Nearshore Scavenging Phenomenon Elucidated by $^{234}\text{Th}/^{238}\text{U}$ Disequilibrium in the Coastal Waters off Western Taiwan

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Dissolved and particulate ^{234}Th activities in surface seawater were determined at 27 stations along the coastline of western Taiwan during 19–23 November 2004. Contrasting scavenging settings were observed between the northern and southern regimes of the nearshore water off western Taiwan, separated by the Cho-Shui River. The northern regime is characterized by a large quantity of suspended load contributed by northward transport of a suspension plume from the Cho-Shui River, while the southern regime, low in suspended load and high in chlorophyll concentration, is a system controlled by biological activity. A scavenging model that takes account of the physical transport was used to estimate the ^{234}Th budget in order to estimate the scavenging and removal rates from the nearshore water. The scavenging and removal rates ranged from 21 to 127 dpm $\text{m}^{-3}\text{d}^{-1}$ and from 36 to 525 dpm $\text{m}^{-3}\text{d}^{-1}$, for dissolved and particulate ^{234}Th , respectively. The removal fluxes of particulate organic carbon (POC) and particulate organic nitrogen (PON) were estimated by multiplying the particulate ^{234}Th removal flux to the organic carbon/ ^{234}Th and nitrogen/ ^{234}Th ratios in suspended particles, which ranged from 4.5 to 275.2 mmol-C $\text{m}^{-2}\text{d}^{-1}$ and from 1.3 to 50.1 mmol-N $\text{m}^{-2}\text{d}^{-1}$, respectively. These fluxes resulted in residence times of 1–20 days for the POC in the surface water of nearshore water off western Taiwan.

Keywords:

- ^{234}Th ,
- particle scavenging,
- nearshore environment.

1. Introduction

The growing awareness of the need to build a sustainable habitat has made it more urgent for environmental authorities to manage the pollution problems and promote various monitoring campaigns in the coastal ocean of Taiwan. A major problem of the huge data sets collected from these monitoring programs is that they often lack of quantitative rate information to quantify the pollutant transport, which is necessary when determining element budget of interest. Since many pollutants are particle-reactive, e.g., Pb, Hg, Cu, PCBs, and DDTs, it is important to comprehend marine particle and POC dynamics and flux in the coastal water in order to understand the fate of these elements or compounds when they are discharged from land. In addition to the context of environmental protection, coastal waters form a complex system with allochthonous characteristics and a wide range of particle concentrations, which provides strongly contrasting biogeochemical settings for investigating the

mechanism of scavenging and removal processes by which particle-reactive elements are scavenged and removed.

The coastal waters of western Taiwan are subject to the influence of four major river systems, characterized by their high suspended loads from high denudation rates of western Taiwan (Dadson *et al.*, 2003). From the north to the south of western Taiwan, the Dan-Shui River (DSR) receives large amounts of sewage effluent discharged from metropolitan Taipei and is severely threatened by environmental degradation (Wen *et al.*, 2008); the Cho-Shui River (CSR) is laden with the highest particulate matter concentration among all rivers of Taiwan; the Jen-Wen River (JWR) is known for its barrier islands and lagoon settings (Liu *et al.*, 2000); and the southernmost Kao-Pin River (KPR) collects the municipal sewage effluents discharged from Kaohsiung, the second largest city of Taiwan, and serves as the major source of fluvial sediments in the shelf-slope region off southwestern Taiwan (Kao *et al.*, 2006).

The naturally-occurring isotope ^{234}Th ($t_{1/2} = 24.1$ days) is one of the most particle-reactive elements with a particle-water distribution coefficient (i.e. K_d , the ratio

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of particulate to dissolved concentration) ranging from 10^6 to 10^7 ml g⁻¹ in estuarine and coastal waters (Honeyman *et al.*, 1988). ^{234}Th is continuously produced from its parent, ^{238}U , and has been widely used as a tracer for particles in coastal marine environments (Santschi *et al.*, 1979; Minagawa and Tsunogai, 1980; Tanaka and Tsunogai, 1983; Wei and Murray, 1992; Baskaran and Santschi, 1993; Niven *et al.*, 1995; Radakovitch *et al.*, 2003). ^{234}Th - ^{238}U disequilibria can provide quantitative estimate of scavenging and removal rates by simple mass balance calculation if both dissolved and particulate ^{234}Th concentrations are measured, and the removal rates of associated elements in the particles can then be estimated by multiplying the ^{234}Th flux by the ratio of the specific element or compound (e.g., particulate organic carbon: Murray *et al.*, 1989; PAH: Gustafsson *et al.*, 1997; trace metals: Weinstein and Moran, 2005) to ^{234}Th . The rate information derived from ^{234}Th - ^{238}U disequilibria can therefore be used to quantify the removal rates of reactive heavy metals and pollutants discharged through rivers.

The wide range of particle concentration and variable particle composition in coastal waters provides the opportunity to investigate the influences of this important master variable on metal recycling processes. The purpose of this study is to use short-lived ^{234}Th to estimate the particle scavenging and removal rates in the coastal marine environment of Taiwan. In addition, using ^{234}Th as a proxy for POC and PON, we have attempted to estimate the removal fluxes of particulate organic carbon and particulate nitrogen from the nearshore water.

2. Methods

2.1 Sampling

At the stations shown in Fig. 1, seawater samples were taken during 19–23 November 2004 from the R/V Ocean Researcher II (cruise #1248). Seawater was collected at a depth of 2 meters using 10 L Teflon-coated Go-Flo water samplers (General Oceanic Inc., USA) mounted on a Sea-Bird CTD (SBE 9/11, Sea-Bird Electronics Inc., USA) rosette system.

2.2 Instruments and chemical analyses

Vertical distributions of fluorescence and light transmission in the water column were measured by attaching a fluorometer (Chelsea Instrument Aquaparcka MK-III) and a deep water transmissometer (Chelsea Instrument Alphaparcka MK-II) with 25 cm light path length to the CTD system, respectively. The beam attenuation coefficient (BAC) was calculated from the percentage transmission (T%) as $-(\ln(\text{Tr}/100))/0.25$. Along the cruise track, continuous measurements of fluorescence and light transmission were also measured by the underway sys-

Fig. 1. Location of sampling station of ORII-1248 during 19–23 November 2004. The five rivers that contribute the highest particulate matter into the west coast of Taiwan are shown as: DSR (Dan-Shui River), HLR (Ho-Lung River), CSR (Cho-Shui River), JWR (Jen-Wen River), and KPR (Kao-Ping River). Three stations shown with squares are reference stations used for the calculation in the model (see text).

tem (Wetlabs C-Star) installed onboard. The intake of the underway system is about 2 meters below the surface.

Silicate was determined on board ship by the modified standard molybdenum blue method with a flow injection analyzer (Grasshoff, 1999).

Subsamples for the determination of chlorophyll-a and phaeopigments were filtered on board ship through 47-mm Whatman GF/F glass fiber filters. The filters were stored at -20°C and returned to the shore base laboratory for further processing. In the laboratory the filters were ground and extracted with 90% acetone. The chlorophyll-a and phaeopigments in the extract are then measured fluorimetrically using a Turner model 10-AU-005 fluorometer (Strickland and Parsons, 1972; Gong *et al.*, 1993).

POC and PON were determined by filtering 2 L seawater through a precombusted (450°C) Whatman 25 mm GF/F filter. The filter was wrapped in aluminum foil

Table 1. Stations, longitude, latitude, water depth, salinity (S), temperature (T), total suspended matter concentration (TSM), dissolved (DTh), particulate (PTh) ^{234}Th , particulate organic carbon (POC) and particulate nitrogen (PON) along the coast line of western Taiwan. All hydrographic and ^{234}Th data are collected at 2 m depth.

Station	Longitude	Latitude	Water depth	S	T	TSM	DTh	PTh	POC	PON
	E	N	m		°C	mg L ⁻¹	dpm L ⁻¹	dpm L ⁻¹	μM	
B3	121.47	25.28	29	33.631	21.696	6.24	0.021 ± 0.001	0.991 ± 0.049	10.8	2.2
D4	121.38	25.19	25	33.429	22.236	10.08	0.225 ± 0.014	1.092 ± 0.052	10.6	1.9
B5	121.23	25.13	13	32.956	22.114	7.96	0.213 ± 0.014	0.450 ± 0.024	8.3	1.7
B6	120.99	25.00	24	33.417	22.679	11.58	0.086 ± 0.005	0.472 ± 0.024	15.4	2.9
B7	120.90	24.87	30	33.905	23.722	4.02	0.337 ± 0.021	0.118 ± 0.007	10.2	1.9
B8	120.79	24.70	24	34.084	23.780	6.44	0.457 ± 0.021	0.276 ± 0.015	8.4	1.6
B9	120.73	24.63	11	33.896	22.762	18.10	0.169 ± 0.010	0.494 ± 0.028	16.5	3.1
B10	120.64	24.51	22	33.932	22.897	8.97	0.085 ± 0.005	0.230 ± 0.014	9.6	1.8
B11	120.52	24.34	15	34.099	23.113	6.44	0.449 ± 0.024	0.185 ± 0.011	8.1	1.5
B12	120.43	24.20	10	33.570	23.527	13.61	0.367 ± 0.023	0.323 ± 0.017	13.3	2.6
B13	120.32	24.07	9	33.799	21.944	17.34	0.117 ± 0.007	0.424 ± 0.026	12.3	2.0
D14	120.20	23.88	13	34.339	22.795	18.09	0.178 ± 0.010	0.212 ± 0.012	n.d.	n.d.
B15	120.14	23.71	8	34.321	22.172	31.46	0.185 ± 0.010	0.353 ± 0.021	20.5	3.4
B17	120.08	23.34	12	34.198	22.689	13.95	0.151 ± 0.010	0.311 ± 0.015	16.8	3.0
B18	120.04	23.26	17	34.457	24.024	7.45	0.095 ± 0.006	0.374 ± 0.019	9.8	1.8
D19	120.05	23.03	10	34.517	24.873	3.86	0.041 ± 0.002	0.358 ± 0.019	9.3	1.7
B20	120.14	22.96	13	34.339	24.711	3.91	0.004 ± 0.001	0.207 ± 0.013	11.0	2.0
B21	120.17	22.86	11	34.457	25.380	1.98	0.032 ± 0.002	0.292 ± 0.017	8.6	1.6
B22	120.23	22.69	9	34.358	25.039	3.63	0.069 ± 0.004	0.226 ± 0.013	14.8	2.5
B23	120.28	22.54	17	34.356	25.515	1.70	0.092 ± 0.005	0.250 ± 0.013	5.5	1.0
D24	120.41	22.47	43	34.300	25.409	1.78	0.184 ± 0.008	0.421 ± 0.023	5.5	0.9
B25	120.50	22.40	14	34.276	25.315	1.87	0.027 ± 0.001	0.241 ± 0.014	4.5	0.8
B26	120.63	22.27	21	34.455	25.549	1.21	0.744 ± 0.080	0.183 ± 0.012	2.1	0.5
B27	120.68	22.07	19	34.496	25.154	1.05	0.499 ± 0.022	0.402 ± 0.024	1.1	0.3
HT	120.68	21.96	284	34.491	25.502	1.36	0.521 ± 0.023	0.257 ± 0.015	0.9	0.4

*n.d.: not determined.

and stored at -4°C . In the laboratory the filter was acid-fumed to remove carbonates, then measured with a Fisons elemental analyzer (NA1500). A calibration curve was obtained by running acetanilide ($\text{C}_8\text{H}_9\text{NO}$) standard. The overall procedural errors estimated from duplicates were better than $\pm 2\%$ for both carbon and nitrogen determinations.

Dissolved and particulate ^{234}Th were determined from 20 L seawater collected at each sampling station. Immediately after the seawater was transferred to the PVC pressure drums, compressed air (at 12 p.s.i.) was used to force seawater through preweighed 142 mm Nuclepore filters ($0.45\text{ }\mu\text{m}$ pore size) mounted in a Plexiglass filter holder. The filters were rinsed with approximately 15 ml deionized distilled water to remove sea salt and stored in a petri dish to determine the total suspended matter (TSM) concentration and particulate ^{234}Th activities.

The filtrate was acidified with about 20 ml concen-

trated HCl and spiked with 30 dpm of ^{230}Th and 50 mg of Fe carrier. The samples were bubbled vigorously for at least 3 hours to reach isotopic equilibrium. Concentrated NaOH was then added to raise the pH to 8 and precipitate $\text{Fe}(\text{OH})_3$. The $\text{Fe}(\text{OH})_3$ precipitates, with adsorbed thorium, were collected by siphoning and centrifuging, and then dissolved in concentrated HCl to make the samples 9 N in HCl. These samples were then passed through an anion exchange column (Dowex1X-8) preconditioned with 9N HCl to separate uranium from thorium. Thorium samples were purified by passing the sample through three anion exchange columns pre-conditioned with 8 N HNO_3 . The sample was evaporated down to one drop and was ready for extraction. ^{234}Th and the yield tracer, ^{230}Th , were extracted into a 0.4 M TTA (thenoyltrifluoroacetone)-benzene solution and stippled on a stainless-steel disc. Preconcentration and separation of uranium and thorium from the filtered seawater samples were completed in

three days after samples were collected.

After weighing for the determination of TSM concentration, the filters were decomposed in the laboratory by soaking in ~10 ml of concentrated NH_4OH . The samples were gently heated to evaporate the NH_4OH then fluxed in HClO_4 -HF to thoroughly digest organic and inorganic materials. After digestion, the samples were purified and mounted on stainless-steel discs following the same procedures as for dissolved ^{234}Th samples.

The activities of ^{234}Th were counted by a low background (<1.1 cpm) anticoincidence counter (Tennelec LB-5100) via its β -emitting daughter ^{234}Pa . The chemical yield of thorium was estimated by counting spiked ^{230}Th using silicon surface-barrier detectors (EG&G Ortec 576). The counting efficiencies of the α detectors were calibrated against NIST traceable ^{230}Th (Isotope Products Laboratory 387-67-3) standard plates. Activities of ^{234}Th reported here were corrected back to the sampling time after the ingrowth of ^{234}Th from ^{238}U was subtracted.

3. Results

Stations, longitude, latitude, water depth, salinity, temperature, TSM, dissolved ^{234}Th (DTh), particulate ^{234}Th (PTh), POC and PON are listed in Table 1. Latitudinal distributions of selected parameters are shown and briefly described.

3.1 Temperature, salinity, fluorescence, BAC

Underway measurements of temperature and salinity along the cruise track of water sampling stations are shown in Fig. 2a. Independent measurements of temperature and salinity at 2 m depth by CTD/Rosette deployments at the 27 stations are also plotted in the figure. As a result of interaction between the cold China Coastal Current and warm Kuroshio Branch Current residing in the northern South China Sea during the prevailing northeast monsoon, a temperature front was observed at latitude $23^\circ40'$ N (Jan *et al.*, 2002). The coastal water to the south of the front was about 3°C warmer than that to the north of the front. The salinity of coastal water to the south of the front (station B15) averaged 34.2 and then showed a somewhat decreasing trend northward. Due to the riverine influence, evident spikes of low temperature and low salinity were clearly observed.

Underway measurements of fluorescence (F) and BAC are shown in Fig. 2b. Independent sets of F and BAC data are also available from the CTD/Rosette deployments at the 27 stations and are plotted in the figure. Although some fine structures clearly shown by the underway system were missed, it can be seen that discrete observations of the abovementioned parameters are consistent with continuous data, and general distribution patterns are retained. Fluorescence readings along the cruise track showed an interesting spatial distribution. Two distinct

Fig. 2. Latitudinal distributions of (a) salinity (S, black line) and temperature (T, grey line) and (b) beam attenuation (BAC, black line) and fluorescence (F, grey line) along the coastline of western Taiwan. Discrete symbols represent the four parameters measured independently with the sensors mounted on the CTD/Rosette system at water sampling stations. Locations of the five major river mouths are shown as abbreviations on top of the figure: DS (Dan-Shui River), HL (Ho-Lung River), CS (Cho-Shui River), JW (Jen-Wen River), and KP (Kao-Ping River).

regimes were identified, separated by the temperature front at $23^\circ40'$ N. In the northern regime, F was low and uniform, whereas the F reading in the southern regime was high and variable. In the southern regime, peaks of F were usually found in concert with temperature and salinity extremes, indicating the riverine influence upon the biological activity.

Similar to the F distribution, two distinct regimes separated by the temperature front were observed for the latitudinal distribution of BAC. However, in contrast to the F distribution, it can be seen that the BAC was much higher and more variable in the northern regime than that in the southern one. As shown in the figure, the locations of the four major rivers and Ho-Lung River, with its high suspended load, were associated with a high BAC signal. As expected, the highest BAC signal was associated with CSR, which is the most turbid river in Taiwan. In fact,

Fig. 3. Latitudinal distributions of (a) total suspended matter concentration (TSM, circles) and total pigments (TP, squares), (b) particulate organic carbon (POC, circles) and silicate concentration (SiO_2 , squares), and (c) dissolved (DTh, circles), particulate (PTh, solid squares), and total (TTh, crossed squares) ^{234}Th at water sampling stations along the coastline of western Taiwan.

the name Cho-Shui River means “turbid” river in Chinese.

3.2 TSM, pigments, POC, and SiO_2

Latitudinal distributions of TSM and total pigments (TP = chlorophyll-a + phaeopigments) are shown in Fig. 3a. It can be seen that TSM distribution varies in tandem with BAC (m^{-1}) distribution (cf. Fig. 2b) and the close correlation between TSM and BAC gives a linear correlation equation ($r^2 = 0.97$, $n = 47$):

$$\text{TSM} = -1.08 + 2.35\text{BAC}. \quad (1)$$

TP values also correlate closely with F readings with a linear equation ($r^2 = 0.77$, $n = 47$):

$$\text{TP} = -0.19 + 0.38\text{F}. \quad (2)$$

It is interesting to note that TSM peaks coincide with pigment peaks in the northern regime whereas there is no evident coincidence between the two parameters in the southern regime, which reflects different particle composition between the two regimes. Higher TSM in the northern regime resulted from the northward transport of the CSR river particulate plume and resuspension of bottom sediments.

Latitudinal distributions of POC and SiO_2 are shown in Fig. 3b. The two parameters are generally correlated with each other and show elevated concentrations at the five river mouths, indicating the riverine influence on the biogeochemical settings in the nearshore water. The influence of riverine input is supported by the close correlation of POC and TSM in the region to the north of CSR. It is also noteworthy that elevations of POC and pigment concentrations and depletion of SiO_2 were found in the region between KPR and JWR, indicating enhanced biological production.

3.3 Dissolved and particulate ^{234}Th

Latitudinal distributions of dissolved (DTh) and particulate (PTh) ^{234}Th activities are shown in Fig. 3c. Total ^{234}Th (TTh = DTh + PTh) are also plotted in the figure. Large variability of DTh, ranging from <0.01 to 0.74 dpm L^{-1} , was observed. The PTh does not show latitudinal variability as marked as that of DTh, except at the northernmost stations where much higher PTh was found. Four distinct regimes can be identified from the spatial distributions.

(1) South of KPR: Despite significantly lower activity than the secular equilibrium value ($\sim 2.4 \text{ dpm L}^{-1}$), DTh in this region is the highest among all sampling stations. In this region, most (50–80%) of TTh is in the dissolved phase. As pointed out by Wei *et al.* (2008a), this region is affected by southeastward transport of South China Sea water.

(2) KPR–CSR: In this region, DTh is extremely low ($<0.2 \text{ dpm L}^{-1}$) while PTh ranges between 0.2 and 0.4 dpm L^{-1} . Centered at JWR, the DTh distribution reveals an interesting V-shaped pattern.

(3) CSR–DSR: In between CSR and DSR, both DTh and PTh show a clear M-shaped pattern with a spatial offset. The peaks of PTh and the troughs of DTh are generally located at the river mouths of CSR, HLR, and DSR.

(4) North of DSR: In contrast to the region to the south of KPR, PTh in this region elevates to the highest activity among the sampling stations and is the dominant phase ($>80\%$) of ^{234}Th in seawater.

TTh also show great variability, ranging from an extremely low value of 0.21 dpm L^{-1} at JWR to 1.32 dpm L^{-1} in the northernmost region.

4. Discussion

As shown Fig. 3c, except in the northernmost and southernmost regions, low TTh and hence low $^{234}\text{Th}/^{238}\text{U}$ ratio ($\text{TTh}/\text{U} \approx 0.2$) were commonly found along the coastline, which indicates that an active particle removal process is imposed on the nearshore environment off west coast of Taiwan. Compared with other shallow waters, this low TTh/U ratio is comparable with Narragansett Bay (Santschi *et al.*, 1979), Yangtze Estuary (McKee *et al.*, 1984) and Mecklenburg Bay (Kersten *et al.*, 1998), but higher than Long Island Sound (Aller and Cochran, 1976), Texan Bay (Baskaran and Santschi, 1993) and Casco Bay (Gustafsson *et al.*, 1998). We first quantify the scavenging and particle removal rates by adopting an advection-scavenging-removal model then combine the comprehensive ancillary data to depict the effect of allochthonous characteristic of the nearshore setting on the cycling of ^{234}Th and particulate organic carbon along the nearshore water off western Taiwan.

4.1 ^{234}Th scavenging and removal rates

Since the early use of the simple irreversible scavenging model (Coale and Bruland, 1985; Wei and Murray, 1992), the thorium community has asked for a more realistic model for ^{234}Th budgeting in the surface ocean (Benitez-Nelson *et al.*, 2000). Given the dynamic nature of the nearshore environment, it is necessary to include the physical transport factors when $^{234}\text{Th}/^{238}\text{U}$ disequilibrium is applied to the study of coastal scavenging processes (Savoye *et al.*, 2006 and references therein). The scavenging model adopted here is established based on the rationale given by Gustafsson *et al.* (1998), where horizontal transport is included in the mass balance of ^{234}Th . Assuming a steady-state, the mass balance of DTh and PTh are described by the following equations:

$$\begin{aligned} \frac{\partial \text{DTh}_i}{\partial t} &= 0 \\ &= \frac{V_i(\text{DTh}_{i-1} - \text{DTh}_{i+1})}{\Delta L} + \lambda_{\text{Th}} U_i - \lambda_{\text{Th}} \text{DTh}_i - J_i, \end{aligned} \quad (3)$$

$$\frac{\partial \text{PTh}_i}{\partial t} = 0 = J_i + \frac{V_i(\text{PTh}_{i-1} - \text{PTh}_{i+1})}{\Delta L} - \lambda_{\text{Th}} \text{PTh}_i - R_i, \quad (4)$$

where

U_i = average ^{238}U activity at station i , which is calculated from salinity according to Ku *et al.* (1977) (dpm m^{-3});
 DTh_i = dissolved ^{234}Th activity at station i (dpm m^{-3});
 PTh_i = particulate ^{234}Th activity at station i (dpm m^{-3});
 V_i = average current velocity at station i (m d^{-1});
 ΔL = distance between stations $i-1$ and $i+1$ (m);

λ_{Th} = radioactive decay constant of ^{234}Th (d^{-1});

J_i = net scavenging rate of dissolved ^{234}Th at station i ($\text{dpm m}^{-3}\text{d}^{-1}$);

R_i = net removal rate of particulate ^{234}Th at station i ($\text{dpm m}^{-3}\text{d}^{-1}$).

As the thorough review by Savoye *et al.* (2006) points out, the difficulty of including horizontal advection in budgeting coastal ^{234}Th is mainly caused by acquiring a reliable value for current velocity in this high energy environment. In many studies in the open ocean, the parameters needed for the estimation of physical transport were derived indirectly from the physical model (e.g., Dunne *et al.*, 1997). Different approaches have been adopted in coastal environments. For example, Gustafsson *et al.* (1998) estimated the horizontal velocities using salinity gradients in the Casco Bay, Gulf of Maine, and Benitez-Nelson *et al.* (2000) estimated the horizontal advection velocities from historical current meter measurements in the Gulf of Maine. Recently, Giuliani *et al.* (2007) attempted to use average current measured with a current meter to evaluate the importance of horizontal transport for ^{234}Th removal in the coastal region of the northern Mediterranean Sea, but ended up by neglecting the physical term in calculating the ^{234}Th removal rate. In this study we estimated the mean current velocity from historical shipboard ADCP (sb-ADCP) observations.

Following the procedures of Liang *et al.* (2003), mean longshore current velocity at each station defined by a 10×10 km area during winter (October–December) was estimated by averaging archived shipboard October–December sb-ADCP data (NCOR Data Bank) collected during 1991–2005. At most of the locations, at least 15 data sets were available, which is ample for a reliable estimate of average flow field to filter out the semidiurnal tidal current (Liang *et al.*, 2003). However, at the three stations off the central coast (B15, B17, B18) the number of data sets accumulated in the past two decades is too small (<10) to give reliable averaging current velocity. It is therefore assumed that the current velocity at B15 is the same as at D14 and the current velocity at B17 and B18 is the same as at B19. The resultant current velocities at each station are shown in Table 2.

Since the direction of the average current at the stations to the north of station B9 are generally towards NNE (Table 2), so the dissolved and particulate ^{234}Th activities of 0.13 and 0.43 dpm L^{-1} , respectively, obtained at $121^\circ 30' \text{ E } 25^\circ 30' \text{ N}$ (shown as N30 in Fig. 1, Wei *et al.*, 2008b) were assumed to be the destined DTh and PTh of outflow water from these stations. A Lagrangian observation by drifters deployed in October–November 1997 in the Central Taiwan Strait showed a strong current with a mean speed of $30\sim 50 \text{ cm s}^{-1}$ flowing northward in the Peng-Hu Channel (Chern and Wang, 2000) and, as a result of topographic influence, both the sb-ADCP data

Table 2. E-W (U) and N-S (V) components of current, physical transport of dissolved (V_{DTh}) and particulate (V_{PTh}) ^{234}Th , scavenging (J) and removal (R) rates, residence times of dissolved (τ_{DTh}) and particulate (τ_{PTh}) calculated from the scavenging model. Parallel changing rates and residence times calculated from the model without physical transport are given in italics.

Station	U	V	V_{DTh}	V_{PTh}	J	R	τ_{DTh}	τ_{PTh}	J	R	τ_{DTh}	τ_{PTh}
	cm s ⁻¹		dpm m ⁻³ d ⁻¹				day		dpm m ⁻³ d ⁻¹		day	
B3	25.7	4.1	55	431	122	525	0.2	1.9	68	39	0.3	25.2
D4	7.8	4.3	12	15	73	57	3.1	19.1	62	30	3.7	36.2
B5	-1.2	2.9	-1	4	60	51	3.6	8.8	61	48	3.5	9.4
B6	6.2	27.5	52	-56	118	48	0.7	9.8	66	52	1.3	9.1
B7	2.4	26.6	63	-12	122	107	2.8	1.1	59	56	5.7	2.1
B8	5.7	7.0	2	9	59	60	7.8	4.6	56	48	8.1	5.7
B9	4.7	11.5	-3	-8	61	39	2.8	12.8	64	50	2.6	9.9
B10	2.7	1.5	19	-21	86	58	1.0	4.0	67	60	1.3	3.8
B11	6.9	9.6	70	23	127	145	3.5	1.3	57	51	7.9	3.6
B12	7.8	8.8	-34	38	24	52	15.4	6.2	58	49	6.3	6.7
B13	11.9	13.8	-44	36	21	45	5.5	9.5	65	53	1.8	8.0
D14	3.2	-4.4	8	2	73	69	2.4	3.1	65	59	2.7	3.6
B15	3.2	-4.4	3	20	67	77	2.8	4.6	65	54	2.9	6.5
B17	2.5	2.5	-5	1	60	52	2.5	6.0	65	56	2.3	5.5
B18	2.5	2.5	-10	4	58	51	1.6	7.3	67	57	1.4	6.6
D19	2.5	2.5	-8	-15	61	36	0.7	10.0	69	59	0.6	6.1
B20	1.8	5.8	-2	-15	68	46	0.1	4.5	70	64	0.1	3.2
B21	-5.0	9.4	19	6	88	85	0.4	3.4	69	61	0.5	4.8
B22	-4.6	3.0	8	-5	76	64	0.9	3.5	68	61	1.0	3.7
B23	-2.1	2.0	9	15	76	84	1.2	3.0	67	60	1.4	4.2
D24	1.4	0.2	-3	-0	62	49	3.0	8.5	65	52	2.9	8.0
B25	0.9	1.9	31	-13	100	80	0.3	3.0	69	62	0.4	3.9
B26	0.4	1.5	15	5	64	63	11.7	2.9	49	44	15.3	4.2
B27	5.0	-11.0	67	-22	122	89	4.1	4.5	56	44	8.9	9.1
HT	4.7	-14.0	-17	34	38	65	13.7	4.0	55	48	9.4	5.4

(Liang *et al.*, 2003) and the drifter experiment (Tseng and Shen, 2003) revealed the strong landward flow at 24°10' N. Accordingly, the DTh of 0.21 dpm L⁻¹ and PTh of 0.45 dpm L⁻¹, respectively, obtained at 120°00' E 24°00' N (shown as C17 in Fig. 1, Wei *et al.*, 2008b) were assumed to be the DTh and PTh of incoming water into the stations (B12, B13, D14, and B15) located in the central region. The flow field derived from the sb-ADCP data in the vicinity of JWR agreed with the direct current measured by current meter deployments at the JWR river mouth (our station D19) by Liu *et al.* (2000). The average current at the southernmost station is to the southeast (Table 2, Wei *et al.*, 2008a). Consequently, the DTh and PTh, 0.55 and 0.30 dpm L⁻¹, respectively, measured at S2 in the northern Luzon Strait were used (Fig. 1, Wei and Hung, 1992) as the destined values of the outflow water of the southernmost station (HT).

The results of the model calculations are listed in Table 2. For comparison, the results calculated from the scavenging model without physical transport ($V = 0$ in Eqs. (3) and (4)) are also shown. The net ^{234}Th change due to advection ranges from -44 to 70 dpm m⁻³d⁻¹ and from -56 to 38 dpm m⁻³d⁻¹, for dissolved (first term of right-hand side of Eq. (3), V_{DTh}) and particulate (second term of right-hand side of Eq. (4), V_{PTh}) ^{234}Th (Table 2), respectively. Although the absolute values of the changing rates of DTh and PTh caused by physical transports are generally lower than the scavenging (J) and removal (R) rates, respectively, the contribution by physical transport is still too significant to be neglected for ^{234}Th budgeting in the nearshore water.

Latitudinal variations of J and R are shown in Fig. 4a. Excluding the very high removal rate at the northernmost station B3, J and R calculated from the scavenging

Fig. 4. Latitudinal distributions of (a) scavenging (J , circles) and removal (R , squares) rates of ^{234}Th and (b) residence times of dissolved (τ_{DTh} , circles) and particulate (τ_{PTh} , squares) ^{234}Th calculated by the scavenging model at water sampling stations along the coastline of western Taiwan.

model range from 21 to 127 $\text{dpm m}^{-3}\text{d}^{-1}$ and 36 to 145 $\text{dpm m}^{-3}\text{d}^{-1}$, respectively. The lower R at the river mouths may be a result of higher energy associated with river influx that favors dispersion of fine particles. The exceptionally high removal rate of 525 $\text{dpm m}^{-3}\text{d}^{-1}$ at station B3 is a result of high advective input from the south and very small output of particulate ^{234}Th due to small PTh gradient between station B3 and the destined location ($\text{N}30$) in the northern Taiwan Strait. It can be seen that latitudinal variation of J and R in the nearshore water are very similar to each other. A close correlation between J and R was also found in other coastal environments (Cochran *et al.*, 1995; Baskaran *et al.*, 1996; Trimble and Baskaran, 2005). Interestingly, both J and R appear to be lower at the five major river mouths. It is possible that lower J is caused by the retardation of ^{234}Th scavenging due to riverine input of dissolved organic matter and lower R is caused by the enhancement of sediment resuspension at the river mouths. Unlike the findings from the open ocean (Bacon and Anderson, 1982) and coastal environment (Wei and Hung, 1992), there is no relationship between the J and TSM concentration (cf. Fig. 3b), indicating that the particle composition may also play a significant role in ^{234}Th scavenging in the nearshore water.

Sediments serve as the final repository of riverine

and/or marine particles, so their accumulation of sediments can be envisioned as an ultimate sediment trap to provide a constraint on ^{234}Th removal. Therefore, in order to check the validity of our estimates, we compare the ^{234}Th removal flux, which is calculated by multiplying R by water depth, against the expected accumulation rate from excess ^{234}Th inventory in the sediment. The flux of ^{234}Th into the sediments can be calculated from the inventory of unsupported ^{234}Th in the sediment column. Unfortunately only one core is available from a site near station B11. From the vertical profile of excess ^{234}Th in this core top (not shown), a burial flux of ~ 3000 $\text{dpm m}^{-2}\text{d}^{-1}$ is needed to give a steady-state ^{234}Th inventory in the sediment. This burial flux is reasonably consistent with the vertical ^{234}Th flux of 2175 $\text{dpm m}^{-2}\text{d}^{-1}$ estimated by multiplying the removal rate by the water depth at station B11. If no physical term were included in Eqs. (1) and (2), the ^{234}Th removal flux at this site would be 765 $\text{dpm m}^{-2}\text{d}^{-1}$ ($51 \text{ dpm m}^{-3}\text{d}^{-1} \times 15 \text{ m}$), which is only one third of the burial flux. Hence, it is evident that the physical transport is too important to be ignored in the region of high current speed.

4.2 Residence times of ^{234}Th

The residence times of dissolved (τ_{DTh}) and particulate (τ_{PTh}) ^{234}Th relative to scavenging and removal rates, respectively, can be calculated by the following equations:

$$\tau_{\text{DTh}} = \frac{\text{DTh}_i}{J_i}, \quad (5)$$

$$\tau_{\text{PTh}} = \frac{\text{PTh}_i}{R_i}. \quad (6)$$

Shown in Fig. 4b, the τ_{DTh} and τ_{PTh} range from 0.1 to 15.3 days and during 3.6 to 36.2 days, respectively. Compared with bacterial production data measured from the same cruise, the τ_{DTh} generally decreases with increasing bacterial activity in the nearshore water. Microbial remineralization of particulate matter may increase the concentration of complexing ligand, favoring the formation of transparent exopolymer particles (TEP), which has been shown to be an efficient ^{234}Th scavenger (Passow *et al.*, 2006). In the coastal water off California, Lee *et al.* (1993) also observed enhanced ^{234}Th scavenging when bacterial biomass in particles increases. The τ_{DTh} (<3 hr) at the JWR river mouth is the shortest ever found in a marine environment, indicating extremely active scavenging by the particles residing in this locality. This exceptionally rapid scavenging phenomenon in the JWR system, which is known for its lagoon setting, deserves further investigation. As a result of lower R , the τ_{PTh} at the

Fig. 5. Compilation of the residence time of total ^{234}Th (τ_{TTh}) in the turbid nearshore environments reported in the literature (Mecklenburg: Kersten *et al.*, 1998; Narangansett: Santschi *et al.*, 1979; Hooghly: Somayajulu *et al.*, 2002; Yangtze: McKee *et al.*, 1984; Amazon: McKee *et al.*, 1986; Texas: Baskaran and Santschi, 1993; Hudson: Feng *et al.*, 1999). Horizontal and vertical arrows represent the ranges of TSM concentration and τ_{TTh} , respectively, of reported values.

mouths of the five rivers seems to be longer than in other locations, which is caused by higher PTh due to active resuspension in the vicinity of the river mouths. The residence time of total ^{234}Th (τ_{TTh}) relative to the removal rate is approximated by but not equal to the sum of τ_{DTh} and τ_{PTh} (Coale and Bruland, 1985). τ_{TTh} values shorter than a few days can only be found in nearshore environments with extremely high suspended load. Although limited ^{234}Th data in nearshore environments are available, the compilation of literature data collected in shallow marine waters with high turbidity showed an inverse correlation between τ_{TTh} and TSM concentration (Fig. 5). Since it spans a wider geographic range, it is reasonable that the τ_{TTh} of the nearshore water found in this study showed a wider range, from <1 to 40 days, than the values in other nearshore environments.

4.3 Northern vs. southern regimes

Two distinctive regimes of contrasting environmental settings in the west coast of Taiwan can be identified, separated by the temperature front at $23^{\circ}30'\text{N}$ (Fig. 2a), where the node of tidal oscillation is approximately located (Wang *et al.*, 2003). Clear contrasts between the northern and southern regimes are evidenced by the latitudinal distributions of BAC, F, TSM and total pigments (Figs. 2b and 3a). The particulate concentration in the northern regime is higher than in the southern one, whereas the pigment distribution shows a reverse trend.

Fig. 6. Latitudinal distributions of (a) organic carbon content (POC/TSM, circles) and pigment content (TP/TSM, squares) in the suspended particles and (b) distribution coefficient (K_d , circles) and specific particulate ^{234}Th activity (SPTh, squares) in suspended particles at water sampling stations along the coastline of western Taiwan.

This contrasting distribution of the biochemical parameters is mainly caused by the northward transport of terrestrial particle plume originating from the most turbid CSR. As a result of high water discharge ($\sim 1.58 \times 10^{10} \text{ m}^3 \text{ y}^{-1}$) and heavy suspension loads ($\sim 4.04 \times 10^3 \text{ g m}^{-3}$), CSR provides the largest amount of sediment into the Taiwan Strait. The growth of phytoplankton biomass in the northern coastal water was impeded due to the light-limiting effect of elevated turbidity.

In addition to the contrasting distributions of the abovementioned parameters, the contrasting characteristic between the northern and the southern regimes is also reflected by the difference in the particle composition. Latitudinal distributions of organic carbon content in the suspended particles (POC/TSM in unit of %) and specific concentration of total pigments (TP/TSM in unit of $\mu\text{g mg}^{-1}$) are shown in Fig. 6a. It can be seen that specific concentrations of total pigments were low, averaging at $0.1 \mu\text{g mg}^{-1}$, in the suspended particulates collected from the northern regime then increased sharply to $0.8 \mu\text{g mg}^{-1}$ to the south of JWR. Our measurements of chlorophyll-a and phaeopigment show that about 50% of total pigment is contributed by chlorophyll-a in the southern regimes,

whereas only 20% is contributed by fresh chlorophyll-a in the northern regime. A large variability of POC/TSM is found, ranging from 0.7 to 5.2%. As a result of dilution by riverine input, the POC/TSM is lowered at the river mouths of the five rivers. A sharp POC/TSM maximum of 5.2% exists in the region between JWR and KPR, which is much higher than the organic carbon content of 0.42~0.58 % in the suspended particles collected from the river water of JWR and KPR (Kao *et al.*, 2006). It should be noted that close correlation of DTh and silicate (cf. Figs. 3b and c) ($r^2 = 0.76$, $n = 8$) is found in this region, indicating that diatoms may play an important role in ^{234}Th scavenging. Diatoms are known to produce more polysaccharides, which may coagulate and form TEP in seawater, serving as an efficient scavenger for ^{234}Th (Guo *et al.*, 2002; Santschi *et al.*, 2003; Passow *et al.*, 2006).

The contrasting composition of suspended particles in the northern and southern regimes implies a different degree of influence by rivers. In the northern regime, overwhelmed by northward transport of the large particle input from the CSR and the dilution effect of terrestrial detritus, the particles were low in pigment concentration and organic content. Manganese data obtained during the same cruise showed 5 to 10-fold higher specific Mn concentrations in the suspended particles collected from the northern regime than from the southern one, indicating a substantial influence of terrestrial input from the rivers in the northern regime. On the other hand, as shown by the elevated pigment concentration (Fig. 3a), specific pigment concentration (Fig. 6a) and higher primary production, the particulate matter collected in the southern regime is mainly produced in situ. Based on the stable carbon isotope data in the surface sediments, Kao *et al.* (2006) also concluded that a high percentage of organic carbon (38~48%) in the bottom sediment at the nearshore sites off southwestern Taiwan is derived from marine sources. It can therefore be concluded that the contrasting environmental conditions lead to different removal mechanisms between the southern and northern regimes, i.e., ^{234}Th removal is controlled by the large quantity of terrestrial particles in the northern regimes, whereas enhanced biological processes are decisive in the southern regime.

4.4 ^{234}Th partitioning between particles and water

Latitudinal distributions of specific ^{234}Th activity (SPTh) and partitioning coefficient, K_d ($\text{PTh/DTh} \times 1/\text{TSM} \times 10^6$) are shown in Fig. 6b. The SPTh ranges from 0.01 to 0.4 dpm mg^{-1} , which is comparable with the values found in other nearshore regions, e.g., Jinhae Bay, South Korea (Kim and Yang, 2004), Amazon shelf (McKee *et al.*, 1986) and inner shelf of the Yangtze River (McKee *et al.*, 1984). However, an evident difference of SPTh values was found between the northern and the

southern regimes, concurring with the contrasting phenomenon revealed by other parameters, as discussed above. Except for the two northernmost stations, the SPTh in the northern regime (0.02~0.05 dpm mg^{-1}) is much lower than that in the southern one (0.05~0.38 dpm mg^{-1}).

It can be seen that K_d values range from $10^{4.7}$ to 10^7 ml g^{-1} and the K_d in the region covered by the mouths of KPR and JWR are evidently higher than those in the other region. It should be noted that the particles in this region were enriched with pigments and organic carbon (Fig. 6a). This phenomenon is consistent with the findings of Baskaran and Santschi (1993), who observed that higher biological activity may cause a higher K_d of ^{234}Th in Texan coastal water. The K_d in the nearshore water was also found to increase with increasing bacterial production. This is consistent with the laboratory study on bioaccumulation of thorium with cultured picoplankton, which showed that the bacteria had high affinity for ^{234}Th (Fisher *et al.*, 1987). Lee *et al.* (1993) also reported an elevated volume concentration factor of ^{234}Th with bacterial biomass, indicating that ^{234}Th has higher affinity to particle surfaces as a result of enhanced bacterial activity. In addition to the particle concentration, the partitioning of ^{234}Th is also related to the particle composition and the residence time of particulate matter (Honeyman *et al.*, 1988). Indeed, when compared with the τ_{PTh} in Fig. 4b, K_d seems to be higher with particle removal residence time, implying that the partitioning of Th between dissolved and particulate phases is not in equilibrium in the nearshore water (McKee *et al.*, 1986; Moore *et al.*, 1996).

Although limited data are available on the partitioning of ^{234}Th in shallow nearshore environments, there are a few literature data enable us to gain some insight into ^{234}Th behavior under extremely turbid conditions. The compilation of K_d data obtained from the turbid water covering three orders of magnitude of TSM concentration in nearshore environments show a negative log-log relationship (Fig. 7), consistent with the phenomenon of particle-particle interaction postulated by Honeyman *et al.* (1988) and Honeyman and Santschi (1989). However, it should be pointed out that the TSM- K_d correlation given by Honeyman *et al.* (1988) and Luo *et al.* (1995) resulted in a K_d value almost two orders of magnitude higher at 1 g L^{-1} TSM level. A lower K_d in the nearshore environment than the extrapolated value based on the TSM- K_d relationship obtained from the open ocean, may be a result of the lower affinity of ^{234}Th for the terrestrial clay mineral than algal detritus (Shull and Meyer, 2002). Here we propose a more appropriate TSM- K_d correlation ($n = 73$, $r^2 = 0.51$) for the turbid nearshore water:

$$\log K_d = 6.06 - 0.95 \log \text{TSM}. \quad (7)$$

Fig. 7. Correlation of TSM and distribution coefficient in various nearshore environments (Texan coast: Baskaran and Santschi, 1993; Amazon shelf: McKee *et al.*, 1986; Yangtze shelf: McKee *et al.*, 1984; Jinhae Bay: Kim and Yang, 2004). The linear fitting of the data results in the equation $\text{Log}K_d = 6.06 - 0.95\text{Log}(\text{TSM})$ ($n = 73$, $r^2 = 0.51$).

4.5 POC and PON removal fluxes

The removal fluxes of POC (F_{POC}) and PON (F_{PON}) can be calculated by the following equations:

$$F_{\text{POC}} = R \times D \times \frac{\text{POC}}{\text{PTh}}, \quad (8)$$

$$F_{\text{PON}} = R \times D \times \frac{\text{PON}}{\text{PTh}}, \quad (9)$$

where D is water depth. It is conceivable that reliable estimates of removal fluxes of POC and PON rely on accurate information of POC/PTh and PON/PTh ratios. However, it has been found that the ratios are quite variable as a function of size, space, depth and time, in different oceanic regimes (Buesseler *et al.*, 2006; Santschi *et al.*, 2006 and references therein). Latitudinal distributions of POC/PTh and PON/PTh ratios are shown in Fig. 8a. Both POC/PTh and PON/PTh show great variability and their latitudinal distributions display a similar pattern. The POC/PTh (PON/PTh) ranges from a low value of 2.7 (0.8) $\mu\text{mol dpm}^{-1}$ at the southernmost station to 86.1 (15.7) $\mu\text{mol dpm}^{-1}$ in the region between HLR and DSR. The POC/PTh ratio is closely correlated with TP ($n = 11$, $r^2 = 0.94$, cf. Fig. 3a) at stations to the south of station B18, where SiO_2 concentration was generally low (cf. Fig. 3b) and primary production was high. An increasing POC/PTh ratio in this region may be related to the increasing population of large diatoms. This phenomenon was also observed in the open ocean. For example, Kawakami *et al.* (2004) reported that the POC/PTh ratio increases with increasing population of large diatoms

Fig. 8. Latitudinal distribution of (a) ratios of organic carbon (POC/PTh, circles) and nitrogen (PON/PTh, squares) to ^{234}Th in suspended particles and (b) POC (F_{POC} , circles) and PON (F_{PON} , squares) fluxes calculated by the scavenging model at water sampling stations along the coastline of western Taiwan.

during the bloom. In contrast to the southern regime, there was no correlation between the POC/PTh ratio and pigment concentration in the northern regime, reflecting different particle composition from that in the southern regime, as discussed previously.

Latitudinal distributions of F_{POC} and F_{PON} are shown in Fig. 8b. The F_{POC} and F_{PON} range from 4.5 to 275 $\text{mmol-C m}^{-2}\text{d}^{-1}$ and from 1.3 to 50.1 $\text{mmol-N m}^{-2}\text{d}^{-1}$, respectively. It should be noted that the euphotic depth (80 m) (284 m) was used to calculate the F_{POC} at the southernmost station HT, instead of water depth, which results in 18 $\text{mmol-C m}^{-2}\text{d}^{-1}$, about half of the export flux obtained by Wei *et al.* (2008a). The discrepancy between this study and Wei *et al.* (2008a) is due to the fact that the deficiency of ^{234}Th in the surface water is smaller than the average deficiency in the euphotic layer at this site. In particular, the POC removal fluxes at the five river mouths were lower than the rest of the stations, indicating active resuspension due to higher energy at the river mouths. In order to check the validity of the F_{POC} estimated from the scavenging model, we compare our results with the findings of Kao *et al.* (2006), who estimated the accumulation rate from sedimentation rate and

organic content of the sediments off southwestern Taiwan. The organic carbon accumulation rate estimated by Kao *et al.* (2006) is $37.3 \text{ mmol-C m}^{-2}\text{d}^{-1}$ at the site about 7.8 km to the south of our station B25, in which a similar F_{POC} , $20.9 \text{ mmol-C m}^{-2}\text{d}^{-1}$, is estimated. In addition to the F_{POC} , the total mass flux in this region can be calculated by the quotient of R and SPTh, which gives $8.7\sim 9.7 \text{ g m}^{-2}\text{d}^{-1}$, which is also similar to the sediment accumulation rate of $2.3\sim 13.4 \text{ g m}^{-2}\text{d}^{-1}$ estimated by Kao *et al.* (2006). It can therefore be concluded that the scavenging model provides an accurate estimate of removal flux. Calculated by the quotient of POC and PON concentration and the removal rates, the residence time of POC and PON in the nearshore water off west Taiwan is in the range of 1 to 20 days.

5. Conclusions

The distributions of physical and biochemical parameters allow two distinctive scavenging domains to be identified, separated by $23^{\circ}40' \text{ N}$ in the nearshore water off western Taiwan. As a result of northward transport of riverine particles contributed by the Cho-Shui River, the northern regime is characterized by high suspended load and low biomass, whereas the southern regime is characterized by low suspended load and high chlorophyll.

The scavenging and removal rates of ^{234}Th in the nearshore water were investigated by measuring dissolved and particulate ^{234}Th in the surface water at 27 stations along the coast line of western Taiwan. An advection-scavenging-removal model was applied to these data to estimate the scavenging and removal rates of ^{234}Th in the nearshore water. The model's validity is supported by the agreement of the removal fluxes of ^{234}Th , total mass, and particulate organic carbon between our estimates and other independent approaches, i.e., excess ^{234}Th inventory in the core top and sediment accumulation rate at selected sites.

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