

^{226}Ra , ^{210}Pb and ^{210}Po Distributions at the Sea off Southern Taiwan: Radioactive Disequilibria and Temporal Variations

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ABSTRACT

^{210}Pb and ^{210}Po in both particulate and dissolved phases were measured at the sea off southern Taiwan, where the Kuroshio intrudes as a branch in winter and mixes with the South China Sea water and coastal waters. Although the effects of mixing and circulation, based on hydrographic data, were significant, the removal rates of these two nuclides due to scavenging by sinking particles were evaluated.

Dissolved and total ^{210}Pb and ^{210}Po in surface water increased but particulate ^{210}Pb decreased away from the coast in September, 1994, as well as in January, 1995. On the other hand, both particulate and dissolved ^{210}Po in surface water varied temporally and were higher in January, 1995. The mean residence time calculated for the surface water ^{210}Pb with respect to particulate scavenging was generally longer in January, 1995 (0.57-0.71 yr) than in September, 1994 (0.25-0.41 yr) if a constant atmospheric flux of 2 dpm/cm²/y in the area was assumed. Similarly, for ^{210}Po it was also longer in January, 1995 (0.56-7.7 yr) than in September, 1994 (0.23-2.0 yr).

The ^{210}Pb and ^{210}Po profiles measured from three stations in different periods showed that their concentrations varied temporally and spatially, therefore probably reflecting changes in hydrography and particulate scavenging. In the upper layer where ^{210}Pb was in excess of ^{226}Ra , the mean residence time for ^{210}Pb with respect to the removal by sinking particulates was about 1.3 ~ 3 yrs. In the lower layer where ^{210}Pb was deficient relative to ^{226}Ra , the mean residence time ranged from 26 to 44 yrs resulting from variations in ^{210}Pb and ^{226}Ra inventories or varying ^{210}Pb deficits.

The mean residence time calculated for ^{210}Po in the upper, intermediate and lower layers as defined by ^{210}Po deficit or excess relative to ^{210}Pb also varied in the three sampling periods. These variations were due to the fact that both ^{210}Po and ^{210}Pb profiles varied in response to particulate scavenging and the mixing of several water masses in variable proportions at different times. ^{210}Pb was not recycled, but at two stations in January, 1995,

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^{210}Po was recycled over 100%, requiring a lateral input. This feature was also observed at the sea off northeastern Taiwan.

(Key words: ^{226}Ra , ^{210}Pb , ^{210}Po , The sea off southern Taiwan, Radioactive disequilibria, Temporal variations)

1. INTRODUCTION

Because of the particle-reactive nature of ^{210}Pb and ^{210}Po , the removal rate and mean residence time for the two nuclides can be estimated on the basis of the extent of radioactive disequilibrium between ^{210}Pb and ^{226}Ra and that between ^{210}Po and ^{210}Pb . Furthermore, since the half-lives of ^{210}Pb ($t_{1/2}=22.3$ yr) and ^{210}Po ($t_{1/2}=138.4$ d) are comparable to the time scale involved in particle dynamics in the ocean (*e.g.* transportation, dissolution and settling rate, etc.), these two nuclides are useful for studying the geochemical processes involved in oceanic particulates (*e.g.* Craig *et al.*, 1973 ; Bacon *et al.*, 1976 ; Chung and Craig, 1983 ; Chung, 1987).

The sea off southern Taiwan has long been known as an area where the mainland coastal water, the South China Sea water and the Kuroshio water meet and mix in variable proportions (Chu, 1971). It is recognized that a branch of the Kuroshio current flows into the southern Taiwan Strait via the Bashi Channel and the sea off southern Taiwan in winter (*e.g.* Wang and Chern, 1988). At the sea off southern Taiwan, the surface water is mostly composed of South China Sea water with lower salinity in summer. The salinity of the surface water increases gradually in autumn and reaches a maximum in winter and spring, as a consequence of an inflow of saline Kuroshio water (Fan, 1982; Wang and Chern, 1988).

In this study, ^{210}Pb and ^{210}Po in both the particulate and dissolved phases at the sea off southern Taiwan were measured at different periods to determine if there are temporal variations. The removal rates or mean residence times of these two nuclides were calculated on the basis of the observed extent of radioactive disequilibrium caused most likely by particulate scavenging; however, it should be kept in mind that the mixing and circulation effects are probably significant in this area. The station locations were selected in such an array that the effects of land mass and the Kuroshio water intrusion could be examined. Sea water samples were collected in separate casts for ^{226}Ra (unfiltered) and ^{210}Pb and ^{210}Po (filtered) measurements at the same stations and at the same time during several cruises.

2. SAMPLING AND ANALYTICAL METHODS

Water samples were collected at the sea off southern Taiwan in September, 1994, January and April, 1995, aboard the R/V Ocean Researcher III. The locations and water depths of the sampling stations are listed in Table 1. At Stations 1, 3 and 7, samples were collected in profile ; at other stations, only surface water samples were taken (Figure 1). No ^{226}Ra data were available in September, 1994 because the samples were lost in the processing. Niskin bottles of 20 l size were used to collect profile samples by wire casts. The sampling intervals ranged from 100 m to 300 m, with greater intervals in the deeper layers. Although the water depth at Station 3 was 3000 m, the deepest samples were collected at only 2000 m since the

Table 1. Seawater sampling locations and water depths for ^{226}Ra , ^{210}Pb and ^{210}Po measurements at the off southern Taiwan.

Station	Sampling Year and Month	Location	Depth(m)
1	94/9, 95/1	21° 55.7'N, 120° 18' E	1072
2	94/9, 95/1	21° 43.0'N, 120° 9.5' E	2120
3	94/9, 95/1	21° 29.9'N, 120° 00' E	3000
4	94/9, 95/1	21° 31.2'N, 120° 16.5'E	2400
5	94/9, 95/1	21° 32.2'N, 120° 35' E	1600
6	94/9, 95/1	21° 32.8'N, 120° 50.5'E	470
7	95/1, 95/4	21° 34.0'N, 121° 07' E	987

authors' interests were mainly above the depth of 1000 m. Samples from one cast were filtered for ^{210}Pb and ^{210}Po measurements in both the dissolved and particulate phases; those from another cast were taken for ^{226}Ra measurement unfiltered.

^{226}Ra samples were each treated with 1 ml of 1 M HgCl_2 solution to prevent any growth of organisms. The method used for ^{226}Ra measurement essentially followed that of Chung (1971). Each sample was transferred into a heavy-walled plastic bottle, and ^{222}Rn was stripped with He as a carrier gas. About a month later, the regenerated ^{222}Rn was stripped again and purified for an alpha scintillation counting. Activities of ^{222}Rn and its two alpha-emitting daughters, ^{218}Po and ^{214}Po , were used to calculate ^{226}Ra activity.

The technique for ^{210}Pb and ^{210}Po measurements was basically that of Flynn (1968). Each sample was filtered (0.45 μm pore size, 142 mm diameter) so that the particulate and dissolved phases of ^{210}Po and ^{210}Pb could be measured. The filters with particulates were digested in nitric acid and spiked with a known amount of ^{208}Po as well as a stable Pb carrier at a pH of about 1.5-2.0. Po isotopes in the sample solution were autoplated onto a silver disk, and their activities were determined by alpha spectrometry. Any remaining Po in the solution was removed by an ion exchange column. After at least five to six months, another ^{208}Po spike was added into the solution, and the regenerated ^{210}Po was then again plated together with ^{208}Po . The sample solution was then analyzed by AA to determine the chemical yield for Pb. The ^{210}Pb activity was calculated from the regenerated ^{210}Po activity with an appropriate growth factor and the Pb chemical yield. The filtered water sample was acidified by HCl to a pH of about 2. FeCl_3 solution was added and the sample was spiked with a known ^{208}Po as well as a stable Pb carrier. Pb and Po were then coprecipitated with $\text{Fe}(\text{OH})_3$ by the addition of NH_4OH for subsequent analyses, including the separation of Fe from the sample solution and autoplating for alpha spectrometry.

3. RESULTS AND DISCUSSION

3.1. Surface Water

Surface water ^{210}Pb and ^{210}Po activities in both the dissolved and particulate phases as well as ^{226}Ra activities at the sea off southern Taiwan are listed in Table 2. The errors quoted are one sigma counting statistics propagated. ^{226}Ra activities in the surface water are around 7 to 11 dpm/100kg with no systematic trends (Figure 1). These values are higher than those commonly observed in an open ocean (6-8 dpm/100kg, *e.g.* Chung and Craig, 1980). Terrigenous input was probably responsible for higher ^{226}Ra in this marginal sea.

Particulate ^{210}Pb decreases while dissolved ^{210}Pb increases away from the coast along a NE-SW line (Figure 2), suggesting that the suspended particulate distribution in the surface water probably contributed to this. The suspended particulates indeed decreased away from

Table 2. ^{226}Ra , ^{210}Pb and ^{210}Po activities in the surface water off southern Taiwan.

Station	^{226}Ra (dpm/100kg)	D ^{210}Pb (dpm/100kg)	P ^{210}Pb (dpm/100kg)	D ^{210}Po (dpm/100kg)	P ^{210}Po (dpm/100kg)	$^{210}\text{Po}/^{210}\text{Pb}$
94/9						
1	-	9.3 ± 0.5	2.8 ± 0.2	3.1 ± 0.3	1.7 ± 0.2	0.40
2	-	10.3 ± 0.4	1.3 ± 0.1	-	1.9 ± 0.2	-
3	-	10.4 ± 0.5	0.8 ± 0.1	6.0 ± 0.5	0.6 ± 0.1	0.59
4	-	14.8 ± 1.3	1.0 ± 0.1	3.1 ± 0.3	1.5 ± 0.2	0.29
5	-	13.0 ± 0.7	0.7 ± 0.1	4.3 ± 0.4	0.9 ± 0.1	0.38
6	-	9.3 ± 0.5	0.6 ± 0.1	6.4 ± 0.5	1.0 ± 0.1	0.75
7	-	12.4 ± 0.5	0.7 ± 0.1	8.5 ± 0.7	1.8 ± 0.2	0.79
95/1						
1	8.9 ± 0.3	11.0 ± 0.6	1.3 ± 0.1	2.4 ± 0.3	5.0 ± 0.2	0.60
2	8.4 ± 0.3	13.9 ± 0.7	1.0 ± 0.1	5.6 ± 0.5	2.8 ± 0.2	0.56
3	8.3 ± 0.3	13.8 ± 0.7	0.9 ± 0.1	6.5 ± 0.5	2.1 ± 0.2	0.59
4	9.5 ± 0.3	12.8 ± 0.6	0.6 ± 0.1	7.8 ± 0.6	1.9 ± 0.2	0.72
5	10.6 ± 0.3	12.4 ± 0.9	1.1 ± 0.1	8.6 ± 0.7	4.0 ± 0.2	0.93
6	7.4 ± 0.2	14.7 ± 1.0	0.6 ± 0.1	5.2 ± 0.4	2.5 ± 0.2	0.50
7	8.1 ± 0.3	12.1 ± 0.6	0.5 ± 0.1	8.6 ± 0.7	1.8 ± 0.1	0.83
95/4						
7	9.5 ± 0.3	15.1 ± 0.9	0.8 ± 0.1	1.5 ± 0.2	3.4 ± 0.2	0.31

D: dissolved phase
P: particulate phase
- : lost in processing

the coast (Stations 1, 2 and 3 respectively, 0.95, 0.41, 0.32 mg/kg in September, 1994 and 0.93, 0.61, 0.56 mg/kg in January, 1995). A similar feature was observed before at the marginal sea south of New England by Bacon *et al.* (1988). The dissolved ^{210}Pb activities are higher in January, 1995 than in September, 1994 along this line. However, no systematic trends can be recognized along the E-W line (Figure 2).

Along the NE-SW line, ^{210}Po shows a distribution pattern similar to that of ^{210}Pb , probably also regulated by the distribution of suspended particulates, which in turn was then affected by mixing and circulation (Figure 3). Particulate and dissolved ^{210}Po activities are generally higher in January, 1995 than in September, 1994 for both the NE-SW and E-W lines (Figure 3).

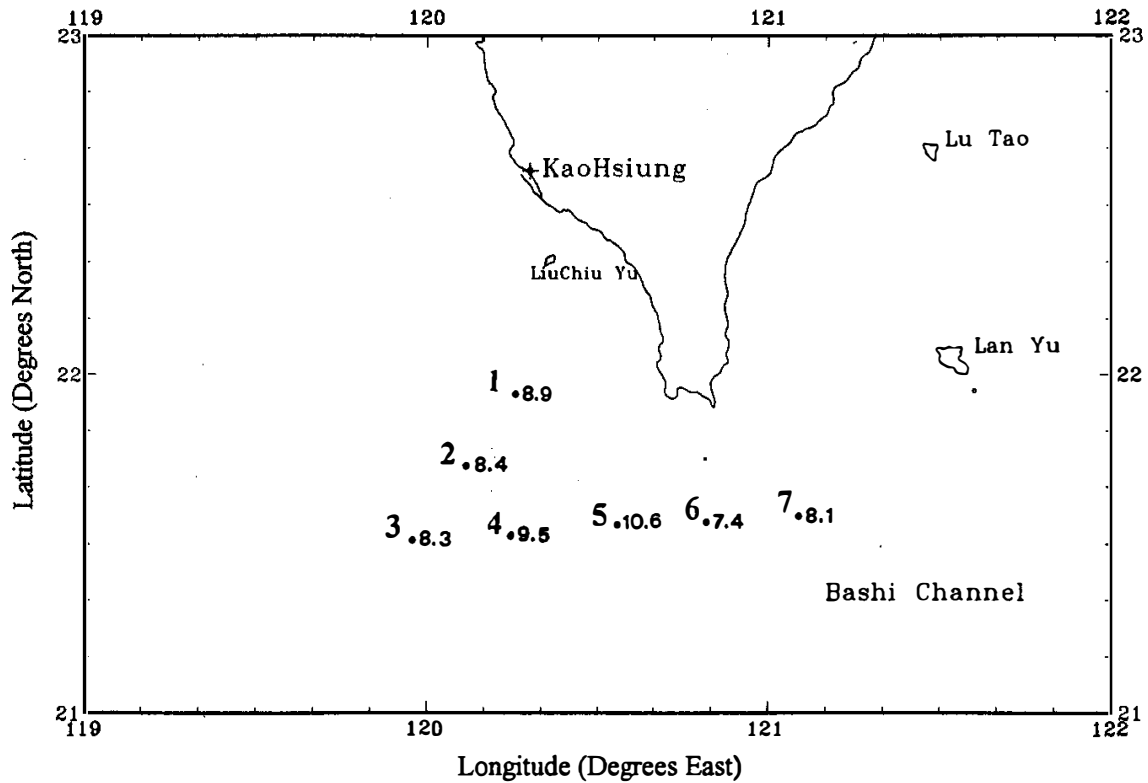


Fig. 1. Sampling locations and ^{226}Ra activities in the surface water in January, 1995.

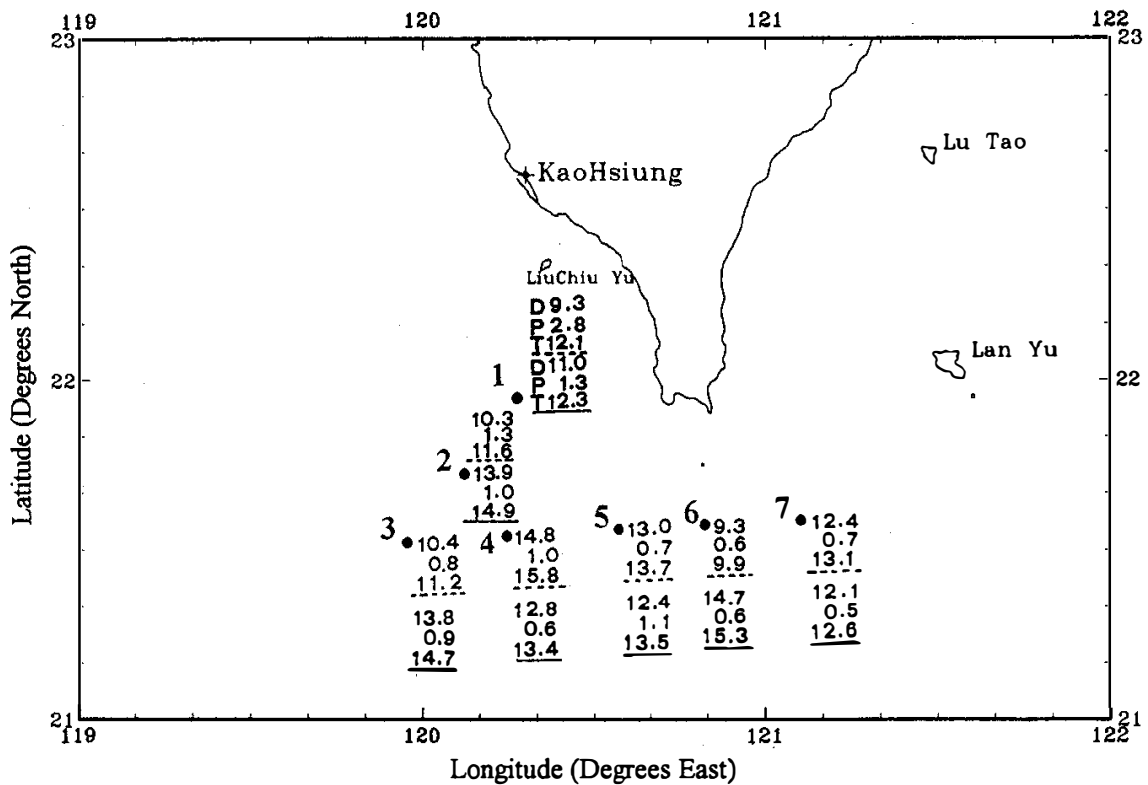


Fig. 2. Distributions of dissolved, particulate and total ^{210}Pb in the surface water in September, 1994 (underlined with a dashed line) and January, 1995 (underlined with a solid line).

Temporal variations are obvious but more pronounced for ^{210}Po than for ^{210}Pb , probably owing to the much shorter half-life of ^{210}Po .

^{210}Pb exceeds ^{226}Ra with the $^{210}\text{Pb}/^{226}\text{Ra}$ activity ratio greater than unity (1.27-2.07) in surface water because of the atmospheric ^{210}Pb flux (Poet *et al.*, 1972). $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios range from 0.29 to 0.79 in September, 1994 and from 0.50 to 0.93 in January, 1995 (Table. 2), indicating spatial and temporal variations of ^{210}Po deficiency relative to ^{210}Pb . Likewise, an unusually large ^{210}Po deficiency in the Kuroshio water was previously observed by Nozaki *et al.* (1990).

The ^{210}Po deficiency with a variable $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio probably resulted from : (1) the temporal variability of the atmospheric ^{210}Pb flux (Yang and Lin, 1992); (2) the changes in suspended matter composition and concentration; and (3) the circulation and mixing of several water masses in variable proportions. Higher $^{210}\text{Po}/^{210}\text{Pb}$ ratios observed in January, 1995 were due to much higher ^{210}Po in the particulate phase during that time (Table 2). The atmospheric ^{210}Pb flux, as determined in southern Taiwan (unpublished data), has shown seasonal variations but with values generally less than that adopted in this study. Although the total suspended matter is known to be variable, its nature and composition were not determined in this study. From September to January of the following year, the Kuroshio water was intruding into the study area, and it generated a progressive mixing with the pre-existing water masses. Thus, it is concluded that the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio was also affected by this mixing process.

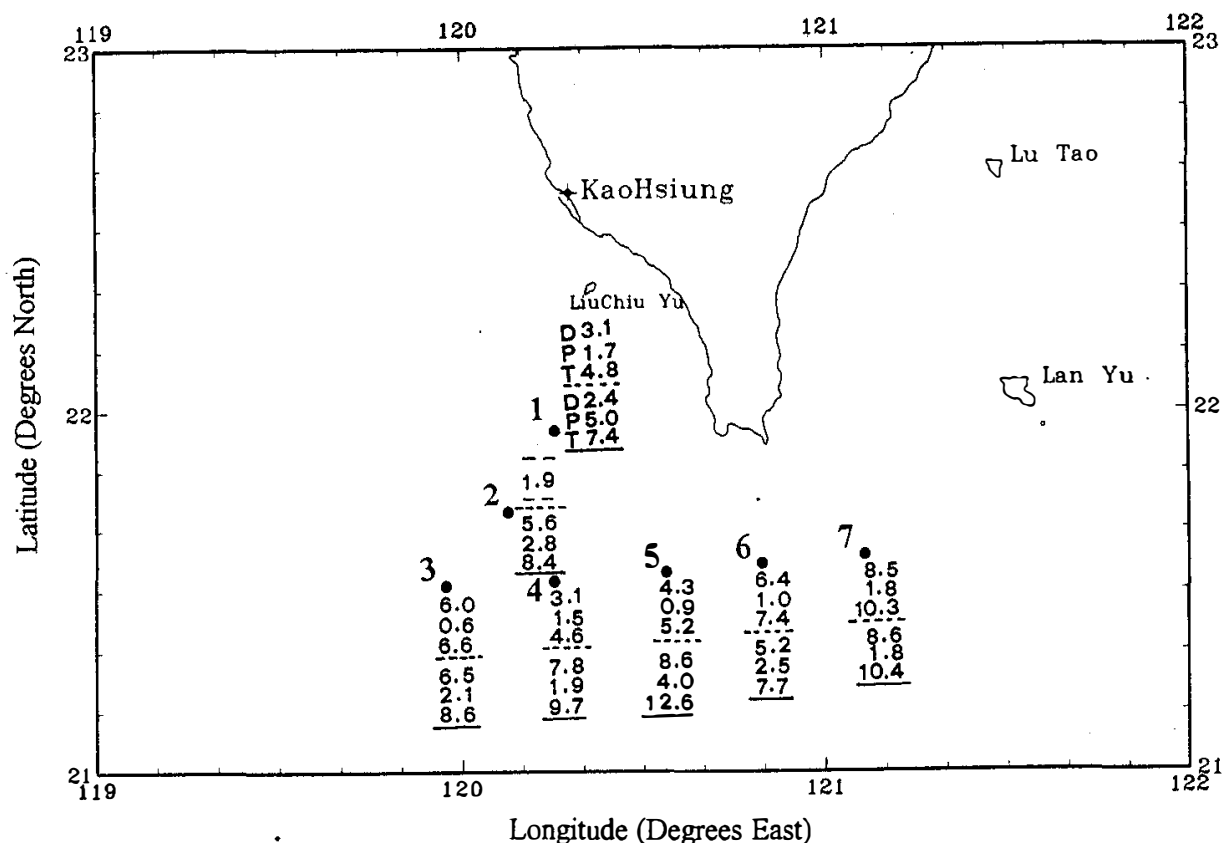


Fig. 3. Distributions of dissolved, particulate and total ^{210}Po in the surface water in September, 1994 (underlined with a dashed line) and January, 1995 (underlined with a solid line)

Calculations of the mean residence times for ^{210}Pb and ^{210}Po in the surface layer followed those of Nozaki and Tsungai (1976) and Bacon *et al.* (1976). The thickness of the surface mixed layer was 50 m in September, 1994, 90 m in January, and 10 m in April, 1995, based on the CTD data. The mean residence times calculated for ^{210}Pb and ^{210}Po in the dissolved and particulate phases as well as in the total, using the surface mixed layer thickness referred to above as reservoirs are listed in Table 3. The mean residence times for total ^{210}Pb and ^{210}Po are longer in January, 1995 than in September, 1994 because the extent of radioactive disequilibrium is smaller and the mixed layer is thicker in January, 1995 than in September, 1994. In September, 1994, the mean residence time of ^{210}Pb in the surface mixed layer is about 0.3yr or slightly over one season, probably representing a similar case to the one in summer. In January, 1995, the ^{210}Pb mean residence time increases to about 0.6yr mainly because the thickness of the mixed layer is 90 m, almost twice that in September, 1994. The thicker winter mixed layer provides a greater reservoir for the nuclide and, hence, a longer mean residence time. In April, 1995 when the mixed layer is only 10 m, the ^{210}Pb mean residence time in this layer is

Table 3. Mean residence time calculated for ^{210}Pb and ^{210}Po in the surface water off southern Taiwan.

Station	$\tau_{\text{TPb}}(\text{y})$	$\tau_{\text{DPb}}(\text{y})$	$\tau_{\text{PPb}}(\text{y})$	$\tau_{\text{TPo}}(\text{y})$	$\tau_{\text{DPo}}(\text{y})$	$\tau_{\text{PPo}}(\text{y})$
94/9						
1	0.31	0.24	0.072	0.36	0.27	0.13
2	0.30	0.26	0.033	--	--	--
3	0.29	0.27	0.020	0.79	0.75	0.071
4	0.41	0.38	0.026	0.23	0.15	0.073
5	0.35	0.33	0.018	0.34	0.27	0.058
6	0.25	0.24	0.015	1.62	1.21	0.22
7	0.34	0.32	0.018	2.02	1.19	0.35
95/1						
1	0.57	0.51	0.060	0.83	0.15	0.56
2	0.69	0.65	0.047	0.71	0.37	0.24
3	0.68	0.64	0.042	0.78	0.49	0.19
4	0.62	0.59	0.028	1.44	0.85	0.28
5	0.63	0.57	0.051	7.70	1.24	2.43
6	0.71	0.69	0.028	0.56	0.30	0.18
7	0.58	0.56	0.023	2.60	1.34	0.45
95/4						
7	0.08	0.08	0.020	0.25	0.06	0.17

Atmospheric ^{210}Pb flux taken to be 2 dpm/cm²/yr (Turekian et al., 1977)

τ_{TPb} : mean residence time for total ^{210}Pb

τ_{DPb} : mean residence time for dissolved ^{210}Pb

τ_{PPb} : mean residence time for particulate ^{210}Pb

τ_{TPo} : mean residence time for total ^{210}Po

τ_{DPo} : mean residence time for dissolved ^{210}Po

τ_{PPo} : mean residence time for particulate ^{210}Po

about 0.1 yr. The particulate ^{210}Pb mean residence time is much less than a month in each case, suggesting rapid ^{210}Pb removal by particulates with irreversible scavenging. The ^{210}Po mean residence time in the surface mixed layer varies widely between 0.2 and 8 yrs, but most values

Table 4. ^{226}Ra , ^{210}Pb and ^{210}Po measured in profile from Stations 1, 3 and 7 at 3 different periods.

Nominal Depth (m)	^{226}Ra dpm/100kg	D ^{210}Pb dpm/100kg	P ^{210}Pb dpm/100kg	D ^{210}Po dpm/100kg	P ^{210}Po dpm/100kg
Station 1 (21 ° 55.7'N, 120 ° 18'E) depth: 1072m					
September, 1994					
0	-	9.3±0.5	2.8±0.2	3.1±0.3	1.7±0.2
100	-	10.3±0.5	1.7±0.1	8.6±0.8	4.1±0.3
300	-	13.4±0.8	1.0±0.1	4.2±0.4	4.4±0.3
500	-	13.1±0.8	0.4±0.1	10.5±0.9	2.1±0.1
750	-	10.2±0.5	0.4±0.1	8.5±0.7	1.2±0.1
1000	-	11.3±0.8	0.4±0.1	6.2±0.5	3.2±0.2
January, 1995					
0	8.9±0.3	11.0±0.6	1.3±0.1	2.4±0.3	5.0±0.2
100	10.4±0.4	12.0±0.9	0.7±0.1	9.6±0.8	1.5±0.1
300	9.6±0.3	11.2±1.0	0.4±0.1	12.2±1.1	1.1±0.1
500	14.1±0.4	6.7±0.4	0.5±0.1	8.5±0.7	1.3±0.1
750	15.8±0.4	7.0±0.4	0.8±0.1	6.1±0.5	1.5±0.1
1000	21.3±0.5	9.6±0.8	0.7±0.1	5.3±0.4	1.7±0.2
Station 3 (21 ° 29.9'N, 120 ° 0'E) depth: 3000m					
September, 1994					
0	-	10.4±0.5	0.8±0.1	6.0±0.5	0.6±0.1
100	-	13.1±0.8	0.5±0.1	1.9±0.3	0.7±0.1
250	-	11.4±0.6	0.6±0.1	2.6±0.3	6.4±0.3
500	-	6.8±0.5	0.3±0.1	4.0±0.3	1.7±0.1
800	-	9.7±0.7	0.6±0.1	3.3±0.3	1.9±0.1
1100	-	7.5±0.6	0.4±0.1	2.1±0.1	1.5±0.1
1400	-	6.4±0.5	0.4±0.1	2.1±0.1	2.4±0.1
1700	-	7.9±0.5	0.3±0.1	1.2±0.1	1.0±0.1
2000	-	8.8±0.6	0.7±0.1	-	1.4±0.1
January, 1995					
0	8.3±0.3	13.8±0.7	0.9±0.1	6.5±0.5	2.1±0.2
100	8.7±0.3	15.3±0.8	0.7±0.1	7.2±0.6	1.1±0.1
250	15.0±0.5	15.5±0.8	0.7±0.1	7.3±0.6	1.0±0.1
500	15.5±0.4	10.6±0.6	0.6±0.1	10.0±1.0	1.3±0.1
800	16.3±0.4	6.6±0.4	0.7±0.1	6.9±0.6	1.7±0.2
1100	17.5±0.4	8.3±0.5	0.6±0.1	10.0±0.9	1.6±0.2
1400	19.2±0.4	5.1±0.4	0.5±0.1	-	0.8±0.1
1700	19.7±0.4	8.6±0.7	0.5±0.1	6.8±0.5	1.5±0.1
2000	-	13.0±0.9	0.8±0.1	-	2.3±0.2

Table 4. cont'd

Nominal Depth (m)	²²⁶ Ra dpm/100kg	D ²¹⁰ Pb dpm/100kg	P ²¹⁰ Pb dpm/100kg	D ²¹⁰ Po dpm/100kg	P ²¹⁰ Po dpm/100kg
Station 7 (21 ° 34'N, 121 ° 7'E) depth: 987m					
January, 1995					
0	8.1±0.3	12.1±0.6	0.5±0.1	8.6±0.7	1.8±0.1
100	9.8±0.2	11.9±0.6	-	11.3±0.9	-
250	9.3±0.2	16.3±0.9	0.7±0.1	18.0±1.2	1.0±0.1
400	15.3±0.3	10.2±0.8	0.8±0.1	10.0±0.9	1.6±0.1
700	17.1±0.3	9.5±0.8	0.7±0.1	7.9±0.7	1.5±0.2
900	18.6±0.5	8.7±0.5	1.0±0.1	-	1.2±0.2
April, 1995					
0	9.5±0.3	15.1±0.9	0.8±0.1	1.5±0.2	3.4±0.2
100	9.7±0.3	13.5±1.0	0.6±0.1	11.2±0.9	1.8±0.1
250	11.7±0.3	12.9±0.9	0.7±0.1	13.3±1.0	1.9±0.1
400	17.1±0.4	11.5±0.8	0.8±0.1	8.8±0.7	1.5±0.1
700	17.8±0.4	9.0±0.6	0.7±0.1	6.5±0.6	1.8±0.1
900	26.1±0.5	6.8±0.5	0.7±0.1	2.8±0.2	2.2±0.2

D: dissolved phase

P: particulate phase

-: lost in processing

are less than a year. Besides the effect of a thicker mixed layer, the longer mean residence time may suggest ²¹⁰Po recycling within the mixed layer.

3.2. Water Column

²²⁶Ra, ²¹⁰Pb and ²¹⁰Po in profiles were measured from three stations (1, 3, 7) during the three sampling periods. The data are given in Table 4 and are plotted in Figure 4. The errors quoted are again one sigma counting statistics propagated, as in Table 2. As no ²²⁶Ra data are available for September, 1994, those of January, 1995 are plotted and used to evaluate the ²¹⁰Pb deficiency in September, 1994. Each ²²⁶Ra profile shows a downward increase from less than 10 dpm/100kg at the surface to about 20 dpm/100kg or more near the bottom, indicating the underlying sediments as the main source.

Dissolved ²¹⁰Pb profiles are more variable but generally show a subsurface maximum and may increase toward the bottom (Figure 4). The subsurface maximum probably indicates a lateral transport. The increase toward the bottom at Stations 1 and 3 contradicts the common trend. As ²¹⁰Pb is unlikely to be released from particulates or resuspended bottom sediments, this feature is not yet fully understood. It might also represent a lateral transport or a higher production rate from ²²⁶Ra. A deviation from this pattern is the Station 7 profile of April, 1995 which shows a decrease from the maximum at the surface toward the bottom. The particulate ²¹⁰Pb at Station 1 in September, 1994 decreases downward from the surface at 2.8 dpm/100kg to a near constant at 0.4 dpm/100kg below 500 m (Table 4, Figure 4). All the other particulate

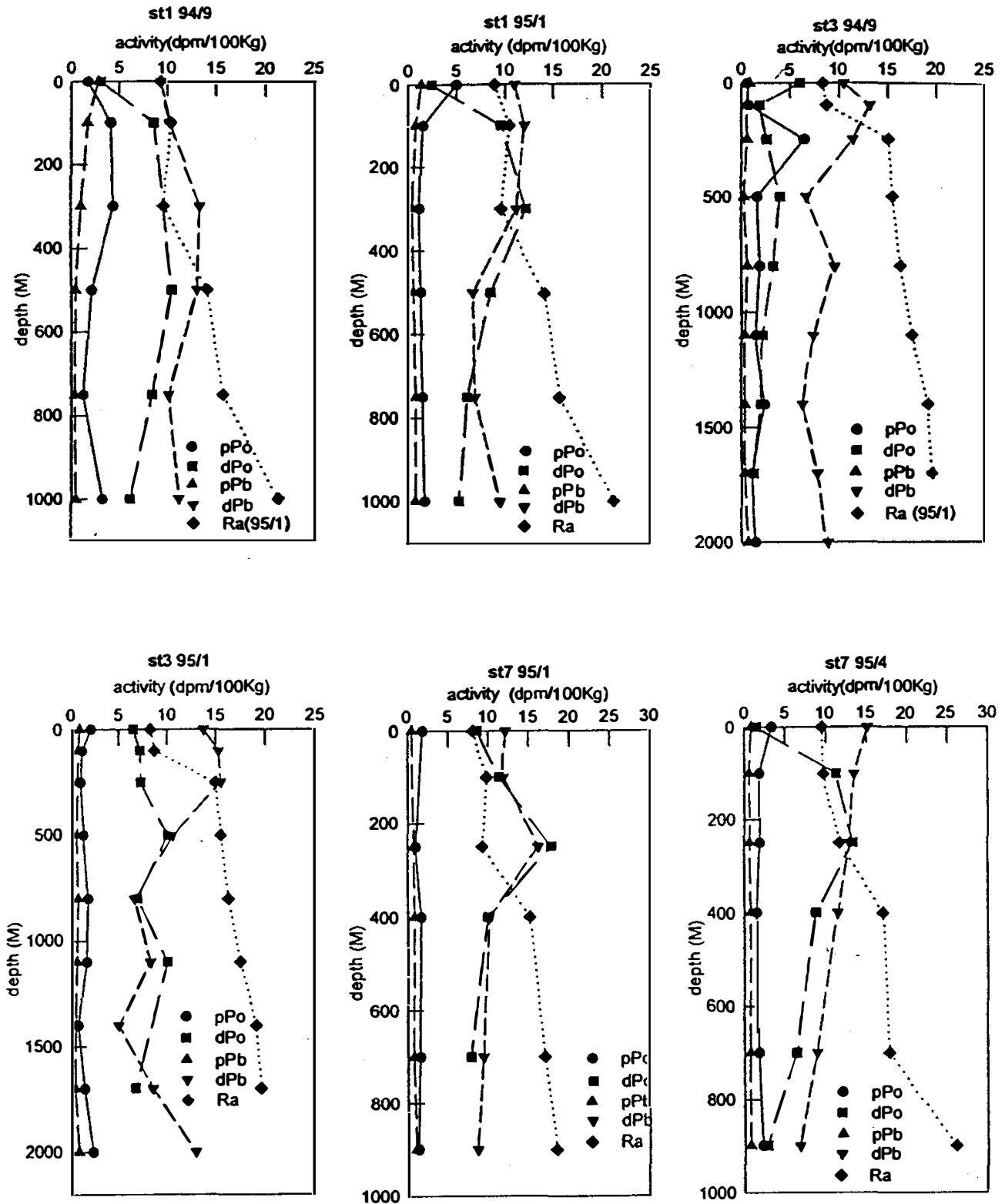


Fig. 4. Profiles of ^{226}Ra , ^{210}Pb and ^{210}Po in dissolved and particulate phases at Stations 1, 3 and 7 at three sampling periods.

^{210}Pb profiles show a fairly constant distribution at about 0.7 dpm/100kg (Table 4, Fig. 4).

Each dissolved ^{210}Po profile shows a subsurface or mid-depth maximum but at different depths and with different values. Except for the surface value, the dissolved ^{210}Po profile of Station 3 in September, 1994 has values less than half of those of the other profiles (Figure 4).

The ^{210}Po maximum is generally attributed to the dissolution of sinking organic particles and/or lateral transport. The particulate ^{210}Po profiles at Stations 1 and 3 in September, 1994, also show a subsurface maximum and much higher activities (Figure 4) due to higher particulate concentrations. The other particulate ^{210}Po profiles show a fairly uniform distribution at about 1-2 dpm/100kg below the surface water.

The $^{210}\text{Pb}/^{226}\text{Ra}$ and $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios were used to indicate the extent of radioactive disequilibria between these daughter-parent pairs and to discuss the behavior of the daughter nuclides in a water column. Figures 5 and 6 show variations of the $^{210}\text{Pb}/^{226}\text{Ra}$ and $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios with depth, respectively. As stated earlier, the January, 1995 ^{226}Ra data were used for calculating the $^{210}\text{Pb}/^{226}\text{Ra}$ activity ratio in September, 1994. The $^{210}\text{Pb}/^{226}\text{Ra}$ activity ratio is greater than unity from the surface to a depth between 200m and 500m (Figure 5), indicating the penetration depth of the atmospheric ^{210}Pb flux. The ratio is less than unity

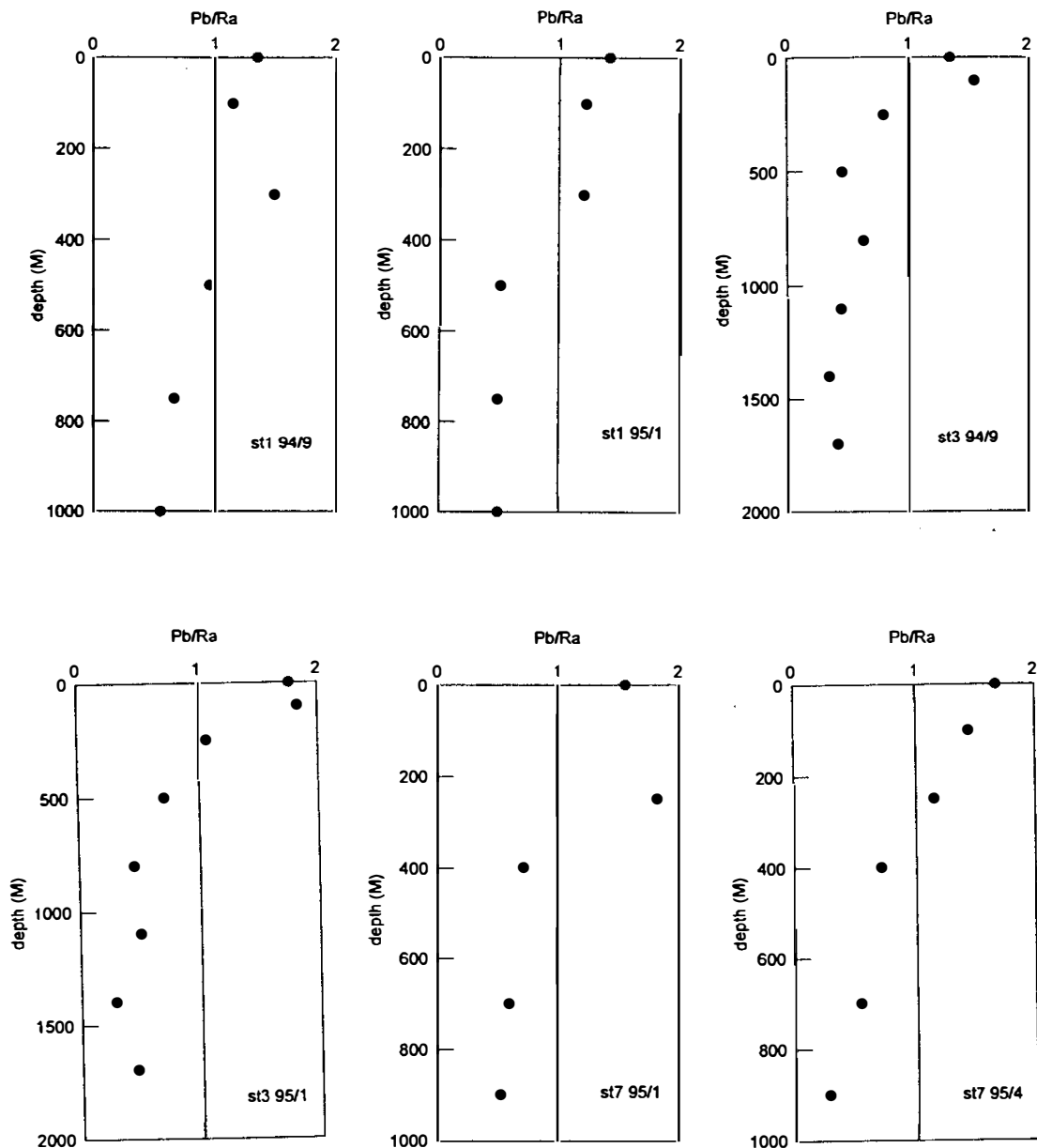


Fig. 5. Variations of $^{210}\text{Pb}/^{226}\text{Ra}$ activity ratios with depth at Stations 1, 3 and 7 at three sampling periods.

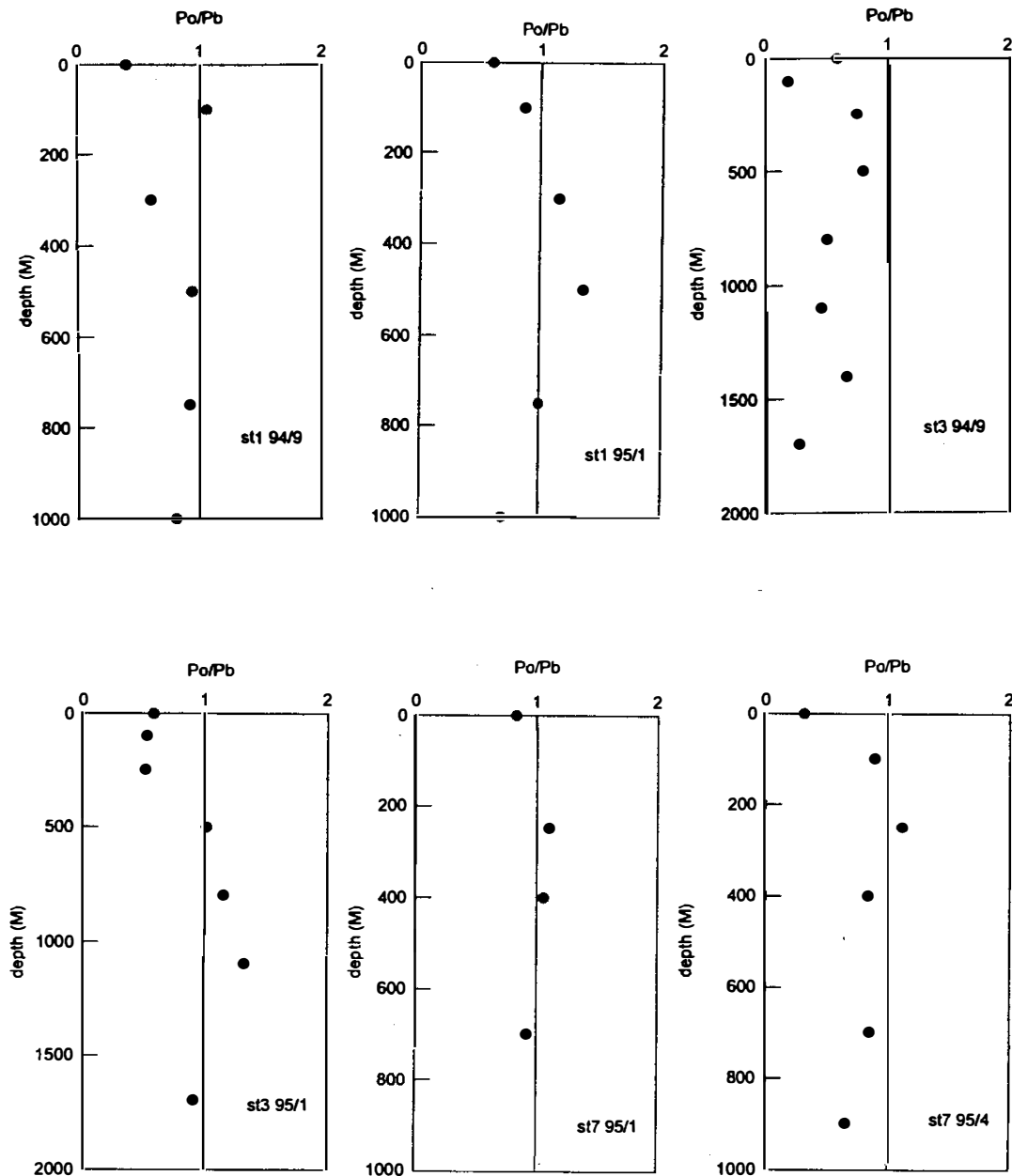


Fig. 6. Variations of $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios with depth at Stations 1, 3, and 7 at three sampling periods

below the penetration depth and indicates that the ^{210}Pb deficit is due to removal by particulate scavenging. These features have commonly been observed in open oceans (*e.g.* Chung and Craig, 1983 ; Chung, 1987).

The $^{210}\text{Po}/^{210}\text{Pb}$ activity ratios for Stations 1 and 3 in January, 1995 show a surface ^{210}Po deficiency (ratio < 1), a mid-depth excess (ratio > 1) and a deficiency again below (Figure 6). The remaining ratios show little or no mid-depth excess. In September, 1994, ^{210}Po is deficient in the entire water column of Station 3. The associated particulate matter concentrations are much higher, at about 0.8mg/kg (data not shown here). Similarly, a ^{210}Po deficit was also previously observed in the East China Sea and northwest Pacific (Nozaki *et al.*, 1991; Nozaki and Tsunogai, 1976). ^{210}Po is higher than ^{210}Pb in the particulate phase of the entire water column at each station, suggesting that the suspended particles tend to scavenge ^{210}Po prefer-

Table 5. Mean residence time of ²¹⁰Pb and ²¹⁰Po in years and ²¹⁰Po recycling efficiency in the intermediate layer at Stations 1, 3, and 7 at three sampling periods. Layer thickness is in meters.

Station	Layer	Thickness	τ_{DPb}	τ_{PPb}	Layer	Thickness	τ_{DPo}	τ_{PPo}	E(%)
1 94/9	U	500	3.0	0.4					
	L	500	44.2	1.7		1000	0.9	0.6	-
1 95/1	U	400	2.5	0.2	U	275	0.6	0.6	116%
					I	425	2.6	0.2	
	L	600	27.5	2.6	L	300	1.2	0.5	
3 94/9	U	220	1.3	0.1					
	L	1480	29.8	1.8		2000	0.3	0.2	-
3 95/1	U	300	2.3	0.1	U	700	0.7	0.1	43%
					I	900	2.6	0.3	
	L	1400	26.1	2.1	L	400	0.9	0.4	
7 95/1	U	350	2.5	0.1	U	150	2.7	1.3	166%
					I	250	10.2	0.6	
	L	550	40.6	4.0	L	300	2.7	1.0	
7 95/4	U	300	2.2	0.1	U	250	0.9	0.4	-
	L	600	26.0	2.2	L	650	1.1	0.5	

Atmospheric ²¹⁰Pb flux taken to be 2 dpm/cm²/yr (Turekian et al., 1977)

τ_{DPb} : mean residence for dissolved ²¹⁰Pb

τ_{PPb} : mean residence for particulate ²¹⁰Pb

τ_{DPo} : mean residence for dissolved ²¹⁰Po

τ_{PPo} : mean residence for particulate ²¹⁰Po

E(%) : recycling efficiency for ²¹⁰Po

U : upper layer

L : lower layer

I : intermediate layer

entially over ²¹⁰Pb in this area. Since Po is more reactive than Pb to organic particulates, a significant portion of the suspended particles may be of an organic nature.

3.3. Mean Residence Time

Based on the radioactive disequilibria observed between ²¹⁰Pb and ²²⁶Ra, as well as between ²¹⁰Po and ²¹⁰Pb, each of these water columns may be divided into two and three layers,

respectively. The box model proposed by Bacon *et al.* (1976, 1980) and Wei and Murrery (1994) could well represent an effective means to estimate the mean residence time and recycling efficiency for ^{210}Pb and ^{210}Po in each layer without having to take into consideration the hydrographic layering which may be different and variable. The atmospheric ^{210}Pb flux of 2 dpm/cm²/yr (Turekian *et al.*, 1977) was used in the calculations although lower and variable values have earlier been observed (unpublished data). The effect of a lower ^{210}Pb flux is an increase in the ^{210}Pb mean residence time in the upper layer where ^{210}Pb is in excess. The mean residence times of ^{210}Po and ^{210}Pb in each defined layer and ^{210}Po recycling efficiency for the intermediate layer where ^{210}Po is in excess were calculated for Stations 1, 3 and 7 for the three sampling periods and are listed in Table 5. These results should be interpreted in a relative manner since these nuclide profiles show seasonal variabilities and are at best in a quasi-steady state.

The mean residence times of dissolved and particulate ^{210}Pb in the upper layer where ^{210}Pb is in excess are about 1.3-3.0 yr and 0.1-0.4 yr, respectively. These values are nearly proportional to the thickness of the upper layer, and are comparable to those reported for the open oceans (Bacon *et al.*, 1976). The mean residence times of dissolved and particulate ^{210}Pb in the lower layer where ^{210}Pb is deficient are between 26 and 44 yr and between 1.7 and 4 yr, respectively. Compared to the ^{210}Pb mean residence time determined in the deep Atlantic (~40 yr) and Pacific (~54 yr) (Craig *et al.*, 1973), the values shown in Table 5 are lower probably the result of a greater particle scavenging rate at marginal sea. No ^{210}Pb recycling occurs in the lower layer during the three sampling periods (Table 5).

The mean residence times of dissolved and particulate ^{210}Po in the upper layer are about 0.6-2.7 yr and about 0.1-1.3 yr, respectively. These large ranges reflect greater ^{210}Po variability. In the intermediate layer, they are, respectively, between 2.6 and 10.2 yr, and between 0.2 and 0.6 yr. In the lower layer, the mean residence time of dissolved ^{210}Po is about 0.9-2.7 yr, while that of particulate ^{210}Po is about 0.4-1.0 yr. The mean residence time of ^{210}Po in the intermediate layer where it is in excess of ^{210}Pb is longer than that in the upper or lower layer, probably due to recycling. The ^{210}Po recycling efficiency calculated for the intermediate layer is 116%, 43% and 166% at Stations 1, 3 and 7, respectively, in January, 1995. For values greater than 100%, an additional input from lateral transport is required. A similar feature was also observed in previous studies at the sea off northeast Taiwan (Chung and Wu, 1995).

4. CONCLUSIONS

As shown in this study, in surface water, the dissolved and total ^{210}Pb and ^{210}Po increase, whereas particulate ^{210}Pb and ^{210}Po decrease away from the coast of southern Taiwan. The particulate ^{210}Pb is higher but the particulate, dissolved and total ^{210}Po is lower in September, 1994 than in January, 1995, indicating temporal variations. The mean residence times of total ^{210}Pb and ^{210}Po in surface water are shorter in September, 1994 than in January, 1995.

The ^{210}Pb and ^{210}Po profiles at three stations vary with sampling periods, and thus, the radioactive disequilibrium between the two nuclides in a water column also varies. Mixing and circulation may have modified the hydrography and the suspended particulate matter distribution. The latter probably controls the scavenging or removal of the reactive nuclides.

Based on the radioactive disequilibria among ^{210}Po , ^{210}Pb and ^{226}Ra , the water column can be divided into layers for calculation of the mean residence times of ^{210}Po and ^{210}Pb . The calculated results for the three sampling periods were variable, probably reflecting hydrographic changes caused by mixing and circulation. The mean residence time of ^{210}Pb was less variable than that of ^{210}Po because ^{210}Po has a short mean life and is biophilic. ^{210}Pb was not recycled in the entire water column. As its activity exceeds ^{210}Pb at mid-depth, ^{210}Po was recycled in this intermediate layer at Stations 1 and 7 in January, 1995. Mass balance consideration requires an additional ^{210}Po input from lateral transport.

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