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Lead-210 and Polonium-210 Across the Frontal Region Between the Kuroshio and the East China Sea, Northeast of Taiwan

CHAO-YEUH YANG * and HUI-CHING LIN*

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ABSTRACT

Concentration profiles of Pb-210, Po-210 were measured on several KEEP (Kuroshio Edge Exchange Process) program stations. Results of both Pb-210 and Po-210 of surface water along B transect (from station 6115 to station 4531) showed an increasing trend toward deeper water. This reflected the more efficient removal of Pb-210, Po-210 from the shelf water. Vertical profiles of deep water station (i.e. 4531, 5131) did show Po-210 deficiencies, Po-210/Pb-210 < 1, at upper layer (< 100m); and Po-210 excess in the middle layer (200m-500m).

With Box Model calculation, the recycling efficiency of Pb-210 and Po-210 in the middle layer is found to be 2.9% and 85.4%. Vertical profile of the near shelf-break station 512A, where upwelling or intrusion of Kuroshio water occurred, showed the feature of richness in Po-210. The profile of the station 5323, located at a very steep upper slope region, showed the feature of Pb-210 excess all over the water column. Some sediment particles which contains excess Pb-210 might re-export from the outer shelf to slope water. The shelf water station was characterized with Po-210 excess in most of the water column except the very top layer (< 20m).

1. INTRODUCTION

Pb-210 and Po-210 are members of the naturally-occurring uranium decay series. Both of them are highly reactive and tend to associate with particulate phases. Pb-210 is produced throughout the water column following the decay of Ra-226. In shallow water, the predominant source of Pb-210 is atmospheric fallout [Benniger, 1978], where it is formed by decay of Rn-222. Almost all of the Po-210 in seawater is formed in situ by the decay of Pb-210, so its rate of supply can thus be accurately determined. Pb-210 is strongly depleted relative to Ra-226 by bottom and boundary scavenging processes (Bacon *et al.*, 1976; Nozaki *et al.* Spencer *et al.*, 1981; Cochran *et al.*, 1983). Po-210 is more biologically active than Pb-210 (Kharkar *et al.*, 1976), however, Po-210 is also recycled more effectively than Pb-210. Due

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* Institute of Oceanography, National Taiwan University, Taipei, Taiwan, R.O.C.

to atmospheric Pb-210 input, the ratio of Po-210/Pb-210 is less than one in all surface waters. In the subsurface water, where biological recycling of Po-210 dominates, a layer that is rich with Po-210 relative to Pb-210 is found. In the deep water, since the half-life of Po-210 (138 days) is much shorter than its parent Pb-210 (22.3 years) , a secular equilibrium is reached, so the Po-210/Pb-210 activity ratio becomes unity. The deviation of the activities of Po-210 and Pb-210 from their equilibrium values have been used to determine the removal rates of these nuclides from the ocean. Using a box model, by material balance caculations for Ra-226, Pb-210 and Po-210 (Bacon *et al.*, 1976), we are able to study the efficiency of Pb-210, Po-210 recycling in the oceans.

The Kuroshio, a conduit linking the East China Sea to the Pacific Ocean proper, flows northward in the areas just off northeast Taiwan. The blocking of the Kuroshio by the continental shelf-break causes it to turn slightly to the right. A shelf-slope circulation pattern with lateral water movement is generated to result in a major exchange between the East China Sea and the Kuroshio. The topographically-induced upwelling of the Kuroshio subsurface water at the shelf break near northeast of Taiwan appears to be a permanent feature (Chern *et al.*, 1990), probably as a result of the periodical meandering and shelf-ward migration of the Kuroshio. Sometimes the Kuroshio also intrudes onto the shelf of the East China Sea, thus having a major effect on the composition of the shelf waters.

The large concentration gradient of Pb-210 across the frontal region dividing the slope water from the shelf water at south of New England was observed by Bacon (Bacon *et al.*, 1988). This phenomenon implies the continental shelf may be an important sink of Pb-210 from the open ocean. Dissloved Pb-210 and Po-210 may be exported from the Kuroshio to the East China Sea where they are transferred to the particulate phase and eventually buried in the sediment. Recently Nozaki reported an unusually large Po deficiency relative to Pb near our study area (Nozaki, 1990). The other available paper (Lin *et al.*, 1991) presents the distributions of Pb-210 and Po-210 and the extent of their radioactive disequilibrium in areas off eastern and northeastern Taiwan. This paper presents our effort to study the partition of Pb-210 and Po-210 between the dissolved and particulate phases in KEEP area. The determination of residence time, scavenging rate and recycling efficiencies of Pb-210 and Po-210 are the major focuses of this study.

2. EXPERIMENTAL

2.1 Sample collection

Twelve stations were occupied during KEEP (Kuroshio Edge Exchange Process) cruise (No. 249, 258, 264, 276) by R/V Ocean Researcher I in 1991-1992 (Figure 1). A listing of the characteristics of the stations is shown in Table 1. Most stations were along B transect, normal to the 200-m isobath at the shelf-break just off northeast Taiwan. Samples of surface water were collected at all stations. Samples were also collected at various depths at stations 4531 and 5131 in the Okinawa Trough, station 6115 in the shelf of the East China Sea and station 5323 and 512A at the shelf-break.

2.2 Experimental methods

The analytical procedures used are similar to those described by Fleer and Bacon (1984). A more detailed description of experimental procedure is described in Yang *et al.* (1991). Briefly, for each sample 20L water was collected and filtered in succession with an in-line AMF Cuno Micro Wynd filter and Gelman A/E glass fiber filter. The filtered sample was acidified with concentrated HCl to a pH of 2. Then, spike known dpm of Po-209 and 5mg of Pb (as a lead nitrate solution) were added to the water sample. Those tracers were allowed

Table 1. Remarks on the seawater samples

Station	Cruise No.	Location	Date
4324	249	24° 30'N 122° 40'E	1990-08-10
6021	249	26° 00'N 122° 10'E	1990-08-09
5221	249	25° 20'N 122° 10'E	1990-08-10
5131	249	25° 10'N 123° 10'E	1990-08-12
6115	258	26° 10'N 121° 50'E	1990-10-24
5521	258	25° 50'N 122° 10'E	1990-10-23
5125	258	25° 10'N 122° 50'E	1990-10-23
4531	258	24° 50'N 123° 10'E	1990-10-22
512A	260	25° 05'N 122° 15'E	1990-11-07
4531a	260	24° 50'N 123° 10'E	1990-11-05
6115	264	26° 10'N 121° 50'E	1990-12-06
5323	276	25° 30'N 122° 30'E	1991-04-10
4531b	276	24° 50'N 123° 10'E	1991-04-10

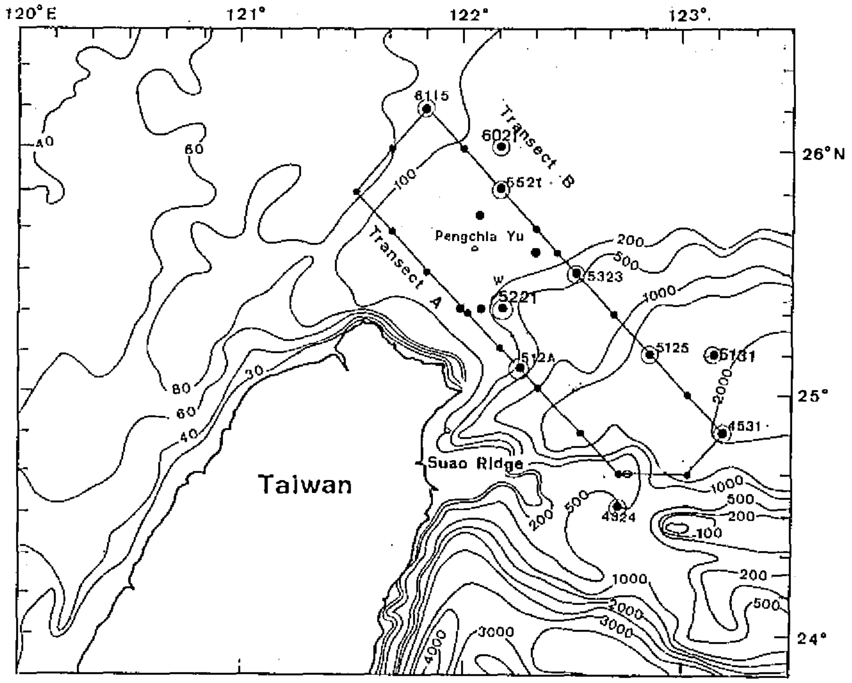

Fig. 1. Sampling location from different cruises.

to equilibrate for a minimun of 12 hours. Then, a cobalt and APDC were added to the sample. The ppt formed was collected by filtering the sample in succession through Gelman A/E glass fiber filter and a Whatman glass microfiber filter. The precipitates were redissolved in a mixture of concentrated nitric acid and hydrochloric acid. The Po-209 and Po-210 in the solutions were then plated into a silver disk and analyzed by alpha spectrometry. The remaining solution which contains Pb-210 will be stored for a known amount of time of no less than six months so that a new batch of Po-210 may be formed as the daughter of the Pb-210. This grow-in Po-210 again will be deposited onto a silver disk and determined by alpha spectrometry. The Pb-210 activity was calculated from the Po-210 value as described in our earlier report (Yang *et al.*, 1991). For the determination of particulate Pb-210 and Po-210 , the collected particles were spiked with Po-209 tracer then dissolved with the concentrated nitric acid and perchloric acid gently, in a Telfon beaker, evaported to near dryness, then 2ml hydrochloric acid were added, followed by the same procedure as measuring dissolved Pb-210 and Po-210. The system uncertainties in the analytical method includes one sigma counting errors and other errors are 5% and 10% each for dissolved and particulate phases analyses.

3. RESULTS

All of the samples analysed were collected during KEEP cruise (No. 249, 258, 260, 264, 276) in August 1990-April 1991. Results of the Pb-210, Po-210 analyses are listed in Table 2 and 3.

Table 2. Total activities of Pb-210 and Po-210 from KEEP cruise

Station	Depth	Pb-210 (dpm/100kg)	Po-210 (dpm/100kg)	Po-210/Pb-210
4324	0	9.2 ± 0.5	4.5 ± 0.2	0.49
	100	14.8 ± 0.7	12.6 ± 0.6	0.85
	200	14.2 ± 0.7	15.4 ± 0.8	1.08
	500	7.9 ± 0.4	10.8 ± 0.5	1.37
	700	7.6 ± 0.4	7.5 ± 0.4	0.99
5131	0	9.2 ± 0.5	4.5 ± 0.2	0.49
	50	8.6 ± 0.4	6.3 ± 0.4	0.73
	100	12.0 ± 0.6	7.8 ± 0.4	0.65
	200	10.4 ± 0.5	11.4 ± 0.6	1.10
	500	10.8 ± 0.5	11.0 ± 0.6	1.02
	1000	4.0 ± 0.2	2.1 ± 0.1	0.53
	1500	3.7 ± 0.2	2.8 ± 0.1	0.76
4324	0	9.2 ± 0.5	4.5 ± 0.2	0.49
6021	0	8.2 ± 0.4	4.5 ± 0.2	0.55
5221	0	8.9 ± 0.4	6.8 ± 0.3	0.76
5131	0	15.1 ± 0.8	7.8 ± 0.4	0.52
6115	0	10.7 ± 0.5	6.1 ± 0.3	0.57
5521	0	13.8 ± 0.7	6.8 ± 0.3	0.49
5125	0	16.7 ± 0.8	10.3 ± 0.5	0.62
4531	0	20.8 ± 1.0	13.8 ± 0.7	0.66
4424	0	20.0 ± 1.0	10.9 ± 0.5	0.54

Table 3. Activities of dissolved and particulate Pb-210, Po-210 from KEEP cruise

Station	Depth	Pb-210 (dpm/100kg)		Po-210 (dpm/100kg)		Po-210/Pb-210	
		(D)	(P)	(D)	(P)	(D)	(P)
5323	0	6.5±0.3	5.9±0.6	3.7±0.2	3.8±0.4	0.57	0.65
	50	6.2±0.3	2.6±0.3	5.1±0.3	1.2±0.1	0.82	0.46
	100	12.3±0.6	3.5±0.4	6.7±0.3	0.9±0.1	0.54	0.26
	200	10.7±0.5	2.3±0.2	6.2±0.3	0.8±0.1	0.58	0.35
	250	10.0±0.5	2.2±0.2	8.2±0.4	1.0±0.1	0.82	0.45
4531b	0	15.5±0.8	1.6±0.2	7.8±0.4	2.2±0.2	0.50	1.38
	10	16.2±0.8	1.7±0.2	8.0±0.4	2.9±0.3	0.49	1.71
	30	15.0±0.8	1.5±0.2	8.1±0.4	1.4±0.1	0.54	0.93
	40	10.5±0.5	1.2±0.1	7.1±0.4	4.0±0.4	0.71	3.33
	60	15.1±0.8	1.6±0.2	8.0±0.4	1.7±0.2	0.53	1.07
	80	16.2±0.8	1.8±0.2	8.4±0.4	2.1±0.2	0.52	1.17
	100	15.4±0.8	3.5±0.4	9.6±0.5	1.4±0.1	0.62	0.40
	200	11.5±0.6	1.2±0.1	14.0±0.7	0.3±0.03	1.22	0.25
	300	10.3±0.5	1.8±0.2	13.1±0.7	1.2±0.1	1.27	0.67
	500	8.0±0.4	0.8±0.1	7.5±0.4	1.7±0.2	1.07	2.13
6115	4	4.1±0.2	0.9±0.1	1.4±0.1	1.8±0.2	0.34	2.00
	20	2.5±0.1	1.1±0.1	2.5±0.1	.4±0.1	1.00	1.27
	40	2.1±0.1	1.5±0.2	3.1±0.2	1.3±0.1	1.48	0.87
	75	1.9±0.1	2.4±0.2	4.2±0.2	1.7±0.2	2.21	0.71

D:Dissolved, P:Particulate

4. DISCUSSION

(1). Distribution of Pb-210 and Po-210 along B transect

Results of Pb-210 and Po-210 of surface water along B transect (as shown in Figure 2) showed an increasing trend toward deeper water station. This reflected the more efficient removal of Pb-210 and Po-210 from the shelf water.

(2). Vertical Distribution Profiles of Pb-210 and Po-210 on KEEP stations

The profiles of total activity of Pb-210 and Po-210 on station 4324 and 5131 are shown in Figures 3, 4. They are in agreement with the profiles of station R19 and R23 in the paper of Lin *et al.*, (1991). All these four stations share a common feature in that they are very close to the Kuroshio axis. The Kuroshio flows northward along the east coast of Taiwan (passes R23 station first), then squeezes through the passage over a ridge between Taiwan and the Yonakunijma Island (R19 and 4324 are nearby) and enters the Okinawa Trough (where 5131 station is). It is interesting to point out : the Kuroshio water after being altered by squeezing through the passage over the ridge still has the same Pb-210 and Po-210 profiles. Station 512A is located very close to the shelf-break. The profile of dissolved and particulate Po-210 activities on station 512A (Figure 5) nevertheless are very unusual. Due to topographic effect and possibly the intrusion of Po-210 rich Kuroshio middle layer water, the maximum Po-210 activities for both dissolved and particulate phases are observed at 200-m depth. The result agrees with what Bacon (1988) reported : " Some of the Po-210 regeneration on the outer shelf may occur at the sediment-water interface ".

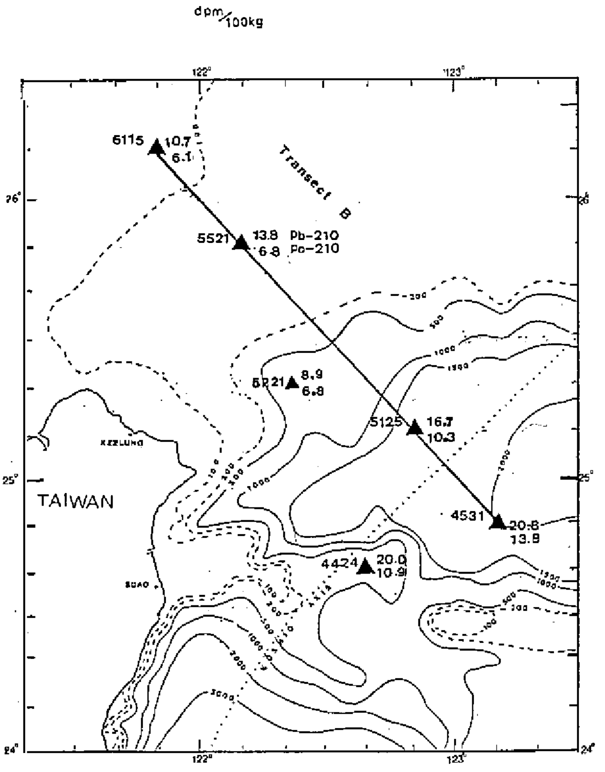

Fig. 2. Distribution of Pb-210 and Po-210 in surface seawater along B transect.

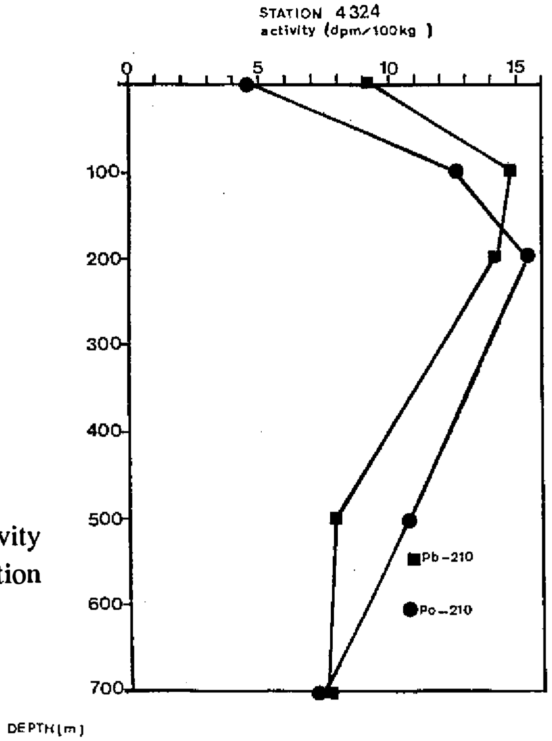

Fig. 3. Vertical profiles of total activity of Pb-210 and Po-210 at station 4324.

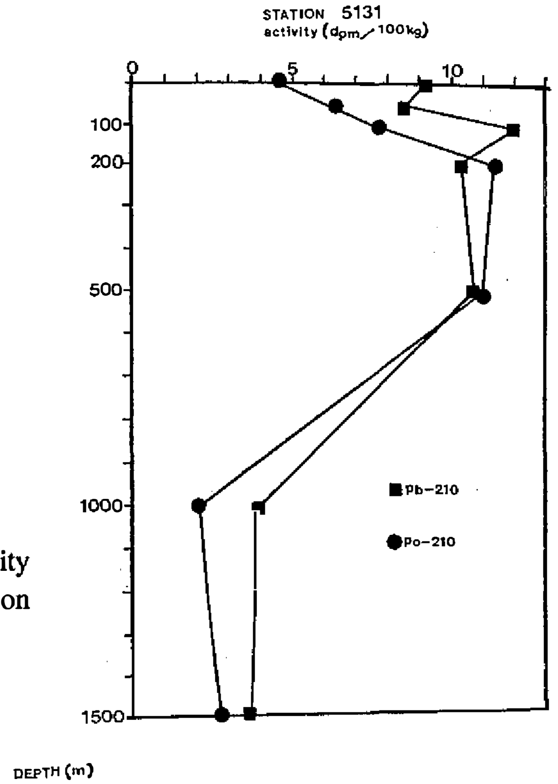

Fig. 4. Vertical profiles of total activity of Pb-210 and Po-210 at station 5131.

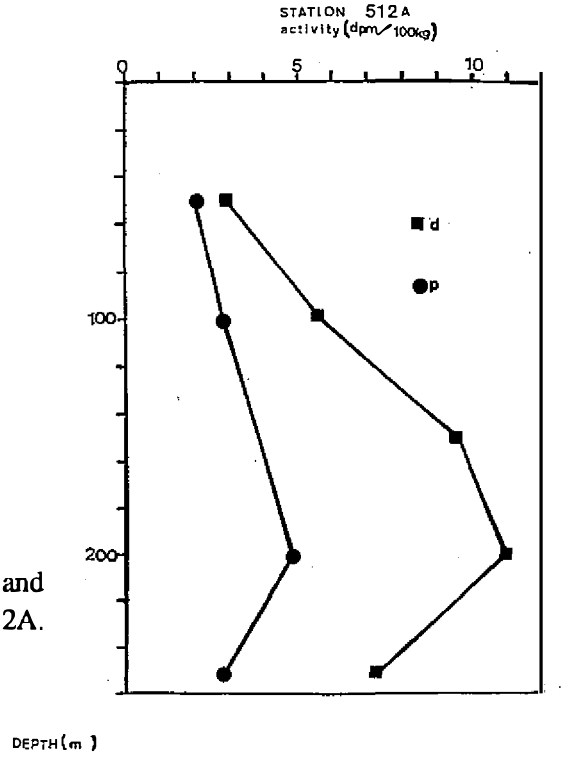

Fig. 5. Vertical profiles of dissolved and particulate Po-210 at station 512A.

Three vertical profiles of Pb-210 and Po-210 on station (4531, 5323, 6115) along B transect are shown in Figures 6, 7 and 8. The dissolved activities of Pb-210 and Po-210 profiles of station 4531 are as shown in Figure 6. We confirmed what Nozaki (1990) reported : " Unusually large Po-210 deficiencies relative to Pb-210 in the Kuroshio current of the East China Sea and Philippine Seas" . Yet our data value are smaller than theirs. Vertical profile on station 4531 indicated Po-210 deficiencies (Po-210/Pb-210<1) at upper layer (<100m) and Po-210 excess in the middle layer (200m-500m). The vertical profiles of dissolved activities of Pb-210 and Po-210 on station 5323 is as shown in Figure 7. The special feature of this profile is that there is no intersection of Pb-210 and Po-210. The concentration of suspended matter at this station is much higher than that of station 4531. With higher productivity, these biogenic particles might take up Po-210 easily to diminish the activity of dissolved Po-210. Since Po-210 is preferentially taken up by plankton but Pb-210 remains in solution to cause no intersection of Pb-210 and Po-210 occurred in their profiles. Station 5323 is located at a very steep continental slope area. Within one mile range, the depth could increase from 300-m to 500-m. Here the seafloor contour from sonar system shows drastically up and down changes. Possibly a fraction of slumping sediment particles which contains excess Pb-210 may re-export from the East China Sea to the Kuroshio by isopycanal mixing or horizontal advection to cause high particulate Pb-210 activity on station 5323. The high activity value of dissolved Pb-210 relative to Po-210 on station 5323 is due to high atmospheric input (Turekian *et al.*, 1997) and the North Pacific gyre circulation results in the deepest thermocline for Pb-210 penetration. Vertical profile of station 6115 is as shown in Figure 8. Dissolved Pb-210 and Po-210 got one intersection at 20m depth. The profile of this station was characterized by Po-210 excess relative to Pb-210 in most of the water column, i.e. over 20m in depth. This profile indicates either the regeneration of Po-210 dominates in this region, or the intrusion of Kuroshio middle layer water.

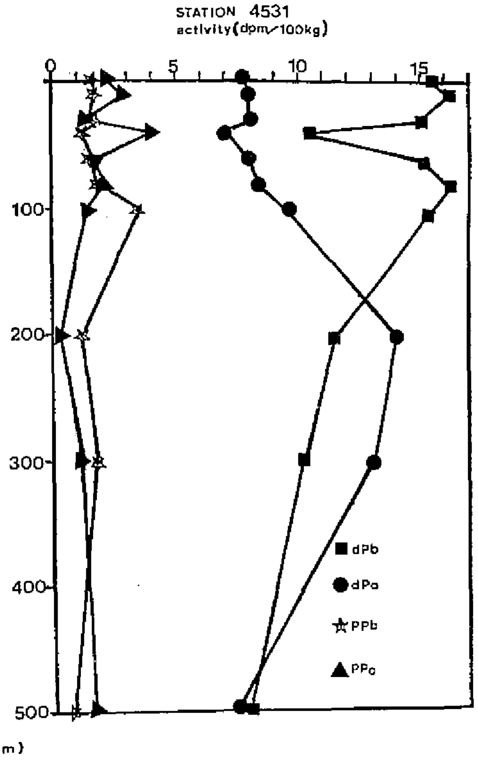

Fig. 6. Vertical profiles of dissolved and particulate Pb-210 and Po-210 at station 4531.

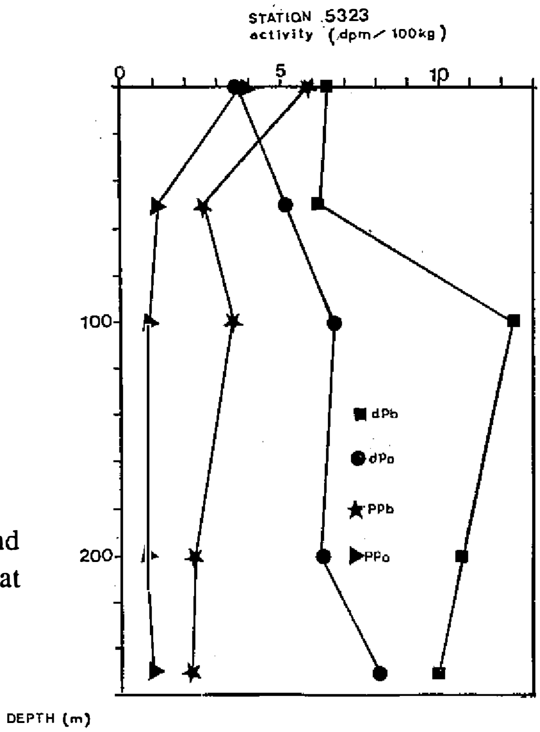

Fig. 7. Vertical profiles of dissolved and particulate Pb-210 and Po-210 at station 5323.

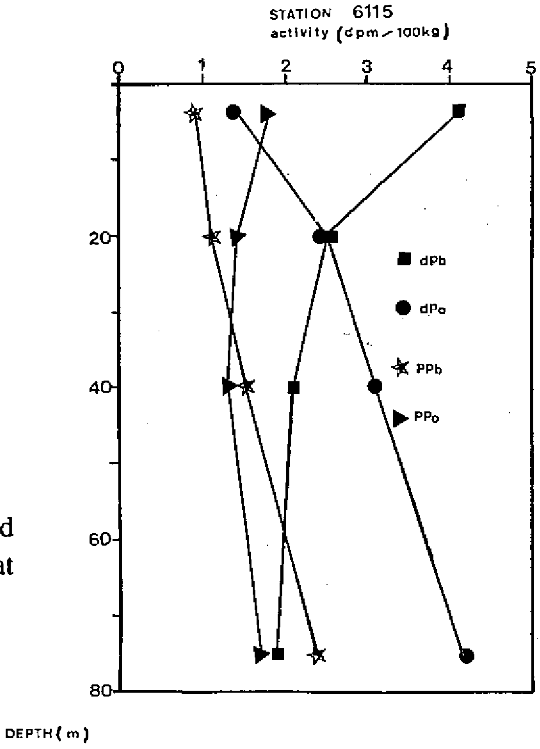

Fig. 8. Vertical profiles of dissolved and particulate Pb-210 and Po-210 at station 6115.

(3). The Scavenging Residence Time of Po-210 and Pb-210 and Its Significance

Assuming a steady state and negligibly small mixing effect, by applying a mass balance model (Santschi *et al.*, 1979), the scavening residence time of Pb-210 and Po-210 in a body of water can be estimated. Thus, for Po-210:

$$T_c = \frac{(A_d/A_p)}{[1-(A_d/A_p)]}(1/\lambda_d)$$

where A_d/A_p is the activity ratio osf Po-210 and Pb-210 ,λd is the Po-210 decay constant (1.82 yr^{-1}), T_c is the scavenging residence time for Pb-210:

$$I + \lambda Ra\ NRa = \lambda_{P}b\ N_{P}b + L_c\ N_{P}b$$

where I is the atmospheric input flux. L_c is the biochemical removal rate constant

$$I + A_p = A_d + L_c(A_d/\lambda_d)$$

$$L_c = \frac{I + A_p - A_d}{A_d/\lambda_d}$$

$$= \frac{[(I/A_p) + 1 - (A_d/A_p)]\lambda_d}{A_d/A_p}$$

$$T_c = 1/L_c$$

$$= \frac{1}{\lambda_d} \times \frac{(A_d/A_p)}{[(I/A_p) + 1 - (A_d/A_p)]}$$

I and A_p are estimated to be $2dpm/cm^2 \cdot yr$, $0.061dpm/L$ The scavenging residence times calculated are tabulated in Table 4.

Table 4. Scavenging residence time of Pb-210 and Po-210 in surface waters

station	Pb-210 residence time (yr)	Po-210 residence time (yr)
4324	1.50	0.52
6021	1.33	0.67
5221	1.45	1.74
5131	2.54	0.59
6115	1.76	0.73
5521	2.31	0.52
5125	2.84	0.86
4531	3.61	1.10
4424	1.76	0.64

Po-210 is scavenged from the surface mixed layer at all stations. The mean ratio of Po-210/Pb-210 is 0.56. The calculated mean residence time is 0.7 year. The calculated Pb-210 residence time is all over one year, longer than that of Po-210. Furthermore, the scavenging residence time in shelf waters is shorter then that in the slope water. This variation in the scavenging residence time is indicative of the effectiveness of the ocean margins as a sink for particle-reactive species.

(4). Box Model Calculation for station 4531 Profile

With a box model, material balance calculations for Ra-226, Pb-210 and Po-210 adopted by Bacon *et al.* (1976) are used to calculate the residence time, output and re-cycled fluxes of Pb-210 and Po-210 for the vertical profile of station 4531.

The standing crops of Ra-226 (cited from Nozaki's data), Pb-210 and Po-210 in each of the two layers are listed in Table 5. The values are obtained by computing the arithmetic averages of all data points in the given depth intervals multiply the density of sea water and length of the layer. The calculation procedures are similar to what Bacon *et al.*, 1976, Lin *et al.*, 1991 did. The result is shown in Table 6.

The Pb-210 scavenged by sinking particles is not being recycled back to the dissolved phases. On the contrary, the Po-210 removed from the upper layer is largely recycled in the middle layer by dissolution of the sinking organic particles or by desorption of Po-210 from sinking particles.

The mean residence times are 1.49 and 0.66 years for Pb-210 and Po-210 within the upper layer. These values are comparable to 1.7 and 0.6 years obtained by Nozaki (1976) for the surface water in the North Pacific. The residence time of Po-210 is shorter than that of Pb-210, possibly because Po-210 is being preferentially taken up by plankton.

Table 5. Inventory of Ra-226, Pb-210 and Po-210 in the upper and middle layers for station 4531 in the Kuroshio water. Values are the arithmetic averages of all data points in the given depth intervals.(units are dpm/cm)

Depth interval (m)	I(d) (Ra-226)	I(d) (Pb-210)	I(p) (Pb-210)	I(d) (Po-210)	I(p) (Po-210)
upper layer 0-100 m	0.63	1.52	0.19	0.83	0.23
middle layer 100-500 m	3.19	4.08	0.52	4.74	0.63

d:dissolved, p:particulate

Table 6. Results of Box model calculations for station 4531 in the Kuroshio water

F (Pb-210) (dpm/cm .yr)	mean residence times of nuclides in the upper layer τ (yr)			recycling efficiency of nuclides in the middle layer (%)	
	Pb-210 (d)	Po-210 (d)	Po-210 (p)	Pb-210	Po-210
1.01	1.49	0.66	0.19	2.9	85.4

d:dissolved, p:particulate

(5). The Interpretation of K_d and F Values at Station 4531 and 5323

Similar to what Bacon *et al.*, (1988) reported, the distribution coefficient K_d is given by

$$K_d = C_p/(C_d * TSM)$$

where C_p and C_d are concentrations of the nuclides in particulate and dissolved forms (Table 3), and TSM is the total suspended matter concentration (Table 7). The fractionation factor F(Po/Pb) is simply the ratio of K_d(Po)/K_d(Pb). Values are plotted in Figures 9-12.

For Po-210, in the upper layer (0-100m), the $logKd$ values are slightly larger than that of Pb-210 at station 4531, as shown in Figure 9. The fractionation factors are larger than unity indicating the preferential uptake of Po-210 relative to Pb-210 by plankon (Shannon *et al.*, 1970). But our F values are not so big as Bacon's (1988), probably due to the concentration of aluminosilicates in particulates in our study area (Chester *et al.*, 1976) being larger than theirs. Minimum values in K_d(Po) and K_d(Pb) are found at 200m depth, due to destruction of the Po-bearing particles and regeneration of Po-210. This depth is much deeper than that found by Bacon (1988) for SEEP-I stations. For Pb-210, 10w K_d values are found in the upper layer, slightly higher K_d value in middle layer (100-500m). Station 5323, located at upper slope region, has relatively higher K_d value for Pb-210 than Po-210 (almost all over the water column) as shown in Figure 11, $logF$ values are less than 0. A Pb-210 excessive particulate, due to slumping or turbid current, might export from outer shelf and function as a second source for Pb-210.

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Table 7. Total suspended matter concentrations

station	depth (m)	TSM (mg/kg)
5323	0	0.57
	50	0.20
	100	0.19
	200	0.16
	250	0.15
4531	0	0.35
	10	0.47
	30	0.43
	40	0.40
	60	0.38
	80	0.37
	100	0.33
	200	0.25
	300	0.22
	500	0.25

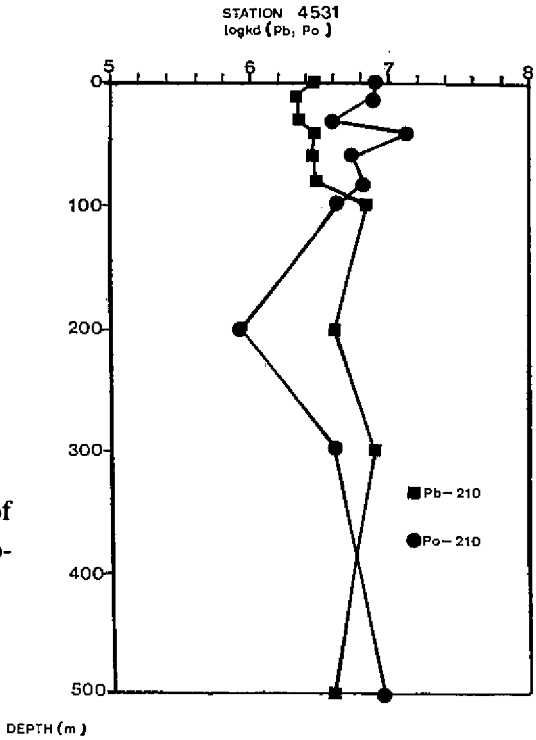

Fig. 9. Vertical profiles of log values of the distribution coefficient of Pb-210 and Po-210 at station 4531.

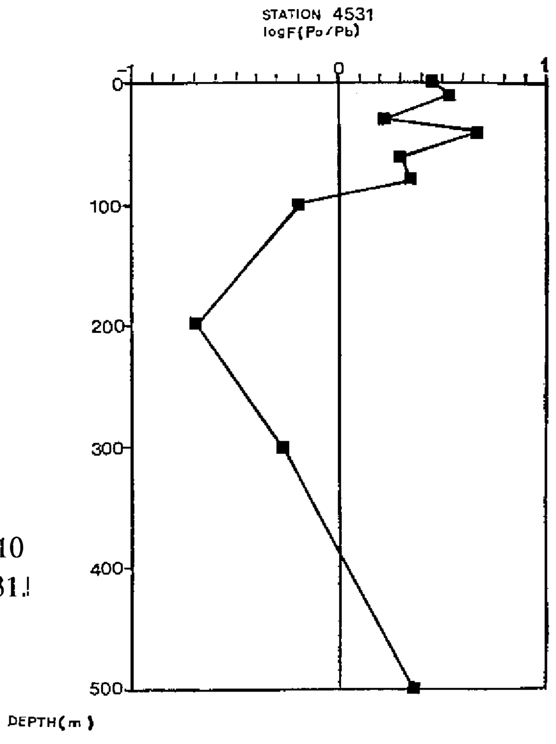

Fig. 10. Vertical profiles of Po-210/Pb-210 fractionation factor at station 4531.

臺灣東北部黑潮與東海交界鋒面
鉛-210，釙-210之研究

楊肇岳、林慧菁
國立臺灣大學海洋研究所

摘要

吾人測量幾個〝黑潮邊緣交換過程〞大型計劃測站的鉛-210和釙-210的垂直剖面。在B橫斷陸棚路線測站的表水中，鉛-210和釙-210的活性有由淺水站往深水站漸增的趨勢，這反應了在陸棚水中的鉛-210和釙-210被較有效率的清除。而 4531,5131 的深水站垂直剖面確顯示在上層水(深度小於100米)釙-210係缺乏，釙-210比鉛-210小於1，而在中層水釙-210係超量(深度200米至500米)。以簡單的盒子模式計算，可求出由表層去除的釙-210有85.4%在中層水中釋放而再循環，鉛-210則僅有2.9%再循環。512A站近陸棚邊緣斷崖，時有湧升或入侵的黑潮水，造成此站釙—210活性富集。5323站位於大陸棚坡上端，極爲陡峭，此站鉛—210在整個水柱中均較釙-210過量，係由帶過量的鉛-210沉積顆粒由陸棚外緣往棚坡水域輸送之結果。陸棚淺水站則可見大部份水柱中，釙-210係過量，僅表面上層(深度小於20米)除外。