Sediment Trap Experiments in the Water Column off Southwestern Taiwan: ²³⁴Th Fluxes

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The activity of ²³⁴Th ($t_{1/2} = 24.1$ days) in dissolved, particulate and sediment trap samples was determined in the water column off southwestern Taiwan during 2– 4 October, 1993. Vertical ²³⁴Th fluxes measured by the free-floating sediment traps ranged from 363 to 2290 dpm m⁻² d⁻¹ in the upper 450 m. Th-234 fluxes predicted from the irreversible scavenging model concur with those measured by the sediment traps. Comparison of the residence times of particulate ²³⁴Th and particulate organic carbon showed that their respective values differ by a factor of approximately 2~3, which suggests organic carbon is preferentially recycled relative to ²³⁴Th in the euphotic zone.

1. Introduction

Downward flux of particulate materials from ocean surface waters is the controlling mechanism for bringing particle-reactive elements into the interior of the ocean (Goldberg, 1954). In recent years, oceanographers have been devoted to quantifying the material exchange rate between the surface layer and deep ocean. The disequilibria of uranium-thorium series radionuclides serve as a powerful tool for investigating the scavenging and particle removal processes, while sediment traps provide the only direct method of measuring vertical flux of particulate materials in the water column.

Multiple sediment traps mounted either on a free-floating (surface-tethered) or an anchored (bottom-tethered) mooring line have been used to study the particle dynamics in the upper ocean (Coale and Bruland, 1985, 1987; Murray *et al.*, 1989; Wei and Murray, 1991, 1992). Depending on the individual site, discrepancies with variable extents exist between measured and expected ²³⁴Th fluxes. For example, in the eastern Equatorial Pacific Ocean (Murray *et al.*, 1989) and the Black Sea (Wei and Murray, 1991) where free-floating traps were used, measured ²³⁴Th fluxes were greater than those calculated from the disequilibria of ²³⁴Th/²³⁸U profiles. In Dabob Bay, a fjord in Puget Sound (USA), where anchored sediment traps were deployed, measured ²³⁴Th fluxes were smaller than calculated ²³⁴Th fluxes (Wei and Murray, 1992). Buesseler (1991) evaluated the existing ²³⁴Th data and observed that the overall discrepancy can be as high as 3~10 folds. No definite explanation has been given for the disagreement between the measured and calculated ²³⁴Th fluxes. This study was designed to directly measure the vertical ²³⁴Th fluxes and ²³⁴Th/²³⁸U disequilibria in the water column to see how the two approaches compare to each other.

In a previous paper (Hung and Wei, 1992) a vertical ²³⁴Th flux of 1220 dpm m⁻² d⁻¹ out of the euphotic zone and a lower limit of 1600 dpm m⁻² d⁻¹ into the bottom sediment off south-western Taiwan was estimated from ²³⁴Th/²³⁸U disequilibria. This region (22°10' N 120°10' E, water depth = 750 m) was revisited to carry out sediment trap experiments and water column sampling. Vertical fluxes of total mass, ²³⁴Th, ²¹⁰Pb, ²¹⁰Po, trace metals, particulate organic carbon

and nitrogen (POC and PON) were measured by sediment traps deployed at five depths in the water column. Vertical profiles of the aforementioned elements were also determined. This paper will report the ²³⁴Th data and address its relationship with particle cycling.

2. Method

This study was carried out during 2–4 October 1993, in the sea off southwestern Taiwan aboard the R/V Ocean Researcher I (cruise #368). Seawater samples and sediment trap samples were collected and processed as follows:

2.1 Seawater samples

Seawater was collected using 20 l Go-Flo bottles mounted on a CTD rosette. To confirm depth, a salinity sample was taken from each bottle and compared to the CTD profile. Samples to determine the concentration of dissolved oxygen, nutrients, chlorophyll a, POC, and PON were collected from separate casts. Compressed air (at 12 p.s.i.) was used to pressure filter seawater through preweighed 142 mm Nuclepore filters (0.45 μ m pore size) mounted in a Plexiglas 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 ²³⁴Th activity. Parallel samples for dissolved and particulate ²¹⁰Pb and ²¹⁰Po results will be reported in a separate paper.

Filtered seawater (~20 l) was transferred into a cubitainer, acidified with approximately 20 ml of concentrated HCl and spiked with 35 dpm 230 Th yield tracer as well as 60 mg Fe carrier. The samples were then aerated for at least 4 hours to help achieve isotopic equilibrium (Wei and Hung, 1993). Without interrupting the aeration, 12 N NaOH was added to raise the pH to 8. The Fe(OH)₃ precipitates, with adsorbed thorium, were collected by siphoning and filtering, and then dissolved in concentrated HCl to make the samples 9 N HCl. These samples were then passed through an anion exchange column (AG1X-8) and preconditioned by 9N HCl to separate uranium from thorium. Thorium passed through the column was collected and stored for later purification in the laboratory.

After weighing, the filters were decomposed in the laboratory by soaking in ~10 ml of concentrated NH₄OH. The samples were gently heated to evaporate the NH₄OH. Digestion and analytical procedures for determining particulate ²³⁴Th are described in Anderson and Fleer (1982).

2.2 Sediment trap samples

Settling particles were collected by particle interceptor traps (PITS) deployed at five depths (23, 63, 83, 202 and 453 m). The sediment trap used in this study was a modified version of the design by Knauer *et al.* (1979). Each set of sediment trap consists of eight Plexiglass tubes with an aspect ratio of 9.53 and a crossframe made of polypropylene. Two types of tube, with and without a filter holder on the bottom, were used. The tubes were snapped into the crossframe which was clamped onto a 500 m surface-tethered mooring line. The mooring line was 3/4" low stretch Dacron (Champion-braid). The duration of trap deployment was approximately 1.5 days. Similar sediment traps were used in other regions (Wei and Murray, 1991, 1992, 1994).

The trap solutions were prepared by adding approximately 800 g of NaCl to 20 l of filtered subsurface seawater taken near the trap deployment location. Approximately 12 ml of 37% formaldehyde was added into each tube to prevent particle decomposition, except for the tubes

used in the POC/PON traps. Upon recovery, the upper layer of seawater in the traps was siphoned off and the remaining trap solution was gravity drained through a pre-weighed 90 mm Nuclepore filter (0.45 μ m pore size), which was mounted on the bottom of the trap. While applying a slight vacuum, the filters were rinsed with approximately 10 ml of deionized distilled water to remove sea salt. Zooplankton swimmers were removed by hand under microscope. Generally, 5~10 copepods were found in the traps deployed in upper 100 m. The filter papers were stored in a petri dish to determine total mass and ²³⁴Th. Total ²³⁴Th activity on the filters was determined by the same procedures as the particulate samples.

The activity of ²³⁴Th in seawater samples and sediment trap samples was counted using a low background (<0.3 cpm) anticoincidence counter (Tennelec LB-5100) via its β -emitting daughter ²³⁴Pa. The purity of the samples was checked by taking repeated counting over a two month period to confirm that the radioactive decay curve was followed (Wei, 1991). Blank determinations of all reagents and filter paper used for this study indicated negligible contribution. Chemical yield of thorium was estimated by counting spiked ²³⁰Th using silicon surface-barrier detectors (EG&G Ortec 576). The counting efficiencies of the α detectors were calibrated against NIST traceable ²⁴¹Am (Isotope Products Laboratory 387-67-2-2) and ²³⁰Th (Isotope Products Laboratory 387-67-3) standard plates. Activity of ²³⁴Th in seawater samples was corrected back to the sampling time after the ingrowth of ²³⁴Th from ²³⁸U was subtracted. Activity of ²³⁴Th in sediment trap samples was corrected back to the mid point of trap deployment.

3. Results

Depth, concentration of total suspended matter (*TSM*), dissolved (*DTh*) and particulate (*PTh*) 234 Th activities are given in Table 1. The uncertainty in the radioisotope data was estimated according to the propagation of counting error ($\pm 1\sigma$). Total mass fluxes, 234 Th fluxes, and calculated specific 234 Th activity in trap particles are listed in Table 2. Standard deviations of mass fluxes were calculated from duplicate samples. Errors of 234 Th fluxes represent propagated

Depth (m)	<i>TSM</i> (mg l ⁻¹)	DTh (dpm l ⁻¹)	<i>PTh</i> (dpm l ⁻¹)
0	0.33	1.665 ± 0.057	0.200 ± 0.008
10	0.37	1.522 ± 0.094	0.128 ± 0.006
20	0.23	1.735 ± 0.067	0.125 ± 0.009
30	0.33	1.903 ± 0.077	0.139 ± 0.007
40	0.24	1.633 ± 0.078	0.167 ± 0.008
50	0.21	1.774 ± 0.060	0.130 ± 0.007
60	0.29	1.769 ± 0.088	0.126 ± 0.009
80	0.27	1.743 ± 0.061	0.206 ± 0.009
100	0.30	1.822 ± 0.071	0.541 ± 0.026
300	0.24	1.892 ± 0.067	0.164 ± 0.012
500	0.24	2.201 ± 0.105	0.296 ± 0.014
700	0.22	2.106 ± 0.100	0.356 ± 0.017

Table 1. Depth, total suspended matter (TSM) concentration, dissolved and particulate 234 Th collected during 2–4 October 1993. Standard deviations are based on propagated counting error ($\pm 1\sigma$).

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Table 2. Depth, total mass fluxes, ²³⁴Th fluxes, and specific ²³⁴Th activity in trap particles collected during 2-4 October 1993. Standard deviations of total mass fluxes are calculated from duplicate samples. Standard deviations of ²³⁴Th fluxes are based on propagated counting error $(\pm 1\sigma)$.

Depth		ss flux		******	²³⁴ Th			²³⁴ Th	1
(m)	(mg ı	m ⁻² d ⁻¹)	(dpm m ⁻	² đ¹)	(dpm m	g~¹)
23	577	7 ± 55			363 ±	25	0	.63 ± 0	.07
63		1 ± 37			802 ±	62	1	$.23 \pm 0.$.12
83		7 ± 56.			1017 ±			$.55 \pm 0.1$	
202		4 ± 47			2290 ±			.68 ± 0.	
453		3 ± 15			1365 ±			.98 ± 0	
σ _θ	21	22	23	24	25	26	27	28	
θ (°C)	, <u>ę</u>	5	10		15	20	25		
S (psu		34.0		34.2	34.4	4	34.6	34.8	
2 3 4 5 6 7	20 - 40 - 60 - 60 - 60 - 60 - 60 - 60 - 6	θ	•	· · · ·	°°°		800		

Fig. 1. Vertical profiles of temperature, salinity, and potential density. Salinities collected from bottle samples and measured by Autosal[®] are indicated by circles.

counting errors $(\pm 1\sigma)$. Hydrographic and nutrient data are not listed but are available upon request.

The vertical distributions of hydrographic data (salinity, potential temperature, and potential density) and dissolved oxygen/nutrients (PO₄, NO₃, and SiO₂) at the sampling station are shown in Figs. 1 and 2, respectively. TSM concentration, dissolved and particulate ²³⁴Th activity with counting error bars are shown in Fig. 3. Total ²³⁴Th activity as the sum of dissolved and particulate ²³⁴Th activities are also included in this figure. The dotted line drawn at 2.42 dpm l⁻¹ represents ²³⁸U activity calculated from the S-²³⁸U relationship from Ku *et al.* (1977).

As pointed out in Hung and Wei (1992), the interleaving salinity structure (Fig. 1) is due to complicated water mixing in this area. The salinity maximum of 34.8 psu at 145 m is caused by the intrusion of the Kuroshio (Shaw, 1989). The main pycnocline occurred at depths of 80–

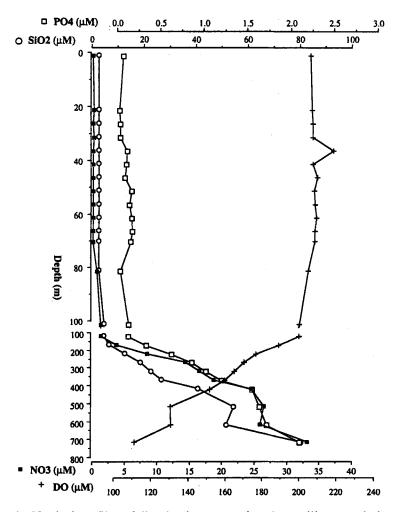


Fig. 2. Vertical profiles of dissolved oxygen, phosphate, silicate, and nitrate.

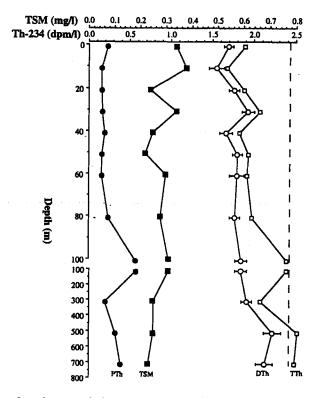


Fig. 3. Vertical profiles of total suspended matter concentration (*TSM*), total (*TTh*), dissolved (*DTh*) and particulate (*PTh*) ²³⁴Th activities. The vertical dashed line at 2.42 dpm l⁻¹ represents ²³⁸U activity calculated from S-²³⁸U relationship. Error bars represent uncertainties based on propagated counting error.

120 m. Nutrients were depleted in the mixed layer, then increased with depth in the pycnocline layer (Fig. 2). Dissolved oxygen decreased from saturation levels in the mixed layer to 185 μ M at S maximum. Below the 200 m depth, dissolved oxygen continued to decrease and nutrients showed a monotonically increasing trend toward the bottom.

In general, vertical distributions of dissolved and particulate 234 Th (Fig. 3) are similar to that of the previous study (Hung and Wei, 1992), except the 234 Th-excess layer was not observed (or was not as evident). It can be seen that all dissolved 234 Th activity was deficient relative to the 238 U activity. Both TSM concentration and particulate 234 Th activity were relatively low throughout the water column and showed little vertical structure except a small particulate 234 Th maximum of 0.5 dpm l⁻¹ at 100 m. Except at 300 m, total 234 Th activity is in equilibrium with 238 U at all depths below 100 m.

Vertical fluxes of total mass and ²³⁴Th are shown in Fig. 4. Total mass flux increases from 580 mg m⁻² d⁻¹ at 20 m to a maximum of 850 mg m⁻² d⁻¹ at 200 m, then decreases to 700 mg m⁻² d⁻¹ at 450 m. The ²³⁴Th flux has a wider range, from 360 to 2290 dpm m⁻² d⁻¹, than the total mass flux. A maximum in ²³⁴Th flux was also observed at 200 m. As shown in Fig. 5, a tight correlation ($r^2 = 0.97$) exists between the total mass fluxes and ²³⁴Th fluxes.

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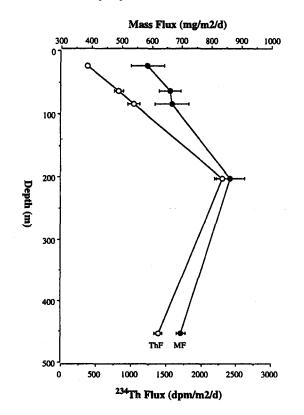


Fig. 4. Vertical fluxes of total mass and ²³⁴Th measured by sediment traps. Error bars associated with total mass fluxes and ²³⁴Th fluxes represent uncertainties based on the standard deviation from duplicate samples and propagated counting error, respectively.

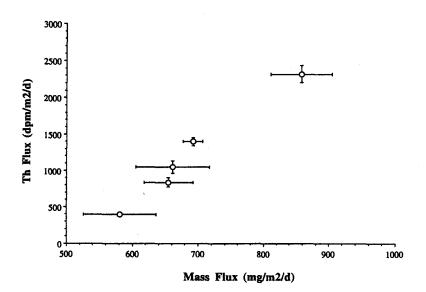


Fig. 5. Correlation of total mass flux and ²³⁴Th flux.

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4. Discussion

In December 1991, vertical profiles of dissolved and particulate ²³⁴Th were determined at a sampling station about 30 km north of this study site. Box-model calculations for the data gave mean residence times of 100 days for dissolved ²³⁴Th and 20 days for particulate ²³⁴Th in the upper water column (100 m) (Hung and Wei, 1992). The same calculations were applied to the profiles obtained from this study and mean residence times of 90 days for dissolved ²³⁴Th and 14 days for particulate ²³⁴Th in the upper 100 m were estimated. The shorter dissolved ²³⁴Th residence time (τ_{DTh}) is consistent with the fact that particle load at our sampling station was about 3 times higher than that of Hung and Wei (1992). The standing stocks of total suspended matter in the upper 100 m of our sampling station and Hung and Wei's (1992) were 28.3 g m⁻² and 11.7 g m⁻², respectively. Partitioning of ²³⁴Th between particulate and solution phases was also affected by the particulate matter concentration. The correlation of distribution coefficient (K_d) of ²³⁴Th and TSM concentrations from this study and the study by Hung and Wei (1992) is shown in Fig. 6. Good correlation of K_d and TSM concentrations indicates internal consistency for the two sets of data obtained from the two cruises.

It should be noted that although the sampling site is near the coast (about 25 km offshore), the τ_{DTh} obtained from this study and Hung and Wei (1992) are both long compared to other coastal regimes, which typically have τ_{DTh} values of days. Relatively long τ_{DTh} implies that the region has oligotrophic characteristics of the open ocean. Coale and Bruland (1985) observed the scavenging rate constant of dissolved ²³⁴Th (reciprocal of τ_{DTh}) is proportional to primary productivity in the California Current. A primary production rate of only 130 mg C m⁻² d⁻¹ was estimated if we assumed the proportionality can be applied to our station. It should be noted that a large uncertainty (~50%) may be associated with the estimate.

Recently, concerns regarding the validity of using PITS type sediment traps to determine the sinking particle flux have been raised. Buesseler (1991) compared the existing ²³⁴Th fluxes

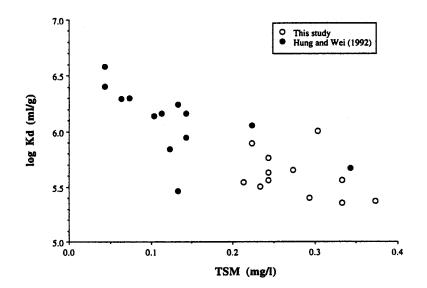


Fig. 6. Correlation between the distribution coefficient (K_d) and total suspended particle (TSM) concentration for data collected from this study (open circle) and Hung and Wei (1992) (solid circle).

measured directly by sediment traps and estimated from the irreversible scavenging model and concluded that the shallow sediment traps cannot provide accurate flux information due to a relatively large discrepancy (3 to 10 folds) between the two fluxes.

According to the irreversible scavenging model the particle removal rate for 234 Th (i.e., *P* flux, Coale and Bruland, 1985) can be calculated by

$$P = \lambda (U - TTh) \tag{1}$$

where λ is the decay constant of ²³⁴Th (0.0288 d⁻¹), U is ²³⁸U activity (dpm l⁻¹), and TTh is total ²³⁴Th activity (dpm l⁻¹). It should be noted that the P flux calculated from (1) has a rate unit, dpm l⁻¹ d⁻¹. In order to compare with measured ²³⁴Th flux data the rate unit should be converted into a flux unit,

²³⁴Th flux =
$$\Sigma[(P_i + P_{i-1})(Z_i - Z_{i-1})/2]$$
 (2)

where P_i is successive P flux calculated from discrete ²³⁴Th data at depth Z_i in the water column. The comparison of modeled ²³⁴Th flux and measured ²³⁴Th flux is shown in Fig. 7.

It can be seen that ²³⁴Th fluxes predicted from the model and measured by sediment traps are almost identical in the upper 100 m. The irreversible scavenging model predicted that the vertical ²³⁴Th flux continues to increase to 3700 dpm m⁻² d⁻¹ at the depth of 300 m. Th-234 flux

²³⁴Th Flux (dpm/m2/d)

Fig. 7. Vertical fluxes of ²³⁴Th measured by sediment traps (open circle) and calculated from the irreversible scavenging model (solid circle). Error bars represent uncertainties based on propagated counting error.

at 200 m measured by the sediment trap falls on the linear interpolation line between the modeled fluxes at 100 and 300 m. For the deeper layer, the residence time of suspended particles is too long to be resolved by the ²³⁴Th/²³⁸U disequilibrium. Much less ²³⁴Th flux measured at 450 m indicates a net decomposition process for the suspended particles or a horizontal transport of particles in the mid-layer (~200 m). However, we suspect the later is important because no elevated TSM concentration was found (Fig. 3). The excellent agreement between the modeled and measured ²³⁴Th fluxes suggests our sediment trap does provide reliable vertical flux measurements in the euphotic layer off southwestern Taiwan. Although the results are very encouraging, potential problems with this type of sediment trap still exist (Buesseler, 1991). Furthermore, using simplified scavenging model to calibrate measured ²³⁴Th flux may not be valid because the model assumes a steady-state and neglects advection/diffusion effects on the distribution of ²³⁴Th.

Based on the findings that the scavenging rate constant of 234 Th correlates with the primary production rate (Coale and Bruland, 1985) or with the flux of particulate organic carbon (Bruland and Coale, 1986), Eppley (1989) suggested that 234 Th can be used as a tracer for organic carbon in the euphotic zone to estimate new production. Murray *et al.* (1989) tested the hypothesis by comparing the residence times of organic carbon and 234 Th in the euphotic layer of the eastern equatorial Pacific. They found that the residence time of 234 Th is approximately one third of carbon, and concluded that using 234 Th as a carbon tracer was not justified. With data available in this study, we compared the residence times of total suspended matter (τ_{TSM}), particulate 234 Th (τ_{PTh}), and particulate organic carbon (τ_{POC}). The residence times were calculated by dividing the inventory of each parameter by flux out. The values for 234 Th were also calculated by the irreversible scavenging model. The results are summarized in Table 3.

The τ_{PTh} in the upper 60 m is 8~12 days when calculated from the water column ²³⁴Th data or 11 days when calculated from the sediment trap flux. Generally, the τ_{PTh} calculated from the two approaches agree well with each other. The τ_{POC} in the same layer is 25 days, approximately 2~3 times the τ_{PTh} . The difference between τ_{PTh} and τ_{POC} seems to be a common feature shared by different oceanic regimes. Coale and Bruland (1985) observed that τ_{PTh} is about half the τ_{POC} in the California Current system while Murray *et al.* (1989) demonstrated that τ_{PTh} is about one third of carbon in the eastern equatorial Pacific. As a result, new production will be overestimated by a factor of 2~3 if τ_{PTh} was assumed to be an estimate of τ_{POC} . The τ_{TSM} (26 days) is similar to the τ_{POC} , implying that the biogenic particles dominate the bulk suspended load in the upper 60 m of the site.

	$ au_{PTh}$ (c			
$ au_{TSM} (day)$	$PTh/\lambda(U-TTh)^*$	ΣPTh/ThF**	$ au_{\rm POC}~({\rm day})$	
26	8-12	11	25	

Table 3. Residence times of total suspended matter (τ_{TSM}), particulate ²³⁴Th (τ_{PTh}), and particulate organic carbon (τ_{POC}) in the upper 60 m.

*Based on the mass balance equation for particulate 234 Th: $0 = dPTh/dt = \lambda(U - DTh) - \lambda PTh - P$. ** ΣPTh calculated by trapezoidal summation from the surface to 60 m.

5. Conclusion

A home-made sediment trap was successfully deployed in the sea off southwestern Taiwan to directly measure the vertical fluxes of total mass and ²³⁴Th. At the same location, large volume seawater samples were also taken to measure the dissolved and particulate ²³⁴Th profiles. A comparison of the ²³⁴Th fluxes predicted from the water column ²³⁴Th data and measured by the sediment trap was made and excellent agreement found between the two values. It was concluded that ²³⁴Th cannot quantitatively trace organic carbon in the euphotic layer, and new production will be overestimated by a factor of 2~3 if the residence time of particulate ²³⁴Th is used as an export rate parameter for carbon.

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