \quad (3)$$

where t_1 and t_2 represent the beginning and the end of the time interval, and $^{238}U_n$ and $^{234}Th_n$ are measured activities in the euphotic zone at t_n.

On the basis of sediment trap data (Honda *et al*., 2002) and satellite data (Murata *et al*., 2002), April 23 was regarded as immediately before the spring bloom; we therefore set t_1 to be April 23 (1999 and 2000). We assumed the vertical distributions of ^{238}U and ^{234}Th on April 23 to be the same as those in winter; we therefore used the winter data (Jan. 2000) for $^{238}U_1$ and $^{234}Th_1$. The other calculations for the fluxes of POC and PON are similar to those of the winter studies.

In the spring bloom, the deficiencies of ^{234}Th relative to ^{238}U were higher than in winter. The fluxes of POC in the bloom in 1999 were several times higher than in winter; the maximum value was 520 mg-C m^{-2}d^{-1} (Table 1). In spring 2000, high fluxes of POC close to those in 1999 were also observed at two stations (Stns. 15 and 22), but most of the POC fluxes were not as high as in the spring of 1999 (Table 1). In particular, the POC fluxes near the eddy and in the Okhotsk Sea were lower than at the rest of the stations, and are the lowest values observed in this study (10–20 mg-C m^{-2}d^{-1}).

In the spring bloom period, the fluxes of POC and the e-ratios were notably higher than in winter, with the highest e-ratio being 70% (Table 1). Tsurushima *et al*. (2002) estimated the net community productivity from May to June in 1999 at Stn. KNOT to be 600 mg-C m^{-2}d^{-1}, which is almost the same as the primary productivity that they measured. This almost 100% e-ratio is concordant with the high e-ratio shown by our measurements. We think these high POC fluxes and e-ratios are related to the increased abundance of phytoplankton (diatoms) in the spring bloom. We assume that the low POC fluxes in spring 2000 were related to the eddy and that these sampling regions were still in "winter mode".

The rates of nitrogen decrease in the euphotic zone (ΔNO_x; NO_x = nitrates + nitrites) were calculated by the following equation:

$$\Delta NO_x = (N_1 - N_2)/(t_2 - t_1), \quad (4)$$

where t_1 and t_2 represent the beginning and the end of the time interval, respectively, and N_1 and N_2 are the measured concentrations of nitrates + nitrites (NO_x) at t_1 and t_2, respectively.

We assumed that t_1 was April 23, and that N_1 was the "winter mode data", similar to the calculation of ^{234}Th fluxes. The "winter mode data" were used at a station where the nutrients were at an almost constant concentration throughout the surface layer (44.8°N 156°E in early spring 1999) (Murata *et al*., 2002). The differences in the NO_x concentrations were calculated from the differences between the vertical distribution of the winter mode data and the spring vertical distribution data.

If the rates of NO_x decrease are assumed to be new productivity, then the e-ratios of nitrogen were high in spring, ranging from 40% to 100% (Table 2). Therefore, the sinking efficiencies of nitrogen in the northwestern North Pacific were also much higher in spring.

The e-ratios in much of the world ocean were estimated to be <5%–10% (Buesseler, 1998). In this study, the stations at which primary productivity was measured

Table 3. Summary of POC export studies based upon the ^{234}Th approach

Site	Term	POC flux (mg-C $m^{-2}day^{-1}$)	E ratio (%)	Reference
North Atlantic (NABE sites)	Apr. 25–May 30, 1989	80–920	8–79	Buesseler (1998)
North Atlantic (BATS sites)	Mar.–Oct., 1993–1995	10–70	1–56	Buesseler (1998)
Equatorial Pacific (EqPac sites)	Feb. 23–Oct. 21, 1992	20–110	2–10	Buesseler (1998)
Polynya, Greenland	Spring, 1992–1993	160–840	41–88	Buesseler (1998)
Antarctic	Spring (bloom), 1992	50–350	16–100	Buesseler (1998)
Arabian Sea	Jan.–Sep., 1995	10–310	1–27	Buesseler (1998)
Subtropical and equatorial Atlantic	May–Jun., 1996	40–510	—	Charette and Moran (1999)
Western and central Equatorial Pacific	Oct., 1994	50–160	—	Dunne *et al.* (2000)
	Apr.–May, 1996	40–150	—	Dunne *et al.* (2000)
Northeastern North Pacific	May, 1996	30–150	8	Charette *et al.* (1999)
	Aug., 1996	30–140	13	Charette *et al.* (1999)
	Feb., 1996, 97	10–70	6	Charette *et al.* (1999)
Northwestern North Pacific	Nov.–Dec., 1997	50–150	—	This study
winter	Nov.–Dec., 1998	20–190	15[a]	This study
	Jan.–Feb., 2000	20–170	45[b]	This study
spring	May, 1999	40–520	70[b]	This study
	May–Jun., 2000	10–560	—	This study

[a] *St. 40N.*
[b] *St. KNOT.*

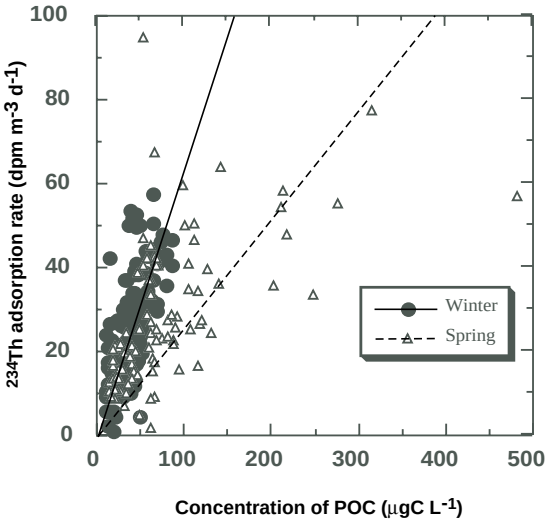

Fig. 9. Relationship between ^{234}Th adsorption rate and POC concentration. The solid and dotted lines are linear regressions for winter data and spring data, respectively.

(Imai *et al.*, 2002) were Stn. 40N in 1998 and Stn. KNOT in 1999 and 2000. The calculated e-ratios in the northwestern North Pacific were 15%–45% in winters and 70% in the spring bloom (Table 1). The e-ratios and POC fluxes in this region were much higher than in much of the world ocean (Table 3), with POC fluxes in the northwestern North Pacific being about three times higher than those in the northeastern North Pacific in spring.

Seasonal variation in the composition of particulate matter

With the negligible advection and diffusion, ^{234}Th adsorption rates were calculated from the following:

$$\partial A^{d}_{Th}/\partial t = A_U\lambda - A^{d}_{Th}\lambda - J, \quad (5)$$

where A_U is the activity of ^{238}U, λ is the decay constant of ^{234}Th, and A^{d}_{Th} is the activity of dissolved ^{234}Th. J is the rate of net adsorption of dissolved ^{234}Th onto particles. The adsorption rates calculated for the spring bloom were based on the non-steady-state model, and those for the other seasons were based on a steady-state condition.

The relationship between ^{234}Th adsorption rate and POC concentration in this study was linear in winter, but showed poor linearity in spring (Fig. 9). The regression line for the winter data is steeper than that of spring. Because ^{234}Th adsorption rates are regarded as proportional to the surface area of POC, they would be proportional to the concentrations of POC if the sizes of POC did not change with time. Thus, the change in the slope in Fig. 9 implies that the size of POC changes in spring, most likely through the appearance of larger phytoplankton. Charette *et al.* (1999) observed in the subarctic Northeast Pacific Ocean that the POC/^{234}Th ratios were two times higher on the large particles collected by the sediment trap when compared with >1-μm particles collected by *in situ* filtration. A similar trend of POC/^{234}Th ratios being higher on large particles than on small particles was found in the South and Equatorial Atlantic Ocean (Charette and

Moran, 1999); they attributed this to the presence of large diatoms that would have a higher POC/^{234}Th ratio relative to smaller phytoplankton.

The dominance of large diatoms in spring is well documented (Imai *et al*., 2002; Liu *et al*., 2002; Mochizuki *et al*., 2002); the dominant species in this region have been estimated to be centric diatoms all the year round, but pennate diatoms increased during the spring bloom. The trends toward higher POC/^{234}Th ratios in spring (Table 1) can thus be attributed to the transition of phytoplankton structure to larger diatoms in spring. These results showed that the increase in POC fluxes in spring was due not only to the increased abundance of phytoplankton, but also to the transition of phytoplankton species.

CONCLUSION

The ^{234}Th/^{238}U disequilibrium in the euphotic zone was used to derive the export fluxes of ^{234}Th and particulate organic carbon and nitrogen in the northwestern North Pacific Ocean. The estimated export fluxes of particulate ^{234}Th were higher, in general, over the regions characterized by higher rates of water column productivity. The ^{234}Th data were used to derive the fluxes of particulate organic carbon and nitrogen out of the euphotic zone.

POC fluxes in winter showed wide variation (20–190 mg-C $m^{-2}d^{-1}$) in the northwestern North Pacific, and tended to be higher in the western region than in the eastern region. The influence of the Oyashio Current and the supply of substances from the continent to the study area appeared to be higher in the western region than in the eastern region. This tendency seemed to influence the primary productivity and biomass in this area. Thus, the horizontal distribution of POC fluxes in winter was influenced by the continental materials. The fluxes of POC and e-ratios in the spring bloom were much higher (up to 520 mg-C $m^{-2}d^{-1}$ and 70%, respectively). Therefore, in the northwestern North Pacific, the export efficiencies of POC from the euphotic zone were higher than in other areas in the world ocean. The increase in POC fluxes in spring depended not only on the increased abundance of phytoplankton, but also on the transition of species of phytoplankton.

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