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Particle-reactive radionuclides (^{234}Th, ^{210}Pb, ^{210}Po) as tracers for the estimation of export production in the South China Sea

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Abstract

The time-series station, SEATS (18° N, 116° E) in the South China Sea was visited six times during October 2006–December 2008 to carry out seawater sampling and floating trap deployments for the determination of distributions and fluxes of POC, PIC, PN, ^{234}Th, ^{210}Pb, and ^{210}Po in the upper 200 m of the water column. Radionuclide deficiencies resulted in removal fluxes from the euphotic layer of 1.1×10^3–1.8×10^3 dpm m^{-2} d^{-1} and 7.1–40.2 dpm m^{-2} d^{-1} for ^{234}Th and ^{210}Po, respectively. Due to atmospheric input, an excess of ^{210}Pb relative to ^{226}Ra is commonly observed in the upper water column. Sinking fluxes of total mass, POC, PIC, PN, ^{234}Th, ^{210}Pb, and ^{210}Po measured at the euphotic depth were low in summer-fall and high in winter-spring, reflecting the seasonal variability of biological pumping. Excluding the suspiciously low primary productivity data point in July 2007, a relatively high e-ratio of 0.28–0.69 was estimated by the ratio of the POC flux at the euphotic depth and the integrated primary productivity. The ratios of ^{234}Th, ^{210}Pb, and ^{210}Po to organic carbon, inorganic carbon, and nitrogen in the sinking particles were combined with the disequilibria of ^{234}Th-^{238}U, ^{210}Pb-^{226}Ra, and ^{210}Po-^{210}Pb to estimate export fluxes of POC, PIC, and PN from the euphotic layer. Compared with measured fluxes by the sediment trap and estimated fluxes by other approaches, it is concluded that the export production in the South China Sea, ranging from 1.8 to 21.3 mmol-C m^{-2} d^{-1}, can be reasonably estimated using ^{234}Th, ^{210}Pb, and ^{210}Po as carbon proxies.

1 Introduction

It is known that carbon dioxide sequestration depends, in part, on the magnitude of carbon removal via particle settling from the surface to the deep layer of the ocean. Thus, the quantification of export flux from the euphotic zone is of great importance to the understanding of carbon cycling. The SEATS site, a time-series station regularly visited by Taiwanese oceanographers since 1999, is located at 18° N 116° E in the

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central basin of the South China Sea (Wong et al., 2007). Although being in a marginal sea, the site shows open-ocean characteristics with low productivity. The SEATS is an ecosystem dominated by picoplankton with a relatively low primary productivity ranging between 300 mg-C m^{-2} d^{-1} in summer to 550 mg-C m^{-2} d^{-1} in winter (Chen et al., 2004; Chen and Chen, 2005). The South China Sea has a thin mixed layer of 30–80 m and a constant euphotic depth of 100 m, which make the diffusive flux of nutrients an important mechanism of supporting the primary production in this oligotrophic system (Tseng et al., 2005).

Despite the fact that previous estimates of the export flux of particulate organic carbon (POC) at the SEATS in the South China Sea were attempted by various approaches, e.g., biogeochemical modeling (Liu et al., 2002), carbon budgeting (Chou et al., 2006), and new productivity measurements (Chen and Chen, 2005; Chen et al., 2004), the magnitude of POC export from the euphotic zone has never directly been measured. Previously, short-lived particle-reactive radionuclides, ^{234}Th ($t_{1/2} = 24.1$ d), ^{210}Pb ($t_{1/2} = 22.4$ a), and ^{210}Po ($t_{1/2} = 138$ d), have been found to be very useful for the estimation of export production in the ocean. Among the three, ^{234}Th has been widely used as a powerful tracer for POC and provided estimates of the export production (see review of Buesseler, 1998). There is only a few studies that used ^{210}Pb to estimate the export production in the ocean, although a strong correlation of ^{210}Pb removal with the POC flux was previously demonstrated (Moore and Dymond, 1988). On the other hand, due to its high affinity with biological particles (Cherry et al., 1975), ^{210}Po is also used as a powerful tracer for particulate matter sinking out of surface ocean (Murray et al., 2005; Friedrich and Rutgers van der Loeff, 2002; Verdeny et al., 2009; Kim and Church, 2001; Buesseler et al., 2008).

Here we report the results of POC, PIC, and PN export fluxes directly measured by floating traps deployed in six cruises to the SEATS during October 2006–December 2008. The export fluxes were also estimated from the distributions of the three particle-reactive radionuclides in the euphotic layer. The goals of this study therefore are (1) to investigate the temporal variability of particle scavenging and removal processes, (2) to compare the geochemical behavior of ^{234}Th, ^{210}Pb, and ^{210}Po, and (3) to estimate the export flux of POC, PIC, and PN by $^{234}Th/^{238}U$, $^{210}Po/^{210}Pb$, and $^{210}Pb/^{226}Ra$ disequilibria in the euphotic layer of the northern South China Sea.

2 Material and methods

Seawater samples were collected by multiple cruises to the SEATS site in the South China Sea on board the R/V *Ocean Researcher I* or *III* (refer to Fig. 1 for the timeline of the cruises).

2.1 Seawater

Seawater was collected using either 10 l (R/V *Ocean Researcher III*) or 20 l (R/V *Ocean Researcher I*) Teflon-coated Go-Flo bottles mounted on a Sea-Bird® CTD (SBE 9/11) rosette system. Except for the first cruise (ORI-812), from which only ^{234}Th activity in unfiltered seawater was measured, filtration of large-volume seawater for the determination of ^{234}Th, ^{210}Pb, and ^{210}Po activities in filtrate and particles was carried out for all samples. At each sampling depth, 40 l seawater was collected and divided into one 20 l and two 10 l subsamples. The 20 l sample was used to determine ^{234}Th and the two 10 l were used to determine ^{210}Pb and ^{210}Po, respectively. Seawater was immediately pressure-filtered by compressed air through a pre-weighed 142 mm Nuclepore filter (0.45 µm) mounted in a Plexiglas filter holder. Filtrate and particles retained on the filter were processed and analyzed for ^{234}Th, ^{210}Pb, and ^{210}Po activities following the procedures described in Wei et al. (2009). Auxiliary standard parameters (e.g., nutrients, primary productivity, etc.) were measured by technical personnel of SEATS program. Nutrients were determined following the methods of Strickland and Parsons (1984). Primary productivity was determined by $Na^{14}HCO_3$ incubation of seawater samples collected from 6–8 depths in the upper 100 m.

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2.2 Floating trap

Particle interceptor trap (Wei et al., 1994) was deployed for the measurement of vertical fluxes of total mass (F_M), ^{234}Th (F_{Th}), ^{210}Po (F_{Po}), ^{210}Pb (F_{Pb}), particulate carbon (F_{PC}), particulate nitrogen (F_{PN}), and particulate organic carbon (F_{POC}), at three depths (30 m, 100 m, 160 m). At each depth, a set of sediment traps consisting of eight Plexiglas tubes filled with trap solution prepared from the mixture of filtered seawater and formaldehyde were snapped on a polypropylene cross frame, which is secured to a mooring line. It should be noted that radionuclide release into overlying waters in unpoisoned traps (even when deployed for less than 1 day) has been documented (Hung et al., 2010; Xu et al., 2011), which can make radionuclide estimates a minimum. The mooring array was left free drifting for 37–48 h. Upon recovery, the upper layer of seawater in the tube was siphoned off and the remaining trap solution was filtered through a pre-weighed 90 mm Nuclepore filter (0.45 µm pore size). The swimmers, mainly macrozooplanktons, were picked under a microscope. The samples then were rinsed by deionized water to remove salts. The filters were then dried in a desiccator and weighed for total mass flux calculation. The filters were analyzed for ^{234}Th, ^{210}Pb, and ^{210}Po activities following the procedures as particulate samples described in Wei et al. (2009).

The samples in tubes used for carbon and nitrogen analyses were filtered through pre-combusted Whatman 25 mm GF/F filters. After swimmers were picked, the filters were wrapped firmly into tin boats and loaded into the autosampler of EA elemental analyzer (EuroEA3000) for total carbon and nitrogen content for F_{PC} and F_{PN} calculations. The filters for organic carbon analysis were fumed for 24 h with concentrated HCl to remove calcium carbonate, then analyzed for carbon content for F_{POC} calculation. Fluxes of particulate inorganic carbon (F_{PIC}) were calculated by difference of F_{PC} and F_{POC}. The overall procedural error are better than ±2 % for both carbon and nitrogen determinations.

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3 Results

3.1 ^{234}Th, ^{210}Pb, and ^{210}Po in seawater

Vertical profiles of ^{234}Th activity concentrations of dissolved ($^{234}Th_d$) and particulate ($^{234}Th_p$) in in the upper 200 m at SEATS station, as well as related TSM concentrations, are shown in Fig. 2. It should be noted that only total ^{234}Th ($^{234}Th_t$) concentrations are available for October 2006 (ORI-812). Except for the subsurface layer in October 2007, the $^{234}Th_d$ values are significantly lower than those of ^{238}U, indicating active scavenging in the upper water column. The $^{234}Th_p$ values fall in the range from 0.05 to 0.75 dpm l^{-1} and sometimes show similar vertical distributions as TSM (Fig. 2). Vertical profiles of ^{210}Pb activity concentrations of dissolved ($^{210}Pb_d$) and particulate ($^{210}Pb_p$) in the upper 200 m at SEATS station, and related TSM concentrations from the filtration of ^{210}Pb samples are shown in Fig. 3. The line showing vertical distribution of ^{226}Ra was estimated from SiO_2 concentration by ^{226}Ra (dpm 100 l^{-1}) = 5.15 + 0.14 SiO_2 (µmol l^{-1}), which is based on the data determined at the site 300 km to the south of the SEATS by Nozaki and Yamamoto (2001). An excess of ^{210}Pb relative to ^{226}Ra in the upper water column is generally found. The $^{210}Pb_p$ concentrations are much lower than the $^{210}Pb_d$ concentrations, and show no systematic variation with depth. Vertical profiles of ^{210}Po activity concentrations of dissolved ($^{210}Po_d$) and particulate ($^{210}Po_p$ in the upper 200 m at SEATS station, and related TSM concentrations from the filtration of the ^{210}Po samples are shown in Fig. 4.

3.2 Sinking fluxes

Sinking fluxes of total particulate matter or mass (F_{Mass}), POC (F_{POC}), PIC (F_{PIC}), PN (F_{PN}), ^{234}Th (F_{Th}), ^{210}Pb (F_{Pb}), and ^{210}Po (F_{Po}) at the three deployment depths (30, 100, and 160 m) are shown in Fig. 5a–g, respectively. The average, range, and standard deviation of sinking fluxes of various parameters are summarized in Table 1. In

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general, all fluxes decreased with depth. Although the fluxes for all components, except ^{210}Pb, show the largest variability with a RSD of >70 % at 30 m, the correlations between fluxes are noticeably high, with correlation coefficients of >0.9 between all combinations of flux parameters. Except for the F_{PIC}, and F_{Po}, all fluxes varied within a relatively small range, with a RSD of ∼40 % at 100 m, the approximate euphotic depth at SEATS.

4 Discussions

4.1 Deficiencies and fluxes of ^{234}Th, ^{210}Po, and ^{210}Pb

There are very few previous open ocean studies that report ^{234}Th, ^{210}Pb, and ^{210}Po data determined from the same site. Among those few, Sarin et al. (1994) reported three vertical profiles of dissolved activity concentrations of ^{234}Th, ^{210}Pb, and ^{210}Po in the northeastern Arabian Sea. Wei and Murray (1991, 1994) compared the geochemical behavior of ^{234}Th, ^{210}Pb, and ^{210}Po in the Black Sea. Shimmield et al. (1995) measured the three radionuclides from the same seawater samples collected in the upper 500 m from the marginal ice zone in the Antarctica. Kim and Church (2001) presented dissolved and particulate ^{234}Th, ^{210}Pb, and ^{210}Po data determined on the same samples collected from the Sargasso Sea. Murray et al. (2005) reported a complete ^{234}Th, ^{210}Pb, and ^{210}Po data set measured from seawater samples and settling particles collected by floating traps deployed in the Equatorial Pacific.

Vertical profiles showing the daughter/parent nuclide ratios of ^{234}Th and ^{238}U, ^{210}Po and ^{210}Pb, and ^{210}Pb and ^{226}Ra in the upper 200 m obtained from the six cruises are given in Fig. 6. Relative to their parent radionuclides, deficiencies of ^{234}Th and ^{210}Po in the euphotic layer were commonly found, indicating evidence for an efficient removal process. A layer with excess ^{234}Th underlying the euphotic layer was found in January 2006, October 2007 and June 2007, which is the result of remineralization (Buesseler et al., 2008). The remineralization phenomenon is also shown as a layer of excess ^{210}Po found in all cruises, except in June and December 2008, corroborating

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the results from other regions (Bacon et al., 1976; Thomson and Turekian, 1976). As a result of atmospheric deposition, excess ^{210}Pb was commonly observed in the upper 200 m during all cruises, consistent with the findings of Obata et al. (2004) and Chung and Wu (2005).

Assuming steady-state and negligible physical transport, the flux of ^{234}Th out of the euphotic depth can be calculated by Eq. (1).

$$F'_{\mathrm{Th}} = \lambda_{\mathrm{Th}} \int_0^{100} ({}^{238}\mathrm{U} - {}^{234}\mathrm{Th_t})dz + V, \tag{1}$$

where F'_{Th} is the removal flux, λ_{Th} is radioactive decay constant (=0.0387 d^{-1}) and V is the net flux of ^{234}Th from physical transport. To justify the assumption of steady-state, the contribution of temporal changes in ^{234}Th activity concentrations in the mass balance needs to be assessed. It is found that the temporal change is only equivalent to 0.1–3.1 % of the contribution by radioactive decay, which is negligible in the flux calculation. The F'_{Th} caused by physical transport can be evaluated based on the unpublished ^{234}Th data obtained from 6 stations in March and July 2000, which covered 3 × 3 degree region of the study area. Following the procedures of Liang et al. (2003), the mean current velocity was estimated by averaging the archived shipboard ADCP data collected during 1991–2006. The results showed that horizontal transport by surface currents contributes only 1.5 % of ^{234}Th in the region. Negligible contribution of ^{234}Th from horizontal advection at the SEATS site corroborates to the work conducted by Chen et al. (2008) in the northern shelf of the South China Sea. Based on the distributions of ^{234}Th and ^{228}Th in the upper 500 m, Cai et al. (2006) also concluded that advection/diffusion is not a significant term in the scavenging model.

The flux of ^{210}Pb out of the euphotic depth can be calculated by Eq. (2).

$$F'_{\mathrm{Pb}} = \lambda_{\mathrm{Pb}} \int_0^{100} ({}^{226}\mathrm{Ra} - {}^{210}\mathrm{Pb_t})dz + I_{\mathrm{Pb}}, \tag{2}$$

where F'_{Pb} is the removal flux, λ_{Pb} is radioactive decay constant of ^{210}Pb

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(=$8.48\times10^{-5}\,\mathrm{d}^{-1}$) and I_{Pb} is the atmospheric ^{210}Pb deposition flux. The I_{Pb} is about 0.4 dpm cm^{-2} a^{-1} according to the model by Feichter et al. (1991). Based on ^{210}Pb chronology in ornithogenic sediments, Xu et al. (2011) estimated the atmospheric ^{210}Pb flux of 0.76 dpm cm^{-2} a^{-1} in the southern South China Sea. Here we assume an average I_{Pb} of 0.6 dpm cm^{-2} a^{-1} (16.4 dpm m^{-2} d^{-1}).

The flux of ^{210}Pb out of the euphotic depth is calculated by Eq. (3),

$$F'_{\mathrm{Po}} = \lambda_{\mathrm{Po}} \int_0^{100} (^{210}\mathrm{Pb_t} - {}^{210}\mathrm{Po_t})\,dz + I_{\mathrm{Po}}, \tag{3}$$

where F'_{Po} is the removal flux, λ_{Pb} is radioactive decay constant of ^{210}Po (=0.005 d^{-1}) and I_{Po} is the atmospheric ^{210}Po deposition flux, which is assumed to be 10 % of the atmospheric ^{210}Pb flux (Turekian et al., 1977). Since the spatial variation of ^{210}Po in the surface water of the South China Sea is small (Yang et al., 2006; Wei et al., 2011), as is the case for ^{234}Th, horizontal transport should be negligible. However, similar to the nutrients, the vertical profile of $^{210}\mathrm{Po_t}$ shows evidence for an increase in the thermocline layer, vertical diffusion may play a significant role in the mass balance of ^{210}Po in the euphotic layer (Sarin et al., 1994). If an eddy diffusion coefficient of 0.55 cm^2 s^{-1} (Nozaki and Yamamoto, 2001) and mean vertical gradient of ^{210}Po between 0 and 300 m were applied to the mass balance equation, the vertical fluxes of ^{210}Po due to upward diffusion would be equivalent to 2–8 % of the total flux, lower than that in oligotrophic North Pacific reported by Verdeny et al. (2008). Hence, the contribution of the vertical diffusion does not need to be included in the flux estimation.

The inventories, deficiencies, and removal fluxes of ^{234}Th, ^{210}Pb, and ^{210}Po in the euphotic layer are summarized in Table 2. Daughter nuclide deficiencies from their parent nuclides resulted in removal fluxes of 1.1×10^3–1.8×10^3 dpm m^{-2} d^{-1} and 7.1–40.2 dpm m^{-2} d^{-1} for ^{234}Th and ^{210}Po, respectively, from the euphotic layer. Due to atmospheric input, an excess of ^{210}Pb relative to ^{226}Ra is commonly observed in the upper water column. As is apparent, the atmospheric input is the dominant source term in Eq. (2), thus, the F'_{Pb} is close to the constant atmospheric ^{210}Pb flux, 16.4 dpm m^{-2} d^{-1}. No systematic variations were found for the temporal values of F'_{Th}, F'_{Pb}, and F'_{Po}. Based on the ^{210}Pb and ^{210}Po content in the surface water, Nozaki et al. (1998) predicted a ^{210}Po/^{210}Pb ratio of 0.58 in sinking particles from the South China Sea. The ^{210}Po/^{210}Pb ratio 0.4–2.5 in the sinking particles exiting from the euphotic layer that was estimated by this study is within the range of the ^{210}Po/^{210}Pb ratio of 0.4–4.5 measured by our sediment traps.

4.2 Trapping efficiency determined by ^{234}Th, ^{210}Po, and ^{210}Pb

It is known that the sediment traps can show trapping efficiencies different from 1 (or 100 %), due to hydrodynamic and swimmer effects (Buesseler, 1991; Buesseler et al., 2007), as well as selective dissolution effects (Hung et al., 2010; Xu et al., 2011). To evaluate the trapping efficiency of the floating trap, the ^{234}Th/^{238}U disequilibria in the upper water column are commonly used to assess the trapping efficiency of the floating trap. The ^{234}Th flux measured by the sediment trap deployed at specific depth should be equal to the removal flux calculated from the deficiency in the water column above the depth. Ideally, following the same concept, the comparison of measured with estimated ^{210}Pb and ^{210}Po fluxes can also be used as an indicator of trapping efficiency of the floating traps. Hence, we have calculated the trapping efficiencies of the floating traps by the ratios of measured and predicted fluxes of ^{234}Th, ^{210}Pb, and ^{210}Po at the three depths. The trapping efficiencies for the six cruises and the average values at the three deployment depths are shown in Table 3. The trapping efficiencies for the traps deployed at 100 m, the euphotic depth, range between 1.06 ± 0.2 and 3.99 ± 3.23 based on ^{234}Th and ^{210}Po data, respectively. Considering the uncertainties associated with sediment trap deployments, these trapping efficiencies are acceptable, as most of the values at 100 and 160 m are close to 1 (Table 3). A large range of trapping efficiency for floating traps, with the ratio of measured and modeled fluxes ranging from 0.1 to 20, is found (Buesseler, 1991; Stewart et al., 2007; Wei and Murray, 1992; Buesseler et al., 2007). A trapping efficiency close to unity and a small variability for the traps deployed at 100 m increase the reliability of the export flux assessment (Buesseler et al., 2007). While it is noted that the traps deployed at 30 m, the mixed depth, generally give significantly higher radionuclide fluxes than expected values and reveal a large variability, they also experience greater hydrodynamic effects, which may be the cause for these phenomena. It is also worth noting that, except for the 100 m trap of July 2007, all trapping efficiencies estimated by F_{Po} and F'_{Po} are greater than unity, which may be related to the fast regeneration rate of ^{210}Po due to particle remineralization in the euphotic layer. Compared with the concentrations of trace metal in the same samples (Ho et al., 2010), it was found ^{210}Po content is highly correlated ($r > 0.8$, $n = 11$) with biophilic elements, P, S, Ca, Zn, Mo, and Cd. Previous studies have shown that ^{210}Po, a sulfur analog found mostly associated with proteins (Fisher et al., 1983), shows higher affinity toward organic particles than ^{210}Pb (Bacon et al., 1976; Shannon et al., 1970), and thus, ^{210}Po is more sensitive to particle decomposition processes than ^{234}Th (Stewart et al., 2007), which is mostly surface-bound (Santschi et al., 2006).

4.3 Export fluxes of POC, PIC, and PN estimated by radionuclide tracers

Export fluxes of POC, PIC, and PN can be indirectly estimated if the ratio of the concentration of carbon and nitrogen to the radionuclides in sinking particles are known. Since the first study in the Panama Basin (Murray et al., 1989), ^{234}Th has been extensively used in the past two decades as a powerful tracer for estimating the export production (i.e., POC flux) from the euphotic layer of the ocean (Buesseler et al., 2006 and refereces therein). For the same purpose, ^{210}Pb and ^{210}Po, as additional tracers, were used to constrain the determination of the export production in the euphotic layer (Shimmield et al., 1995; Stewart et al., 2007; Murray et al., 2005; Friedrich and Rutgers van der Loeff, 2002; Kim and Church, 2001; Buesseler et al., 2008). In the context of treating particle-reactive radionuclides as proxies for POC flux, export production or sinking flux of PIC and PN from the euphotic layer can also be estimated by multiplying the flux of radionuclides, which is obtained from the mass balance of daughter radionuclide relative to its parent radionuclide in the euphotic layer, by the ratio of POC (or PIC, PN) and radionuclide in sinking (or sinkable) particles (Murray et al., 2005; Buesseler, 1998; Bacon et al., 1996), i.e.,

$$F'_{\text{POC-RN}} = F'_{\text{RN}} \cdot \left(\frac{\text{POC}}{\text{RN}}\right), \tag{4}$$

$$F'_{\text{PIC-RN}} = F'_{\text{RN}} \cdot \left(\frac{\text{PIC}}{\text{RN}}\right), \tag{5}$$

$$F'_{\text{PN-RN}} = F'_{\text{RN}} \cdot \left(\frac{\text{PN}}{\text{RN}}\right), \tag{6}$$

where RN is the abbreviation of radionuclide (^{234}Th, ^{210}Pb, ^{210}Po), $F'_{\text{POC-RN}}$, $F'_{\text{PIC-RN}}$, and $F'_{\text{PN-RN}}$ are fluxes of POC, PIC, and PN estimated by RN as proxy, respectively, F'_{RN} is the flux of radionuclide calculated from the deficiencies of RN in the euphotic layer by Eqs. (1)–(3), and POC/RN , PIC/RN, and PN/RN is the ratio of POC, PIC, and PN and the radionuclides in sinking (or sinkable) particles collected at the euphotic depth. Equations (4) and (6) were previously used for ^{234}Th data to estimate the export flux of POC and PN from the euphotic layer of the Atlantic Ocean (Buesseler et al., 1992; Amiel et al., 2002) and the Pacific Ocean (Murray et al., 1996). Limited by the scarcity of sediment trap data on radionuclide to PIC ratios, very few applications of Eq. (5) on PIC export exist. The only application of using ^{234}Th as a PIC proxy is Bacon et al. (1996), who estimated PIC export fluxes based on PIC/^{234}Th ratios in filtered particles in the Equatorial Pacific.

The POC/RN, PIC/RN, and PN/RN in sinking particles collected at the three depths are shown in Fig. 7. The POC/^{234}Th ratios in sinking particles decreased with depth, with average values of 23.9 ± 16.3, 8.7 ± 3.2, and $4.8 \pm 1.8\,\mu\text{mol dpm}^{-1}$ at 30, 100, and 160 m, respectively. The decreasing trend with depth can be attributed to preferential remineralization of organic carbon when particles settle through the water column (Rutgers van der Loeff et al., 2002; Buesseler et al., 2006). Decreasing of POC/^{234}Th ratio with depth were also found for suspended particles in the northern South China

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Sea by Cai et al. (2008) and Chen et al. (2008). A larger variability of this ratio was found at the mixed layer depth, which may be caused by a more active biological activity in the surface layer. It is noted here that the POC/^{234}Th ratio in sinking particles collected at the euphotic depth falls in a small range and values are similar to those reported for more productive regions, e.g., Northern Atlantic (Buesseler et al., 1992) and higher than in the oligotrophic open ocean, e.g., HOTS and BATS (see the compilation of Buesseler et al., 2006). Due to the low scatter of POC/^{234}Th ratio at the euphotic depth, the export production based on Eq. (4) tends to be less variable (Buesseler et al., 2006). Except during July 2007, the POC/^{210}Pb ratio in sinking particles also shows a decreasing trend with depth (Fig. 7b). The average POC/^{210}Pb ratios are 1088 ± 403, 686 ± 352, and 321 ± 131 µmol dpm^{-1} at 30, 100, and 160 m, respectively. These values are comparable to POC/^{210}Pb ratios of 80–268 µmol dpm^{-1} in sinking particles collected by sediment traps at 150 m off the east coast of the United States (Biscaye et al., 1988) and 50–489 µmol dpm^{-1} in the northwestern Mediterranean (Tateda et al., 2003). The POC/^{210}Po ratios in the sinking particles are highly variable, ranging from 120 to 1200 µmol dpm^{-1}, similar to ratios in the sinking particles collected in the coastal Mediterranean Sea (Tateda et al., 2003; Stewart et al., 2007). However, compared to open ocean values (Verdeny et al., 2008 and references therein), our data fall on the high end of these POC/^{210}Po ratios. Different from ^{234}Th and ^{210}Pb, no systematic trends with depth were found for POC/^{210}Po ratios.

Values of the PIC/^{234}Th (Fig. 7d) and PN/^{234}Th (Fig. 7g) ratios show temporal and vertical variations similar to those of POC/^{234}Th, suggesting no preferential association of ^{234}Th with different components of biological particles. The average PIC/^{234}Th ratios are 8.9 ± 6.3, 2.8 ± 1.1, and 1.9 ± 2.4 µmol dpm^{-1} at 30, 100, and 160 m depth, respectively. These ratios are comparable to the limited published sediment trap data, e.g., 0.6 µmol dpm^{-1} at 300 m in the Northwest Mediterranean (Szlosek et al., 2009). The PN/^{234}Th ratios in the lower euphotic zone range between 0.6 and 1.4 µmol dpm^{-1}, which is similar to 0.7–1.3 µmol dpm^{-1} in the Northern Atlantic (Buesseler et al., 1992). Like ^{234}Th data, the PIC/^{210}Pb (Fig. 7e) and PN/^{210}Pb (Fig. 7h) ratios also show

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temporal and vertical variations similar to those of POC/^{210}Pb. The PIC/^{210}Pb ratios are 433 ± 252, 279 ± 51, and 108 ± 96 µmol dpm^{-1} at 30, 100, and 160 m, respectively. The ratios at the lower euphotic depth were very similar to 135–518 µmol dpm^{-1} at 150 m in the Northwest Mediterranean (Tateda et al., 2003). The PN/^{210}Pb ratios obtained from this study are comparable with the ratios in sinking particles from the euphotic layer in the Mid Atlantic Bight (21–279 mmol dpm^{-1}, Biscaye et al., 1988) and the Northwest Mediterranean (4–126 mmol dpm^{-1}, Tateda et al., 2003). Both PIC/^{210}Po (Fig. 7f) and PN/^{210}Po (Fig. 7i) ratios show large seasonal and vertical variability. The average PIC/^{210}Po and PN/^{210}Po ratios range from 91 to 433 µmol dpm^{-1} and 14 to 50 µmol dpm^{-1}, respectively. Compared to the trap data and PIC, PN, and ^{210}Po data of Tateda et al. (2003), our results fall in the same range as the PIC/^{210}Po and PN/^{210}Po ratios from the coastal Mediterranean, i.e., 94–1115 µmol dpm^{-1} and 7–148 µmol dpm^{-1}, respectively,.

Using the ratios of POC, PIC, and PN to the three radionuclides in sinking particles of the 100 m trap, the export fluxes of organic carbon, inorganic carbon, and nitrogen via particle settling from the euphotic layer can be calculated by Eqs. (4)–(6) and plotted for different cruises in Fig. 8. Along with the estimated fluxes based on the three radionuclide proxies, directly measured fluxes of POC, PIC, and PN by sediment traps deployed at 100 m are also shown in the figure. The export fluxes of POC, PIC, and PN from the euphotic layer of the northern South China Sea determined by various approaches are discussed in the following section.

4.4 Comparison of export fluxes by different approaches

As can be seen from Fig. 8, values of $F'_{\text{POC-Th}}$ display a remarkable resemblance to F_{POC} values, both in terms of magnitude and temporal variation. The $F'_{\text{POC-Th}}$ ranges from 9.6 mmol-C m^{-2} d^{-1} to 21.0 mmol-C m^{-2} d^{-1}and the F_{POC} ranges from 9.8 to 18.5 mmol-C m^{-2} d^{-1}. The temporal variation of both F_{POC} and $F'_{\text{POC-Th}}$ generally corroborates with the seasonal variation of primary productivity at SEATS (Tseng et

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al., 2005). In the region further south to SEATS, Cai et al. (2008), using a 3-D scavenging model, estimated the POC export ranged from −10.7 to 12.6 mmol-C m^{-2} d^{-1} calculated from ^{234}Th deficiencies and POC/^{234}Th ratios in pump-collected particles. They attributed the negative values at some stations in the study area to large input of particulate matter by horizontal transport. Although ^{210}Pb has been found useful in estimating the POC sinking flux in the deep ocean (Moore and Dymond, 1988), there are very few studies using the ^{210}Pb/^{226}Ra disequilibrium to estimate the export production in the euphotic layer because a reliable atmospheric ^{210}Pb flux is not available to better constrain the source term in Eq. (2), which is much greater than the input from the radioactive decay of ^{226}Ra. Nonetheless, assuming the atmospheric ^{210}Pb flux of 0.6 dpm cm^{-2} a^{-1} (Feichter et al., 1991; Xu et al., 2010), the $F'_{\text{POC-Pb}}$ of 7.2–21.3 mmol-C m^{-2} d^{-1} can be estimated, which is within the range of F_{POC} and $F'_{\text{POC-Th}}$ values. Since a constant atmospheric ^{210}Pb flux is assumed, the F_{Pb}' values show no seasonal variation (Table 2). Hence, the temporal variations of the $F'_{\text{POC-Pb}}$ are resulting from the POC/^{210}Pb ratio in sinking particles, which may be affected by particle composition. The $F'_{\text{POC-Po}}$ based on the ^{210}Po-^{210}Pb system showed the largest range of POC export, ranging from 1.8 to 20.3 mmol-C m^{-2} d^{-1}, among the three pairs of disequilibria.

Verdeny et al. (2009) recently presented a review of previous studies using ^{234}Th and ^{210}Po as dual tracers to estimate the export production in the ocean. A discrepancy between the POC fluxes estimated by ^{210}Po-^{210}Pb and ^{210}Po-^{210}Pb disequilibria was commonly found. For example, the export production deduced from the ^{210}Po deficiency was two folds higher than that estimated from the ^{234}Th-^{238}U disequilibrium in the Atlantic (Sarin et al., 1994). On the other hand, Shimmield et al. (1995) found the export production estimated by the $F'_{\text{POC-Po}}$ was an order of magnitude lower than that estimated from the $F'_{\text{POC-Th}}$ values in the marginal ice zone of the Antarctica. In the Equatorial Pacific, Murray et al. (2005) reported a generally higher and more variable $F'_{\text{POC-Po}}$ than $F'_{\text{POC-Th}}$ values. Stewart et al. (2007) obtained the $F'_{\text{POC-Th}}$ and $F'_{\text{POC-Po}}$

values of 3.8–8.9 mmol-C m^{-2} d^{-1} and 1.2–2.6 mmol-C m^{-2} d^{-1}, respectively, from the euphotic layer of the northwestern Mediterranean. Recently, Buesseler et al. (2008) used ^{234}Th and ^{210}Po proxies to estimate POC export fluxes from the euphotic layer of the Sargasso Sea and found the $F'_{\text{POC-Po}}$ is about two folds higher than the $F'_{\text{POC-Th}}$. Hence, considering the intrinsic difference in half-life, source function, and geochemical characteristics among the three radionuclides and the uncertainty associated with the scavenging model, the POC fluxes estimated by these proxies in our work can be reliably accepted as representative of the export production in the northern South China Sea.

There is some literature that estimated the export production by various other approaches in the northern South China Sea. These approaches include a ^{228}Ra/NO_3 coupled model (Nozaki and Yamamoto, 2001), biochemical modeling (Liu et al., 2002), carbon budgeting (Chou et al., 2006), ^{15}N incubation measurements of new production (Chen and Chen, 2004, 2005), and phosphorus flux measurements by sediment traps (Ho et al., 2009). The export fluxes by these approaches are summarized in Table 4. Among these studies, based on the P fluxes measured by the sediment traps deployed at the depth underlying the euphotic layer, Ho et al. (2009) obtained the largest range of export fluxes (4.2–70.8 mmol-C m^{-2} d^{-1}) at the SEATS. Some anomalously high fluxes reported by Ho et al. (2009) may be caused by over-trapping of their sediment traps, which were deployed at 160 m on a long mooring line. It can thus be concluded that the export production can be reasonably estimated by using ^{234}Th, ^{210}Pb, and ^{210}Po as carbon proxies in sinking particles. In a more recent paper, Ho et al. (2011) evaluated the POC fluxes estimated from particulate phosphorus fluxes from the moored traps deployed at 120 m and found consistent results with the observations by the floating traps.

The correlation of POC exports obtained by various methods and integrated primary production during the six cruises is shown in Fig. 9. The primary production varied from 9.5 mmol-C m^{-2} d^{-1}in July 2007 to 46.1 mmol-C m^{-2} d^{-1} in October 2007. The export efficiency can be indicated by the ratio of the POC exports and the primary production (Buesseler, 1998), which showed a large range between 0.04 and 2.17 with an average ratio of 0.53. The unreasonably high ratios greater than unity were found for ORIII-1239, from which the primary production was suspected to be underestimated based on the vertical profile of primary productivity. It is found that most of data fall in the range of 0.2 and 0.7, which implies that export efficiency is high in the South China Sea.

The values of F_{PIC} range from 2.1 to 9.5 mmol-C m^{-2} d^{-1} (Fig. 8b), which is significantly higher than in other regions (Berelson et al., 2007). The temporal variation of the F_{PIC} is generally predicted by the fluxes estimated from the other proxies. Based on the ^{234}Th deficiency and the PIC/^{234}Th ratio in pump-collected large particles, Bacon et al. (1996) estimated a PIC export of 0.45–0.80 mmol-C m^{-2} d^{-1} at 150 m and 0.54–0.71 mmol-C m^{-2} d^{-1} at 200 m in the Equatorial Pacific. High F_{PIC} values at SEATS may be attributed to the dominant coccolithophores in the algal community during the winter season in the South China Sea (Chen and Chen, 2005), which enhances the sinking flux due to a ballast effect. Indeed, microscopic examination of trap particles shows that skeletal remains of coccolithophorids and diatoms are abundant during the winter cruises (ORI-821 and ORI-887) (Ho et al., 2010). Calculated from the temporal change of the inventory of alkalinity in the euphotic layer during October 2006 and December 2008, a PIC production ranging from 0.4 to 11.4 mmol-C m^{-2} d^{-1} can be estimated. Direct measurements of Ca concentration from the same samples shows F_{PIC} ranging from 1.6 to 6.2 mmol-C m^{-2} d^{-1} (Ho et al., 2010). All the F_{PIC}, $F'_{PIC\text{-}Th}$, $F'_{PIC\text{-}Pb}$, and $F'_{PIC\text{-}Po}$ values are thus within the PIC production estimates, indicating the PIC export flux was reliably estimated.

The F_{PN} values measured at 100 m of the SEATS ranged from 1.5 mmol-N m^{-2} d^{-1} in July 2007 to 4.1 mmol-N m^{-2} d^{-1} in December 2008, very close to the flux of ∼2 mmol-N m^{-2} d^{-1} derived from a $^{228}Ra/NO_3$ coupled model at the site about 300 km to the south of our station (Nozaki and Yamamoto, 2001). Except for the $F'_{PN\text{-}Po}$ in October 2006 and January 2007, the values of $F'_{PN\text{-}Th}$, $F'_{PN\text{-}Pb}$, and $F'_{PN\text{-}Po}$ are close to the F_{PN} within a factor of three. The export of nitrogen via particle settling in the northern South

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China Sea is also close to the measured PN flux of 1.2–2 mmol-N m^{-2} d^{-1} in the North Atlantic (Martin et al., 1993) and the northeast Pacific (Wong et al., 1999).

5 Conclusions

Temporal data of dissolved and particulate concentrations and fluxes of the particle-reactive radionuclides, ^{234}Th, ^{210}Pb, and ^{210}Po, in the upper water column of the time-series station, SEATS (18° N, 116° E) were obtained during 2006 to 2008. All fluxes (total mass, ^{234}Th, ^{210}Pb, ^{210}Po, POC and PIC) measured at the euphotic depth were lower in summer-fall and higher in winter-spring, reflecting the seasonal variability of biological pumping. Using the three radionuclides as proxies, the disequilibria of $^{234}Th/^{238}U$, $^{210}Pb/^{226}Ra$, and $^{210}Po/^{210}Pb$ were combined with their ratios to POC, PIC, and PN in the sinking particles collected at the euphotic depth to estimate export production in the northern South China Sea. Our results showed that estimated fluxes by the radionuclide proxies are within the range of those measured by sediment traps, which ranged from 1.8 to 21.3 mmol-C m^{-2} d^{-1}, from 1.6 to 9.1 mmol-C m^{-2} d^{-1}, and from 0.2 to 4.5 mmol-N m^{-2} d^{-1}, for POC, PIC, and PN, respectively.

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Table 1. Range, average, and standard deviation of sinking fluxes of total mass (F_{Mass}), POC (F_{POC}), PIC (F_{PIC}), PN (F_{PN}), ^{234}Th (F_{Th}), ^{210}Pb (F_{Pb}), and ^{210}Po (F_{Po}) at three depths during October 2006 and December 2008.

Depth	F_{Mass}	F_{POC}	F_{PIC}	F_{PN}	F_{Th}	F_{Pb}	F_{Po}
m	mg m^{-2} d^{-1}	mmol m^{-2} d^{-1}			dpm m^{-2} d^{-1}		
30	441–3548 1776 ± 1249	8.6–59.1 29.6 ± 19.0	3.7–23.8 11.8 ± 9.1	2.7–10.1 6.0 ± 3.1	694–3395 1778 ± 1187	17.4–36.4 27.6 ± 7.8	33.6–127.4 72.3 ± 45.5
100	597–1215 861 ± 256	9.8–18.5 14.4 ± 3.3	2.1–9.5 5.2 ± 3.1	1.5–4.1 2.4 ± 0.9	1201–2160 1690 ± 399	7.5–33.8 23.0 ± 9.6	10.6–107.6 49.2 ± 39.4
160	285–1095 638 ± 309	3.2–10.8 6.9 ± 2.8	0.4–8.3 2.6 ± 3.2	0.5–1.7 0.9 ± 0.5	1046–1864 1432 ± 304	14.0–37.0 23.1 ± 10.7	11.5–42.4 30.2 ± 15.2

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Table 2. Inventories of radionuclides in dissolved and particulate phases of seawater, deficiencies of total activity of daughter with respect to parent radionuclides, and their removal fluxes from the euphotic layer.

Cruise	Time	Inventory (dpm m^{-2})						Deficiency (dpm m^{-2})			Removal flux (dpm m^{-2} d^{-1})		
		$^{234}Th_d$	$^{234}Th_p$	$^{210}Pb_d$	$^{210}Pb_p$	$^{210}Po_d$	$^{210}Po_p$	($^{238}U-^{234}Th_t$)	($^{226}Ra-^{210}Pb_t$)	($^{210}Pb_t-^{210}Po_t$)	F'_{Th}	F'_{Pb}	F'_{Po}
ORI-812*	Oct 2006	191 000		5924		4828		50 307	−66	1097	1447	16.4	7.1
ORI-821	Jan 2007	149 524	37 578	7534	985	3266	2564	55 080	−2925	2690	1584	16.2	15.1
ORIII-1239	Jul 2007	161 069	14 795	7069	415	3529	744	64 557	−1868	3210	1857	16.3	17.7
ORI-845	Oct 2007	182 687	18 436	5394	741	3465	515	38 424	−278	2156	1105	16.4	12.4
ORI-866	Jun 2008	152 631	34 811	9555	1003	4792	1325	55 640	−4769	4441	1600	16.0	23.9
ORI-887	Dec 2008	156 938	24 882	8053	894	3744	1076	56 537	−3366	7702	1626	16.2	40.2

* Inventories of ^{234}Th, ^{210}Pb, and ^{210}Po represent total values since the radionuclide activities were measured on unfiltered sample.

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Table 3. Trapping efficiencies estimated by the ratios of measured and modeled fluxes of ^{234}Th, ^{210}Pb, and ^{210}Po from the three depths for all cruises. Average value, standard deviation, and relative standard deviation for each depth are listed.

	Depth (m)	Oct 2006	Jan 2007	Jul 2007	Oct 2007	Jun 2008	Dec 2008	Av.	s.d.	r.s.d.
$F_{Th}:F'_{Th}$	30	1.12	5.29	0.79	8.39	0.98	0.75	2.89 ±	3.22	112 %
	100	0.83	1.33	0.84	1.19	1.12	1.06	1.06 ±	0.20	19 %
	160		0.91	0.37	1.26	0.81	1.18	0.91 ±	0.35	39 %
$F_{Pb}:F'_{Pb}$	30		1.33	0.83	2.60	1.61	1.02	1.48 ±	0.69	47 %
	100		0.87	0.69	2.29	3.21	0.89	1.59 ±	1.11	70 %
	160		1.37	1.38	1.27	1.69	1.15	1.37 ±	0.20	15 %
$F_{Po}:F'_{Po}$	30		15.02	2.77	17.85	4.11	5.26	9.00 ±	6.91	77 %
	100		7.64	0.75	1.15	3.35	7.07	3.99 ±	3.23	81 %
	160		2.77	0.39	4.22	1.36	7.84	3.32 ±	2.92	88 %

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Table 4. Export production estimated by various methods in the northern South China Sea.

Method	mmol-C m^{-2} d^{-1}	Reference	Note
^{228}Ra-NO_3 coupling	26.5	Nozaki and Yamamoto (2001)	Estimated diffusive NO_3 flux at 100 m
Biochemical modeling	1.7–3.5	Liu et al. (2002)	Estimated POC flux at 125 m
^{15}N new production	3.8–6.7	Chen (2005)	
Carbon budget	2.9–6.7	Chou et al. (2006)	Net community production at mixed depth
P flux by sediment trap	4.2–71.8	Ho et al. (2009)	Measurement at 160 m
Floating trap	9.8–18.5		Measurement at 100 m
^{234}Th proxy	9.6–21.0	This study	
^{210}Pb proxy	7.2–21.3		
^{210}Po proxy	1.8–20.3		

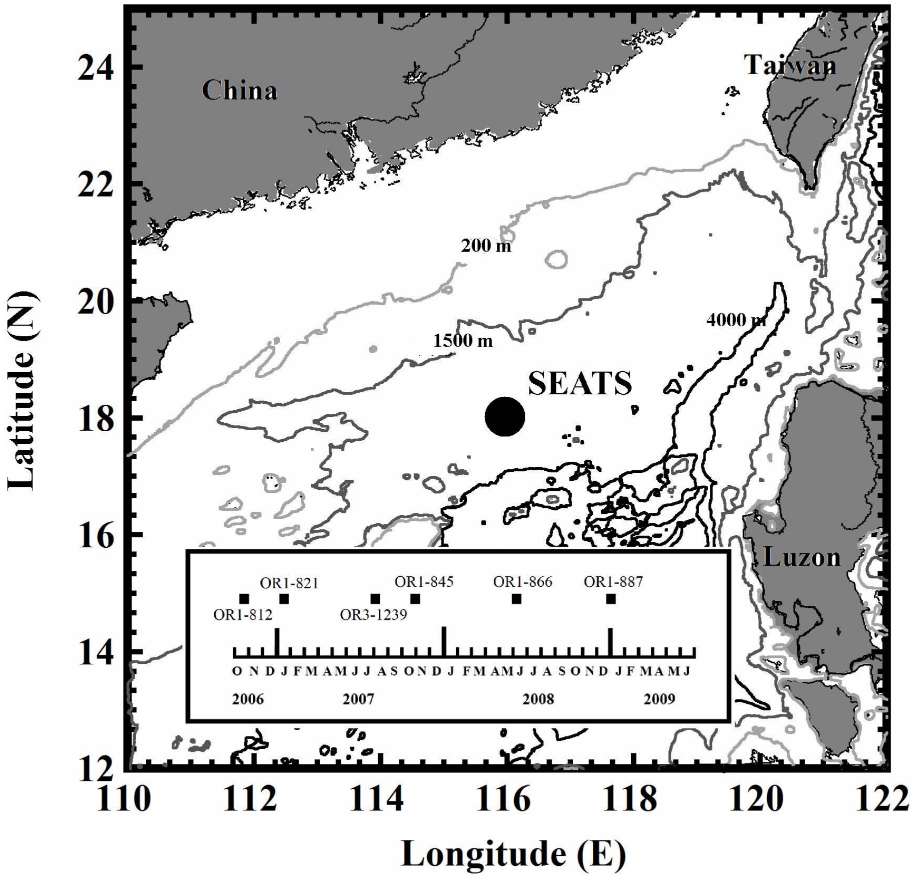

Fig. 1. Location of SEATS station in the South China Sea. Inserted box shows the timeline of the six cruises to the station.

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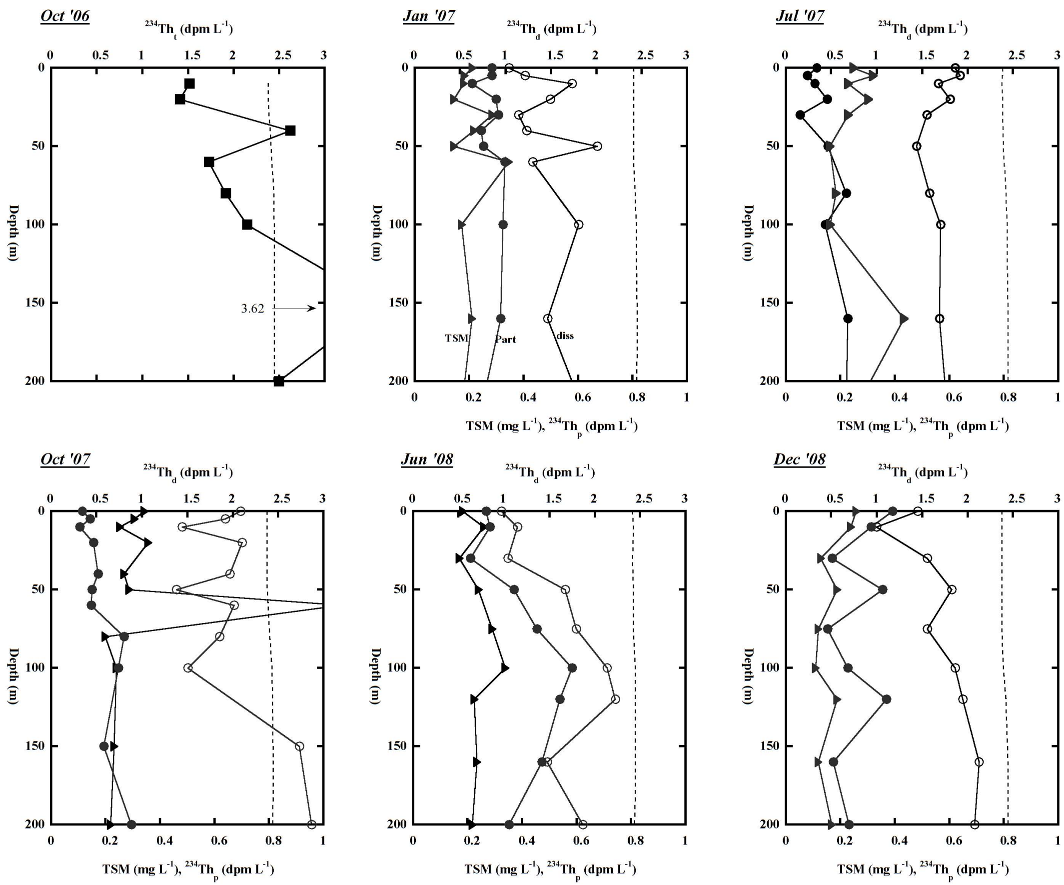

Fig. 2. Vertical distributions of dissolved (open circle) and particulate (solid circle) ^{234}Th, TSM concentration (solid triangle), and the ^{238}U (thick dotted line) calculated from salinity (Ku et al., 1977). Note only total ^{234}Th (solid square) is available for ORI-821 (October 2006).

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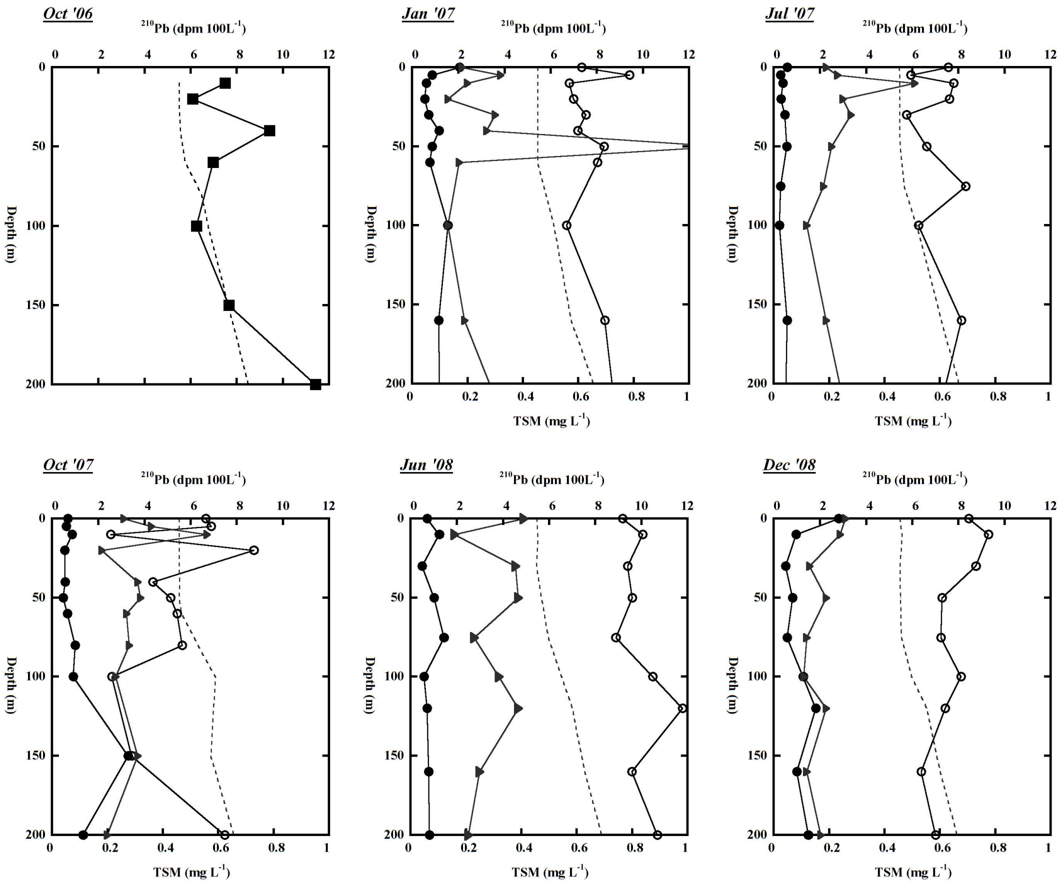

Fig. 3. Vertical distributions of dissolved (open square) and particulate (solid square) ^{210}Pb, TSM concentration (solid circle), and the ^{226}Ra (thick dotted line) estimated from SiO_2 (Nozaki and Yamamoto, 2001). Note only total ^{210}Pb (solid triangle) is available for ORI-821 (October 2006).

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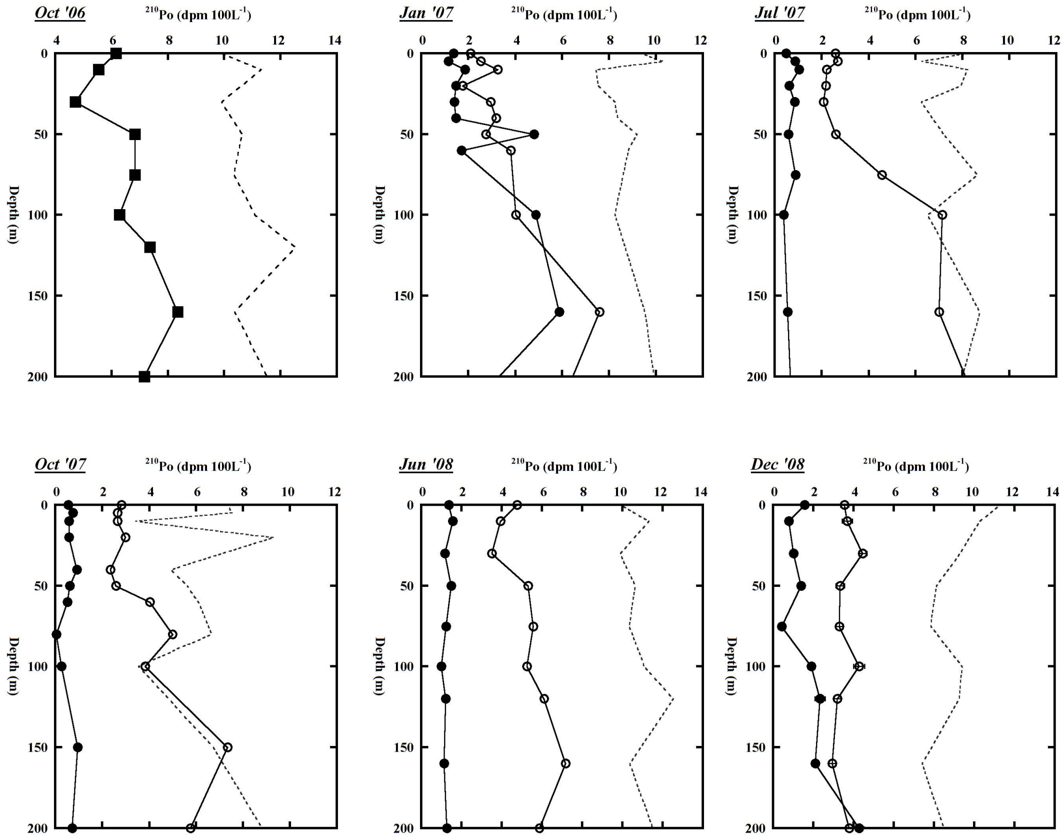

Fig. 4. Vertical distributions of dissolved (open square) and particulate (solid square) ^{210}Po, TSM concentration (solid circle), and total ^{210}Pb (thick dotted line) calculated by the sum of $^{210}Pb_d$ and $^{210}Pb_p$. Note only total ^{210}Po (solid triangle) is available for ORI-821 (October 2006).

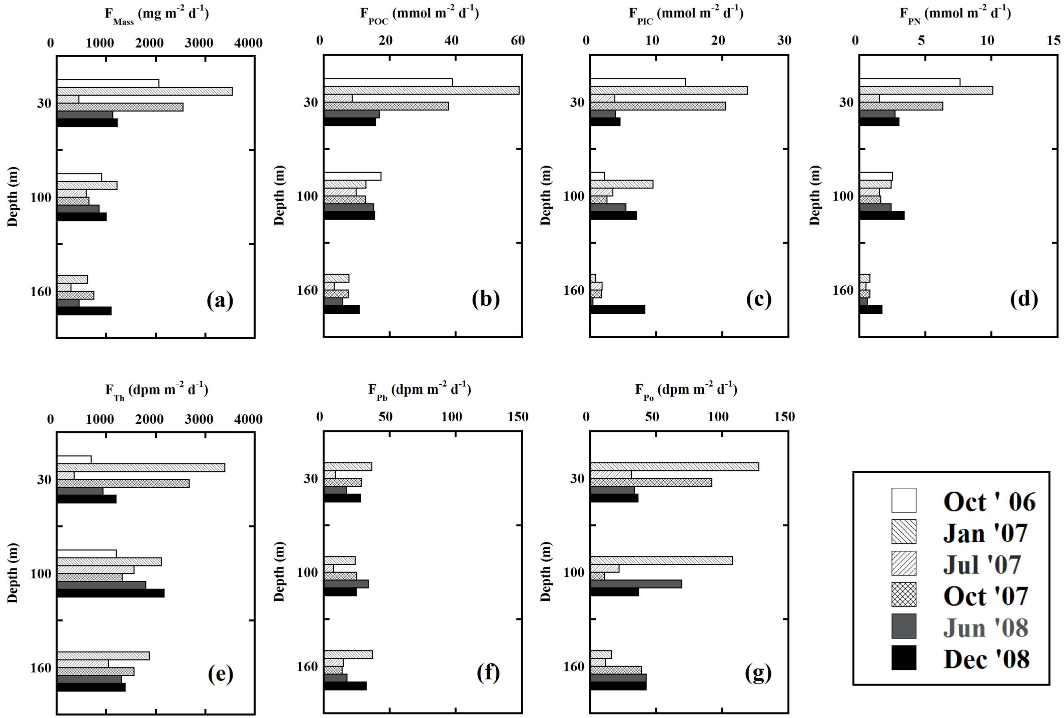

Fig. 5. Fluxes of **(a)** total mass (F_{mass}), **(b)** particulate organic carbon (F_{POC}), **(c)** particulate inorganic carbon (F_{PIC}), **(d)** particulate nitrogen (F_{PN}), **(e)** ^{234}Th (F_{Th}), **(f)** ^{210}Pb (F_{Pb}), and **(g)** ^{210}Po (F_{Po}) measured by the floating traps.

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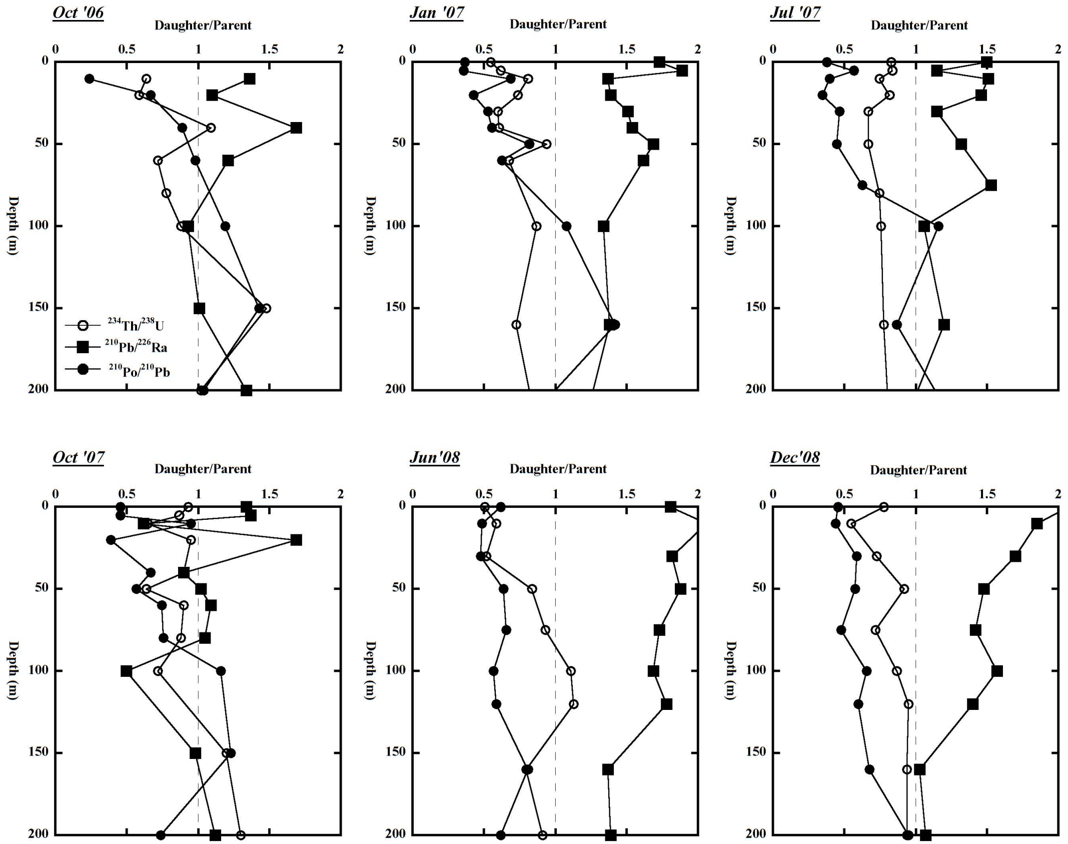

Fig. 6. Vertical distributions of the ratios of total activities of daughter and parent radionuclides obtained from the six cruises. The dotted line represents the secular equilibrium of daughter-parent relationship.

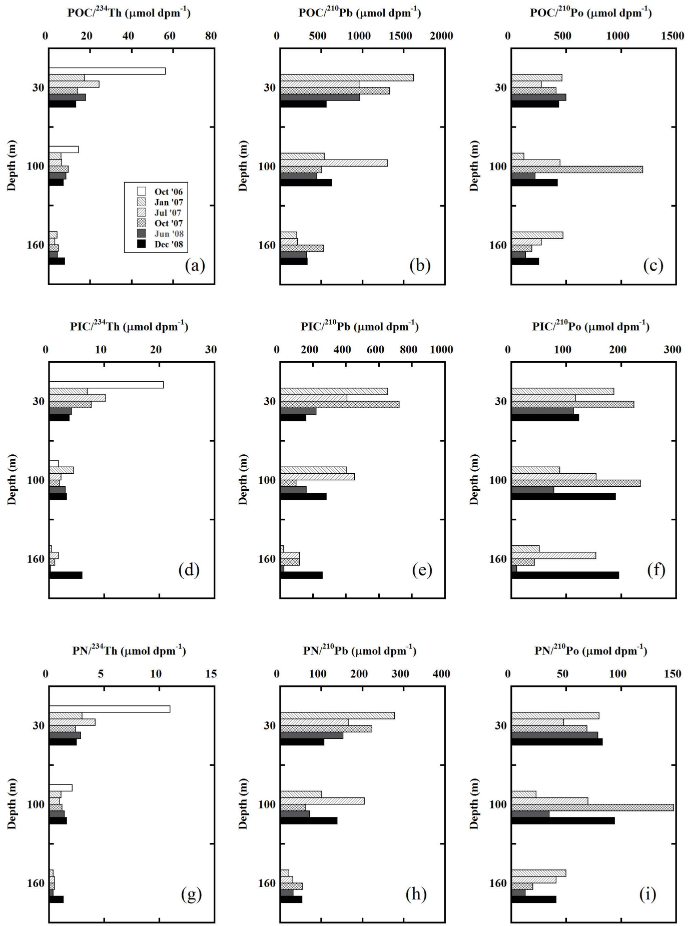

Fig. 7. Ratios of **(a–c)** POC and RN, **(d–f)** PIC and RN, and **(g–i)** PN and RN in settling particles collected by floating traps at the three depths of SEATS.

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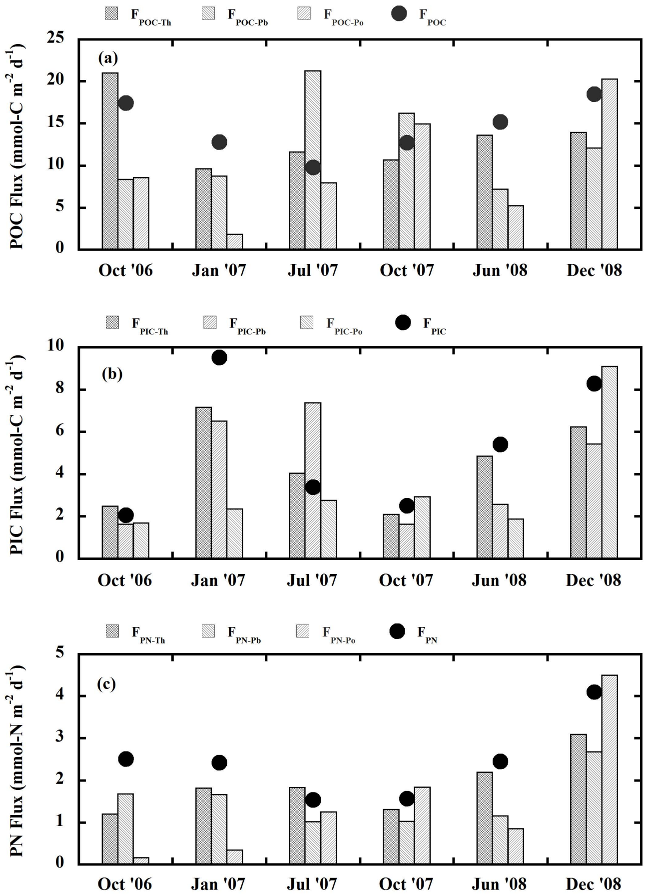

Fig. 8. Export fluxes of **(a)** POC, **(b)** PIC, and **(c)** PN estimated by ^{234}Th, ^{210}Pb, and ^{210}Po as proxy (bars) and measured by the floating traps (circles) from the euphotic layer.

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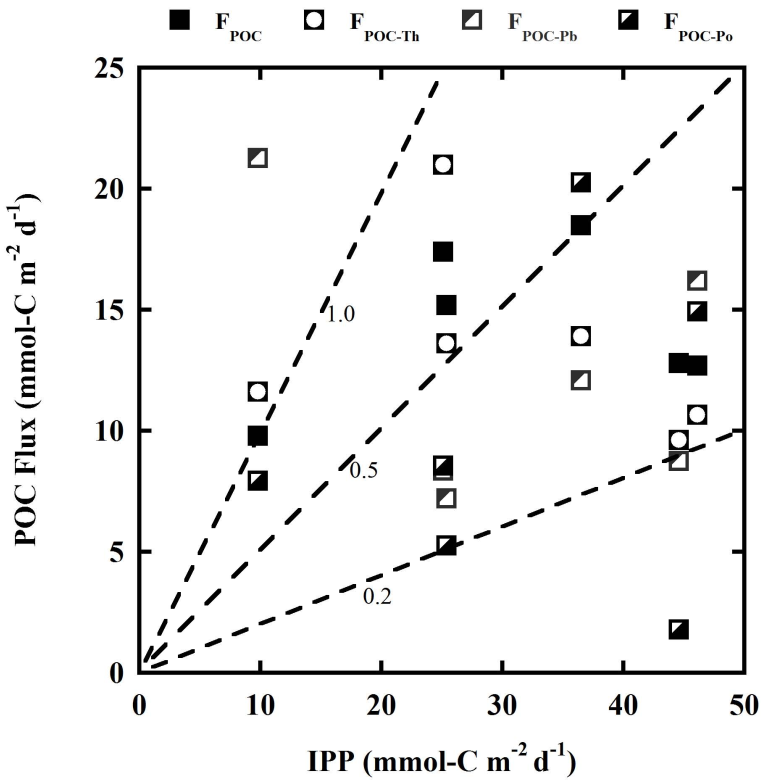

Fig. 9. Correlation of F_{POC}, $F'_{POC\text{-}Th}$, $F'_{POC\text{-}Pb}$, and $F'_{POC\text{-}Po}$ with integrated primary productivity (IPP). Export ratios of 0.2, 0.5, and 1.0 are shown by dashed lines.

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