

Pb-210 and Po-210 Distributions and Their Radioactive Disequilibria in the Kuroshio Waters off Eastern and Northeastern Taiwan

Y. N. LIN¹ AND Y. CHUNG¹

(Received 20 June 1991; Revised 16 August 1991)

ABSTRACT

This paper presents the distributions of Pb-210 and Po-210 and the extent of their radioactive disequilibrium in areas off eastern and northeastern Taiwan where the Kuroshio flows northward and mixes with the East China Sea water. Four vertical profiles of total Pb-210 and Po-210 activities have been measured together with 34 surface water stations within the domain of the Kuroshio and its edge waters.

Three of the profiles along the Kuroshio track share some common features: the Pb-210 maximum occurs at a depth of between 25 m and 100 m; the Po-210 maximum occurs fairly close to the 200 m depth; both nuclides decrease with depth below 200 m and reach radioactive equilibrium below 500 m. Thus, each water column may be divided into three layers based on the activity ratio of these two nuclides. In the upper layer (0 ~ 150 m), the Po-210/Pb-210 ratio is less than unity, suggesting a preferential removal of Po-210. In the middle layer (150 ~ 500 m), the ratio is greater than unity, suggesting a recycling of Po-210 by dissolution or desorption. In the lower layer (below 500 m), the ratio is about unity, indicating a radioactive equilibrium between the two nuclides.

Using a simple box model with some reasonable assumptions for key parameters in a steady state, one calculates a mean residence time of 0.9 and 1.5 years respectively for Po-210 and Pb-210 in the upper layer. As both nuclides are highly reactive they are rapidly removed from the upper layer by particle scavenging. However, at least 40% of the Po-210 removed from the upper layer is recycled within the middle layer, but less than 5% of the Pb-210 is recycled this way. These features are common in an open ocean water column and suggest that the Kuroshio has no influence on the distribution of these nuclides within its domain. The mean residence time for Pb-210 in the lower layer at one station (12 years, H16) is only one tenth of that at another (120 years, R23), suggesting that both boundary scavenging and in-situ scavenging by particulate matter are very effective at H16.

¹ Institute of Marine Geology, National Sun Yat-sen University, Kaohsiung, Taiwan, R. O. C.

In the East China Sea shelf water, the Po-210 excess occurs about 25 m below the surface and extends to the bottom (120 m). This is quite different from the Po-210 excess observed in the Kuroshio water where the excess occurs below the 150 m depth. The Po-210 excess in the shelf water may result from intrusion and upwelling of water from the Kuroshio middle layer in which Po-210 is enriched due to recycling. This is consistent with hydrographic observations which suggest that the subsurface water in this area has been elevated about 130 m.

Both Po-210 and Pb-210 in the surface water increase in concentration from land mass or shelf zone toward the deep sea, reflecting the influence of particulate concentrations and of the boundary scavenging processes. However, the Po-210/Pb-210 activity ratio remains fairly constant at 0.5 ~ 0.6 for all surface water samples, suggesting a mean Po-210 residence time of about 0.5 to 0.8 years. This is quite consistent with a value of 0.9 years calculated for the upper layer using a box model that involves particulate scavenging under a steady-state assumption.

1. INTRODUCTION

In the natural uranium and thorium decay series, the production of a nuclide in the ocean can be determined accurately from its parent activity. Each nuclide has a definite chemical characteristics that allows its partition between seawater (dissolved phase) and suspended particles (particulate phase) with different proportions. The portion that dwells on the particulate matter, if not yet decayed, eventually sinks to the bottom as part of the marine deposits.

The activities of the paired parent-daughter nuclides within the ocean can provide information on chemical or biological processes based on the extent of their radioactive disequilibrium. The removal rates of the particle-reactive nuclides and certain trace elements from the oceanic water column can also be estimated (e.g. Nozaki and Tsunogai, 1976; Bacon *et al.*, 1976; Chung *et al.*, 1982) and the role that ocean boundaries play in the scavenging processes can be assessed (e.g. Nozaki *et al.*, 1976; Bacon *et al.*, 1976; Chung and Finkel, 1988).

The fate of certain reactive elements introduced into the oceans and the biogeochemical processes operating in there can be studied using Pb-210 ($t_{1/2} = 22.3$ years) and its granddaughter Po-210 ($t_{1/2} = 138.4$ days) because both nuclides are particle-reactive and their half-lives are comparable to the time scale of some oceanic processes. Pb-210 is produced throughout the water column by the decay of Ra-226 and its successive short-lived daughters; it is also introduced into the sea surface from decay of atmospheric Rn-222. Virtually all Po-210 in seawater is produced in-situ by the decay of Pb-210 (Bacon *et al.*, 1976). In the oceans both nuclides may be recycled or settle along with solid particles and eventually be deposited in the marine sediments. Figure 1 shows various pathways through which these nuclides may proceed in an atmospheric or marine environment.

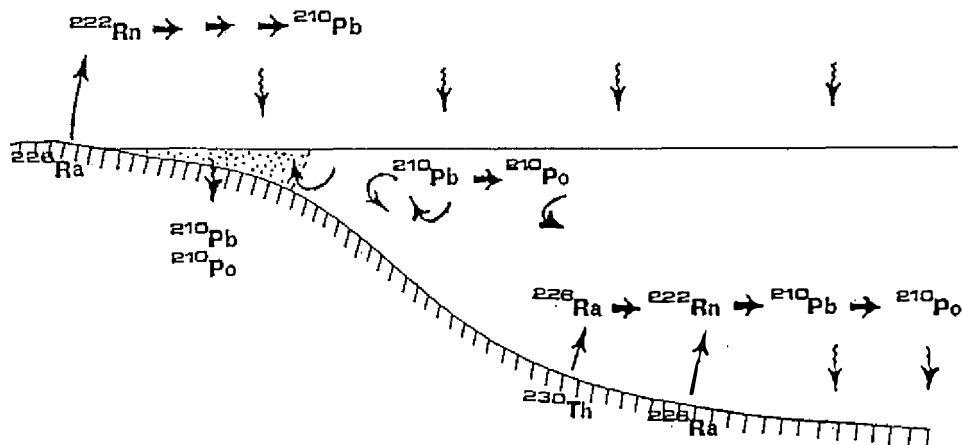


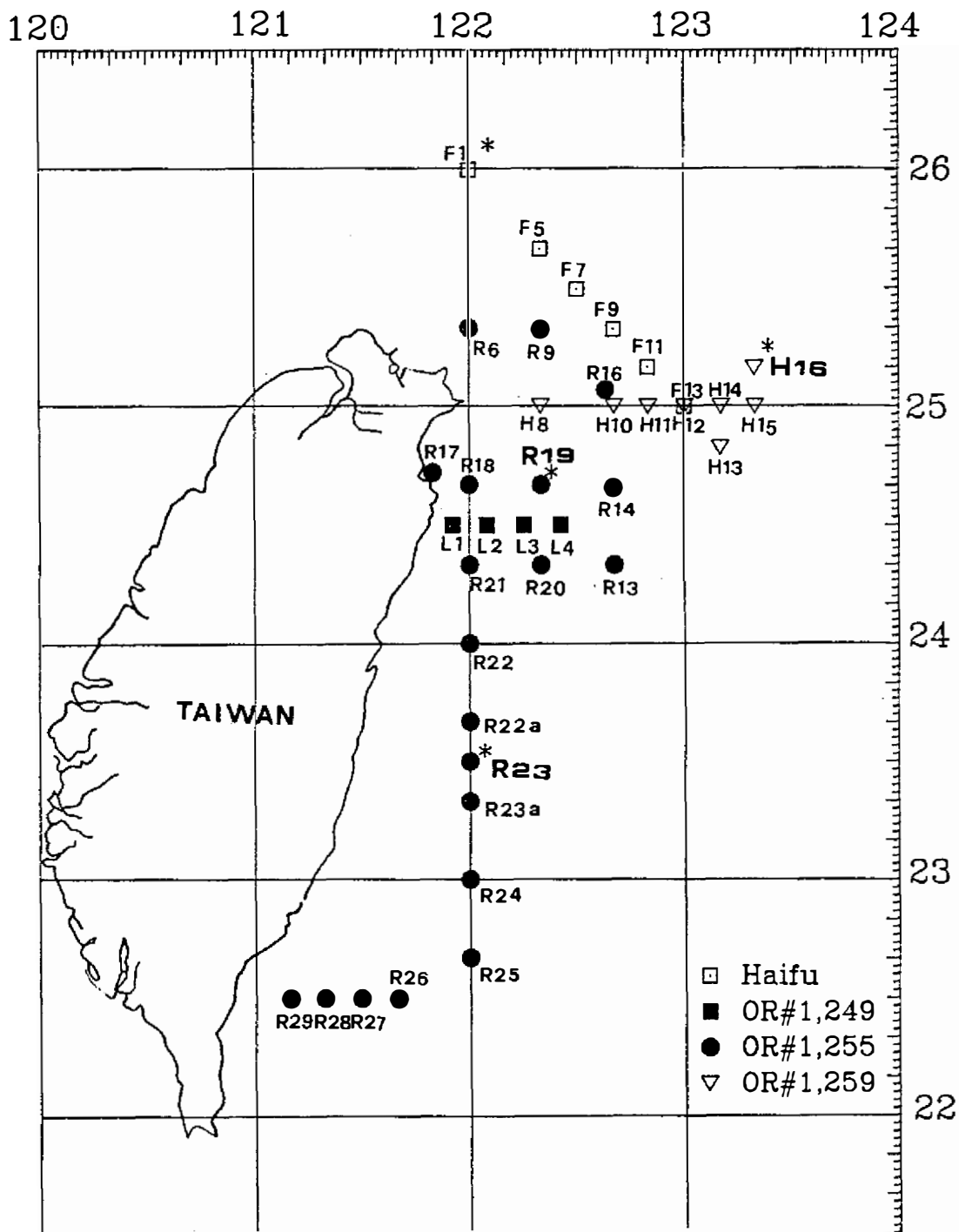
Fig. 1. Various pathways for Pb-210 and Po-210 in a marine environment.

This paper presents for the first time the distributions of Po-210 and Pb-210 and the extent of their radioactive disequilibrium in the areas off eastern and northeastern Taiwan where the Kuroshio current flows northward and mixes with the East China Sea shelf water. The purpose of this study is to examine the scavenging processes influenced, if at all, by the Kuroshio and its exchange with the surrounding waters, using Po-210/Pb-210 disequilibrium as an indicator. Four profiles and 34 surface water stations of total Pb-210 and Po-210 have been measured within the study area.

2. COLLECTION AND ANALYSIS OF SAMPLES

Water samples of 20 l size were collected in profile using Niskin bottles attached to a CTD Rosette at four different stations. Surface water samples were collected with plastic buckets. These samples were all transferred to plastic containers and immediately acidified with HCl to pH~2.

The analytical procedures we adopt have been described in detail by Chung *et al.* (1983). Pb-210 is determined by counting the α activity of the Po-210 that grows from Pb-210 during its storage, using a multi-channel analyzer (MCA, manufactured by EG and G Ortec). A Po-208 spike prepared from a standard solution of NIST was used to determine the Po-210 activity. The Po-210 of a sample was first determined by counting its α activity initially in the water sample and then with decay or growth correction once the Pb-210 activity was determined. Pb-210 was determined by counting the α activity of the Po-210 that grew from the Pb-210 over a known period. Thus, both Po-210 and Pb-210 were measured exactly from the same sample. Figure 2 shows stations where Pb-210 and Po-210 samples were collected and measured.



*:profile

Fig. 2. Sampling stations from different cruises.

3. PROFILES OF Pb-210 AND Po-210 IN THE KUROSHIO CURRENT

The results of Pb-210 and Po-210 measurements from three stations located within the Kuroshio current are listed in Table 1 and plotted in Figure 3. These profiles share some common features: the Pb-210 maximum occurs at a depth of between 25 m and 100 m, ranging from 14.9 and 21.4 dpm/100 kg; the Po-210 maximum occurs fairly close to the 200 m depth, ranging from 14.9 to 19.4 dpm/100 kg; profiles of both nuclides cross over each other around the 150 m depth, creating a point of apparent radioactive equilibrium; both nuclides decrease with depth below 200 m and reach radioactive equilibrium below 500 m.

Each water column may be divided into three layers based on the activity ratio or difference between Po-210 and Pb-210. The activity difference as a function of depth is shown in Figure 4. Within the upper layer (0 ~ 150 m) of the water column, all the data indicate a Po-210 deficiency relative to Pb-210. This is a zone where preferential removal of Po-210 from the dissolved phase takes place, probably due to scavenging by mainly organic particulate matter. In the middle layer (150 ~ 500 m), a Po-210 enrichment relative to Pb-210 is apparent, indicating a net supply of Po-210 by processes other than production from radioactive decay of Pb-210. In the lower layer (below 500 m), the activity difference is nearly zero, indicating a radioactive equilibrium between the two nuclides.

4. BOX MODEL FOR THE VERTICAL PROFILES

Material balance calculations for Ra-226, Pb-210 and Po-210 adopted by Becon *et al.* (1976) are used to evaluate the behavior of these nuclides in the area studied. Assuming the vertical mixing to be negligible and at a steady-state, the mean residence times for Pb-210 and Po-210 in the upper layer, the recycling of these nuclides in the middle layer due to their particle-reactivity and biogeochemical characteristics, and the mean residence time for Pb-210 in the lower layer can be calculated with a box model provided that the partition between the dissolved and particulate phases for these nuclides is known. The information needed but not measured in this study include the Ra-226 and particulate Pb-210 and Po-210 profiles. To overcome these difficulties we shall refer to information available in the literature.

The ratio of particulate Pb-210 to the total Pb-210 (R) is quite variable but generally increases with depth. In the surface water and upper layer it is even more variable. For our simplified model calculations, we assume 2%, 5% and 10%, respectively for the upper layer, middle layer and lower layer by referring to the data in Chung *et al.* (1983) and Somayajulu and Craig (1976).

Table 1. Pb-210 and Po-210 activities from three profiles located in Kuroshio waters (H16, R19 and R23) and one located in shelf water (F1). The errors quoted are one sigma counting errors and other errors combined.

Station	Wire depth(m)	Pb-210(dpm/100 kg)	Po-210(dpm/100 kg)
OR#1 Cruise 259 (Oct.27-Nov. 1, 1990)			
H16 (25° 10'N, 123° 20'E; 1774m):			
	0	19.4±1.0	9.3±0.9
	25	21.4±1.3	10.1±1.1
	50	17.0±1.1	10.6±1.2
	100	16.8±1.0	11.0±1.1
	200	16.4±0.9	19.4±2.1
	400	12.1±0.7	16.7±1.9
	600	11.8±0.7	12.5±1.2
	900	6.0±0.3	4.4±0.5
	1200	4.6±0.3	3.6±0.4
	1495	5.6±0.4	5.4±0.6
	1750	6.4±0.4	6.5±0.7
OR#1 Cruise 255 (Sep.25-Oct. 1, 1990)			
R19 (24° 40'N, 122° 20'E; 870m):			
	0	10.8±0.6	6.4±0.6
	25	12.6±0.7	7.6±0.8
	50	15.1±1.0	7.6±0.8
	100	16.1±0.9	10.9±1.0
	200	12.6±0.6	15.5±1.3
	300	9.4±0.5	12.2±1.3
	500	8.9±0.5	8.0±0.8
R23 (23° 30'N, 122° 00'E; 4250m):			
	0	13.5±0.8	6.1±0.6
	25	13.2±0.9	8.0±0.9
	50	14.9±0.7	7.5±0.8
	100	13.1±0.8	10.8±1.1
	200	12.8±0.8	14.9±1.3
	500	12.6±0.8	12.2±1.2
	750	12.7±0.7	12.4±1.2
	1000	12.3±0.8	13.7±1.4
Haifu (June 18-22, 1990)			
F1 (26° 00'N, 122° 00'E; 120m):			
	0	6.4±0.4	3.8±0.4
	25	6.7±0.4	8.7±0.8
	100	5.1±0.3	11.3±1.2

Note: The errors quoted in Pb-210 are about 6% while those in Po-210 are about 10% due to various corrections required for the Po-210 activities.

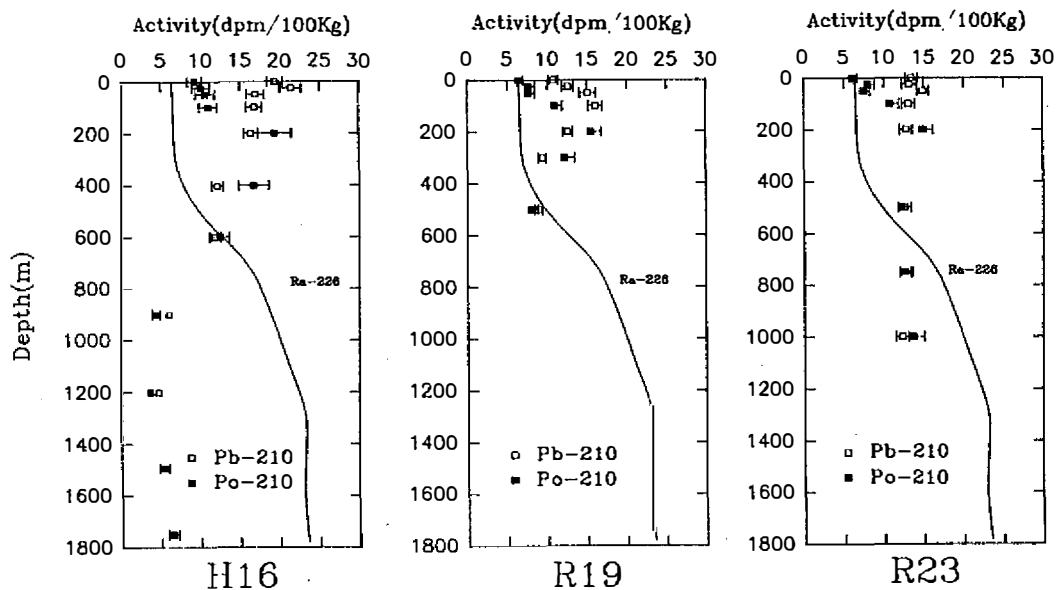


Fig. 3. ^{210}Pb and ^{210}Po profiles from three stations: H16, R19 and R23. The solid curve marked as Ra-226 was calculated by Nozaki *et al.* (1990) based on their Si data obtained near H16 and the Ra-Si relationship that they established.

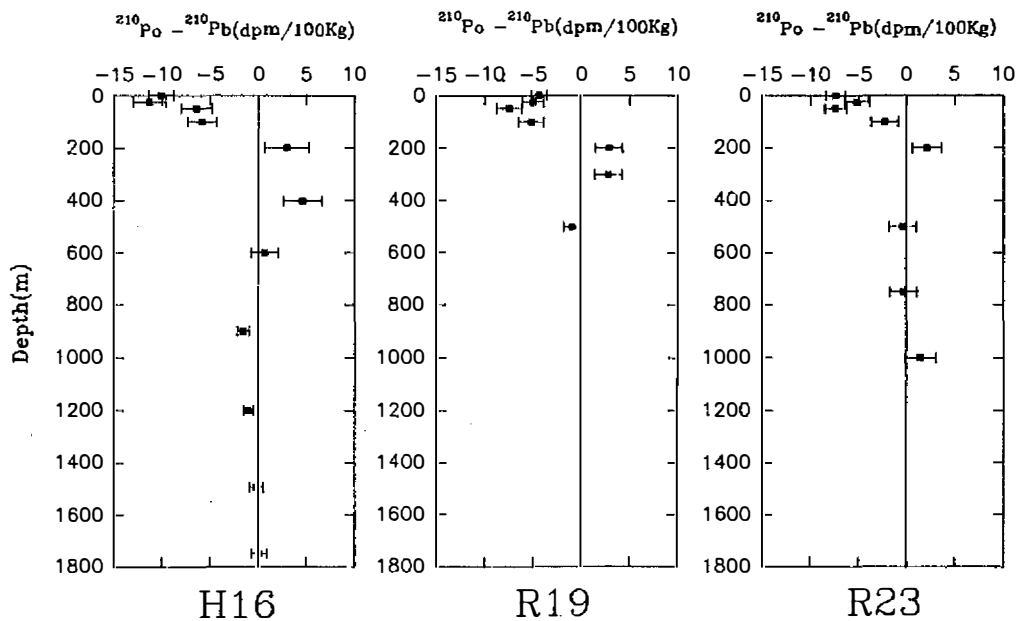


Fig. 4. ^{210}Po and ^{210}Pb differences plotted as a function of depth for the three profiles (H16, R19, R23).

The ratio of Po-210 to Pb-210 in the particulate phase (T) obtained by Nozaki *et al.* (1990) averaged about 1.05. As no Ra-226 profiles were measured at stations where Pb-210 and Po-210 profiles were measured, we estimated the Ra-226 concentration based on a linear Ra-226 and Si relationship established by Nozaki *et al.* (1990) and their Si profile measured near one of our profile stations (H16). The northwestern Pacific GEOSECS Ra-226 data of Chung and Craig (1980) and the associated Si data were used by Nozaki *et al.* (1990) to obtain:

$$[Ra - 226] = 0.062 + 0.00124 [Si]$$

where [Ra-226] is the Ra-226 activity in dpm/l and [Si] the dissolved silica concentration in $\mu mol/l$.

For discussion of the box model and its material balance equations, the following symbols or letters are defined:

λ =decay constant (yr^{-1})

d=dissolved phase

p=particulate phase

J=net rate of transfer or flux ($dpm.cm^{-2}.yr^{-1}$) from the particulate to the dissolved phase, i.e. the production term, taken to be positive, and vice versa

P=removal rate of a nuclide due to particulate matter sinking out of the layer

τ =mean residence time of a nuclide, defined as the time required to remove the nuclide from its reservoir, other than by radioactive decay

F=the rate of input or flux ($dpm.cm^{-2}.yr^{-1}$) from the atmosphere

I=inventory ($dpm.cm^{-2}$) of a nuclide within a defined layer

4.1 Upper layer

The material balance for dissolved Po-210 in the upper layer (0 ~ 150 m) can be expressed as:

$$F_{P_o} + \lambda_{P_o} I_{P_o}^d + J_{P_o} = \lambda_{P_o} I_{P_o}^d \quad (1)$$

which states that the atmospheric flux of Po-210, the production of Po-210 by the radioactive decay of Pb-210 in its dissolved phase, and the production or transfer of Po-210 from the particulate phase to the dissolved phase via dissolution or desorption are balanced by the radioactive decay of Po-210 in the dissolved phase. If Po-210 is removed from the dissolved phase and transferred into the particulate phase, then J, by definition is negative. The flux of Po-210 from the atmosphere is assumed to occur in the dissolved phase and is about 10% that of Pb-210 (Burton and Stewart, 1960; Poet *et al.*, 1972). The mean residence time for Po-210 in the dissolved phase is given by:

$$\tau_{P_o}^d = I_{P_o}^d / |J_{P_o}| \quad (2)$$

The material balance of the particulate Po-210 can be stated such that the supply from decay of particulate Pb-210 and transfer from the dissolved phase into the particulate phase (in this case, $J_{P_o} < 0$) are balanced by radioactive decay and removal by particles from the upper layer, i.e.:

$$\lambda_{P_o} I_{P_b}^p - J_{P_o} = \lambda_{P_o} I_{P_o}^p + P_{P_o} \quad (3)$$

The mean residence time for particulate Po-210 is:

$$\tau_{P_o}^p = I_{P_o}^p / P_{P_o} \quad (4)$$

If the mean residence time of particulate Pb-210 is similar to that of particulate Po-210 (Turekian *et al.*, 1974), then the removal rate of particulate Pb-210 by particles sinking out of the upper layer can be calculated as the inventory of particulate Pb-210 in the upper layer is known:

$$P_{P_b} = (I_{P_b}^p / I_{P_o}^p) P_{P_o} \quad (5)$$

If we consider the Pb-210 flux from the atmosphere, F_{P_b} , to be soluble, then the balance for dissolved Pb-210 is:

$$\lambda_{P_b} I_{R_a} + F_{P_b} + J_{P_b} = \lambda_{P_b} I_{P_b}^d \quad (6)$$

in which J_{P_b} is negative since Pb-210 is transferred from the dissolved phase into the particulate phase by particle scavenging, and the mean residence time for the dissolved Pb-210 is:

$$\tau_{P_b}^d = I_{P_b}^d / |J_{P_b}| \quad (7)$$

The balance for particulate Pb-210 can be written as:

$$-J_{P_b} = \lambda_{P_b} I_{P_b}^p + P_{P_b} \quad (8)$$

since here, J_{P_b} is negative. With the assumptions that particulate Pb-210 is 2% of the total Pb-210 activity and that the activity ratio of Po-210 to Pb-210 for the particulate phase is 1.05, one can calculate the values of F_{P_b} , J_{P_b} , P_{P_b} , J_{P_o} and P_{P_o} (five unknown parameters) using equations (1), (3), (5), (6) and (8), based on the measured total Pb-210 and Po-210 inventories or profiles. Consequently, the mean residence times for Pb-210 and Po-210 in the dissolved phase as well as in the particulate phase can be calculated with equations (2), (4) and (7). The calculated results for the upper layer from the three vertical profiles are given in Table 2. The mean residence times for dissolved Pb-210 and Po-210 at these stations show a systematic increase toward the south (R23),

Table 2. Mean residence times for dissolved Pb-210 and Po-210 and for particulate Po-210 in the upper water layer, and recycling efficiency of both nuclides in the middle layer for stations in the Kuroshio waters calculated with a box model with parameter T at 1.05 and R at 2% and 5% respectively for the upper and middle layers.

Station	F_{P_b}	Mean residence times of nuclides in the upper layer (yr)			Recycling efficiency (%) of unclides in the middle layer	
		$\tau_{P_b}^d$	$\tau_{P_o}^d$	$\tau_{P_b}^p$	$^{210}P_b$	$^{210}P_o$
H16	1.93	1.39	0.83	0.03	3.50	100
R19	1.48	1.48	0.91	0.03	2.00	61.7
R23	1.22	1.67	1.10	0.03	4.50	42.9

Note: F_{P_b} is the atmospheric Pb-210 flux in $dpm/cm^2.yr$ calculated from the box model.

with the calculated atmospheric Pb-210 flux decreasing southward from 1.9 to 1.2 $dpm/cm^2.yr$. The average mean residence times are about 1.5 and 0.9 years respectively for Pb-210 and Po-210 in the upper layer. The mean residence time for particulate Po-210 (as well as for particulate Pb-210) is quite uniform at 0.03 years or about 11 days. Thus, in a layer of 150 m, the mean particle settling rate is about 13.6 m/d or 0.016 cm/s, corresponding to a particle size of about 17 μm (diameter) if Stoke's law applies and the density difference is assumed to be 1 g/cm^3 .

4.2 Middle layer

In the middle layer (150 ~ 500 m), the material balance for dissolved Po-210 is:

$$\lambda_{P_o} I_{P_b}^d + J_{P_o} = \lambda_{P_o} I_{P_o}^d \quad (9)$$

J_{P_o} represents the Po-210 flux from the particulate phase to the dissolved phase. The ratio, J_{P_o}/P_{P_o} , represents the recycling efficiency and indicates the redissolved fraction of the particulate Po-210 flux from the upper layer.

A similar equation can be written for dissolved Pb-210 in the middle layer:

$$\lambda_{P_b} I_{R_o} + J_{P_b} = \lambda_{P_b} I_{P_b}^d \quad (10)$$

and the recycling efficiency for particulate Pb-210 returning to the dissolved phase is J_{P_b}/P_{P_b} .

Thus, equations (9) and (10) are used to calculate the J term in order to obtain the recycling efficiency of Pb-210 and Po-210 within the middle layer as given in Table 2. The recycling efficiency for Pb-210 in the middle layer is less than 4.5%, but that for Po-210 is at least 40%. At station H16 the value

is more than 100%, suggesting that there is an additional source of J_{P_b} in the middle layer.

4.3 Lower layer

As the major source of dissolved Pb-210 in the lower layer (below the 500 m depth) comes from radioactive decay of Ra-226, the Pb-210 removal rate from the dissolved phase other than by radioactive decay is given as: $[\lambda_{P_b} I_{R_a} - \lambda_{P_b} I_{P_b}^d]$

Thus, the mean residence time with respect to removal rate for dissolved Pb-210 is:

$$\tau_{P_b}^d = I_{P_b}^d / \lambda_{P_b} (I_{R_a} - I_{P_b}^d)$$

The removal rate and the mean residence time for dissolved Pb-210 in the lower layer are listed in Table 3. The lower layer is defined here as between 500 m or 600 m in depth and the depth where the deepest Pb-210 sample was measured in the profile. Thus R23 extended to only 1000 m although the station depth is over 4000 m. As the scavenging rate at H16 is more than 10 times as fast as that at R23 while the inventory for both stations is comparable, the mean residence time at H16 is one tenth of that at R23. The extremely short mean residence time of 12 years for H16 may result from bottom boundary scavenging of the resuspended particles since the water depth at this station is only 1770 m. The dissolved Pb-210 at H16 is transferred to the particulate phase at a rate 5 times as fast as that at R23.

Table 3. Scavenging rates and mean residence times for the dissolved Pb-210 in the lower layer calculated with R at 10% for stations H16 and R23.

Station	Depth (m)	Dissolved $^{210}P_b$ inventory (dpm/cm ²)	^{226}Ra inventory (dpm/cm ²)	Scavenging rate (dpm/cm ² .yr)	$\tau_{P_b}^d$ (yr)	ϕ^* (dpm/m ³ .yr)
H16	600~1750	6.57	23.83	0.54	12	4.70
R23	500~1000	5.66	7.18	0.047	120	0.94

*: ϕ is a transfer rate of Pb-210 from the dissolved phase to the particulated phase within the lower layer as defined.

5. DISCUSSION OF BOX MODEL CALCULATIONS

Using R and T values derived from earlier investigators (Somayajulu and Craig, 1976; Chung and Craig, 1983) for the simple box model described in the previous section, we have obtained a mean residence time of 0.9 and 1.5 years respectively for Po-210 and Pb-210 within the upper layer. These values are

comparable to those of 0.5 ~ 5 and 0.6 *years* obtained by earlier investigators for the mixed layer in open oceans (e.g. Shannon *et al.*, 1970; Nozaki *et al.*, 1973; Bacon *et al.*, 1976). In the dissolved phase the mean residence time of Po-210 is shorter than that of Pb-210 because Po-210 is more rapidly removed, perhaps scavenged by sinking organic particles, especially phytoplankton-genetic particles (Shannon *et al.*, 1970).

The model calculations also suggest that at least 40% of the Po-210 removed from the upper layer is recycled in the middle layer by dissolution or desorption of the nuclide from sinking organic particles. This may explain why Po-210 is at its maximum in the middle layer. Similar calculations suggest a much lower recycling efficiency (< 5%) for Pb-210. Thus, the Pb-210 scavenged by sinking particles sinks continuously to the bottom without being recycled back to the dissolved phase. Pb-210 derived from the atmosphere can be observed as an excess relative to the Ra-226 and can penetrate to a depth of 500 to 1000 *m* (Craig *et al.*, 1973). The Pb-210 minimum in our profiles is observed in the lower layer. The mean residence time for Pb-210 in the lower layer at station H16 (12 *years*) is shorter than that at station R23 (120 *years*) by a factor of 10, because the lower layer at H16 is quite close to the ocean floor where bottom boundary scavenging of resuspended particulate matter may dominate.

In order to see how R and T values affect the mean residence time of these nuclides in the upper layer and their recycling efficiency in the middle layer, different R and T values are used for the model calculations. If the R value in the upper layer increases, the mean residence time for the dissolved phase of these two nuclides will decrease, but that for the particulate Po-210 (as well as Pb-210) will increase proportionally or much more sensitively. The mean residence time for particulate Po-210 and dissolved Pb-210 will be doubled if the T value is doubled. However, it is unlikely that T can be twice as much as the reported value of about 1.05. The mean residence time for both nuclides in the dissolved phase, τ_{Pb}^d and τ_{Po}^d , decreases only slightly even if R increases 8-fold (from 2% to 16%) under a normal T value of 1.05. Po-210 recycling in the middle layer vanishes if T is 2.10 and R is equal to or greater than 10%. The Pb-210 recycling efficiency decreases as R increases but it increases as T increases. This is expected since a higher T value means relatively lower Pb-210 activities in the particulate phase, and so higher recycling efficiency.

6. COMPARISON OF THE SHELF WATER PROFILE (F1) WITH THOSE OF THE KUROSHIO WATER (H16, R19, R23)

The Pb-210 and Po-210 data collected in profile in the East China Sea shelf water (F1) are listed in Table 1. The data show that Po-210 exceeds Pb-210 below the 25 *m* depth and the excess extends to the bottom. This is quite different from the Po-210 excess observed at the profiles located in the

Kuroshio water where the Po-210 excess occurs below the 150 m depth. The Po-210 excess in the shelf water may result from intrusion and upwelling of the Kuroshio middle layer water which is in turn enriched with Po-210 due to recycling by dissolution or desorption. The upwelling feature near the shelf edge area northeast of Taiwan has been observed and documented repeatedly (e.g. Fan, 1980; Liu and Pai, 1987; Liu *et al.*, 1988; Chern *et al.*, 1990; Wong *et al.*, 1991) and by the KEEP (Kuroshio Edge Exchange Processes) hydrographic surveys. This upwelling of the Kuroshio subsurface water is now believed to be a permanent feature induced by the shelf edge topography causing the Kuroshio to swing toward the northeast and to exchange with the East China Sea water. Since station F1 is fairly close to the upwelling center, Kuroshio subsurface water is expected to intrude and upwell at this station. The Po-210 excess observed at shallow depths in the F1 profile (below 25 m) suggests that the Kuroshio subsurface water with its Po-210 excess (our middle layer water) has upwelled about 150 m. This is consistent with hydrographic observations in which the 17°C isotherm has ascended 130 m from a depth of 200 m (the depth of maximum Po-210 excess observed in the Kuroshio) to 70 m (Chern *et al.*, 1990).

7. THE DISTRIBUTIONS OF Pb-210 AND Po-210 IN THE SURFACE WATER

Pb-210 in the surface water comes mainly from the atmosphere where it is produced by the decay of Rn-222. Po-210 in the surface water is produced mainly by the decay of Pb-210 within the surface water. Both nuclides may be removed from the surface water by the following processes:

- (1) The nuclides are transferred from the dissolved phase to the particulate phase, either by adsorption onto particles or uptake by living organisms;
- (2) The nuclides attached to particles or organisms may be removed from the surface water by gravitational sinking.

The measurements results of both nuclides in the surface water are listed in Table 4, and the station locations are shown in Figure 2. The Pb-210 activities are about 2.6 to 10.1 *dpm/100 kg* higher than the Po-210 activities, suggesting a rapid removal of Po-210 by sinking particles and an excess of Pb-210 maintained by the atmospheric flux. Surface water Po-210 and Pb-210 across the shelf to the slope off northeastern Taiwan where the Kuroshio flows, increase systematically from station F1 in the shelf area to F13 within the Kuroshio current. Both Pb-210 and Po-210 activities for individual stations are plotted as a function of the distance from station F1 and are shown in Figure 5. Po-210 increases from 3.8 to 10.2 *dpm/100 kg* while Pb-210 increases from 6.4 to 16.8 *dpm/100 kg*. Po-210 is consistently deficient with a maximum deficit of 9.4 *dpm/100 kg* at F9, about 50 nautical miles (nm) away from F1.

Table 4. Pb-210 and Po-210 activities measured from surface water stations. Stations locations are shown in Figure 2. One sigma errors are about 6% for Pb-210 and about 10% for Po-210.

Station	Pb-210(<i>dpm/100 kg</i>)	Po-210(<i>dpm/100 kg</i>)
F1	6.4±0.4	3.8±0.4
F5	8.1±0.4	4.5±0.5
F7	11.6±0.8	5.7±0.6
F9	14.7±1.0	5.3±0.6
F11	14.3±0.9	7.4±0.8
F13	16.8±0.7	10.2±1.0
L1	8.9±0.4	4.8±0.5
L2	7.9±0.5	4.4±0.4
L3	9.6±0.5	5.7±0.6
L4	11.4±0.5	6.9±0.7
R6	7.8±0.5	4.1±0.5
R9	8.6±0.5	5.1±0.5
R13	13.8±0.8	8.3±0.9
R14	12.9±0.8	8.1±0.9
R16	8.5±0.5	4.4±0.4
R17	5.5±0.3	2.2±0.3
R18	6.5±0.4	3.7±0.4
R19	10.8±0.6	6.4±0.6
R20	15.1±0.9	8.7±1.0
R21	11.2±0.6	5.4±0.6
R22	14.1±0.8	7.8±0.8
R22A	13.9±1.0	8.0±0.8
R23	13.5±0.8	6.1±0.6
R23A	17.6±1.0	9.2±1.0
R24	16.2±1.0	10.1±1.1
R25	15.3±0.9	9.8±0.9
R26	13.5±0.9	7.9±0.8
R27	12.1±0.7	6.5±0.7
R28	12.8±0.7	7.2±0.7
R29	8.6±0.4	4.5±0.5
H8	9.4±0.6	5.2±0.6
H10	12.6±0.7	7.3±0.7
H11	11.5±0.7	6.8±0.8
H12	14.7±0.9	8.2±0.9
H13	13.8±0.8	7.5±0.8
H14	13.2±0.7	8.0±0.9
H15	17.5±0.9	10.5±1.1
H16	19.4±1.0	9.3±0.9

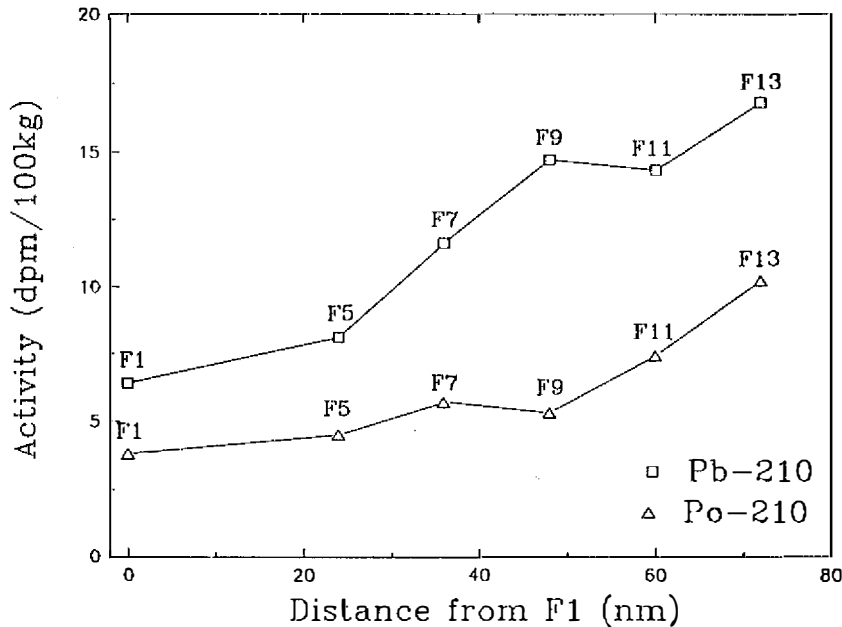


Fig. 5. Systematic surface water Pb-210 and Po-210 increases along a NW-SE transect off Keelung, away from the shelf water at F1, to the Kuroshio water at F13. A Po-210 deficit relative to Pb-210 is a distinct feature.

F9 appears to be at the frontal zone of the Kuroshio where topographically induced upwelling is known to take place (Wong *et al.*, 1991). This particularly low Po-210 value may be due to enhanced productivity and so enriched organic particulates. Since the atmospheric Pb-210 flux is the main source and is presumably fairly uniform within such a small area, this distribution reflects a more efficient removal of these nuclides from the surface water in the shelf area, where it may also be enhanced by boundary scavenging.

In the Kuroshio surface water off eastern and northeastern Taiwan, a similar trend is observed: both nuclides increase from the land mass toward the Kuroshio water or the deep sea, and Pb-210 is consistently higher than Po-210, as can be seen in Figure 6. Although the distribution off eastern Taiwan (Figure 6B) is less than systematic, the trend remains clear if a few low values from the deep water area are excluded, particularly that at R16. Such a distribution pattern may reflect the combined effects of particle scavenging and boundary scavenging, since higher concentrations of particulate matter are often observed in coastal waters (Tsai, personal communications). The Pb-210 and Po-210 distributions in surface waters off eastern and northeastern Taiwan are shown in Figure 7. Both nuclides show a similar pattern with contours turning toward the northeast near $25^{\circ}N$ latitude. This feature coincides with the Kuroshio path which also turns northeast in the same area. Along the Kuroshio current, both nuclides remain fairly constant and the Po-210 deficiency remains fairly

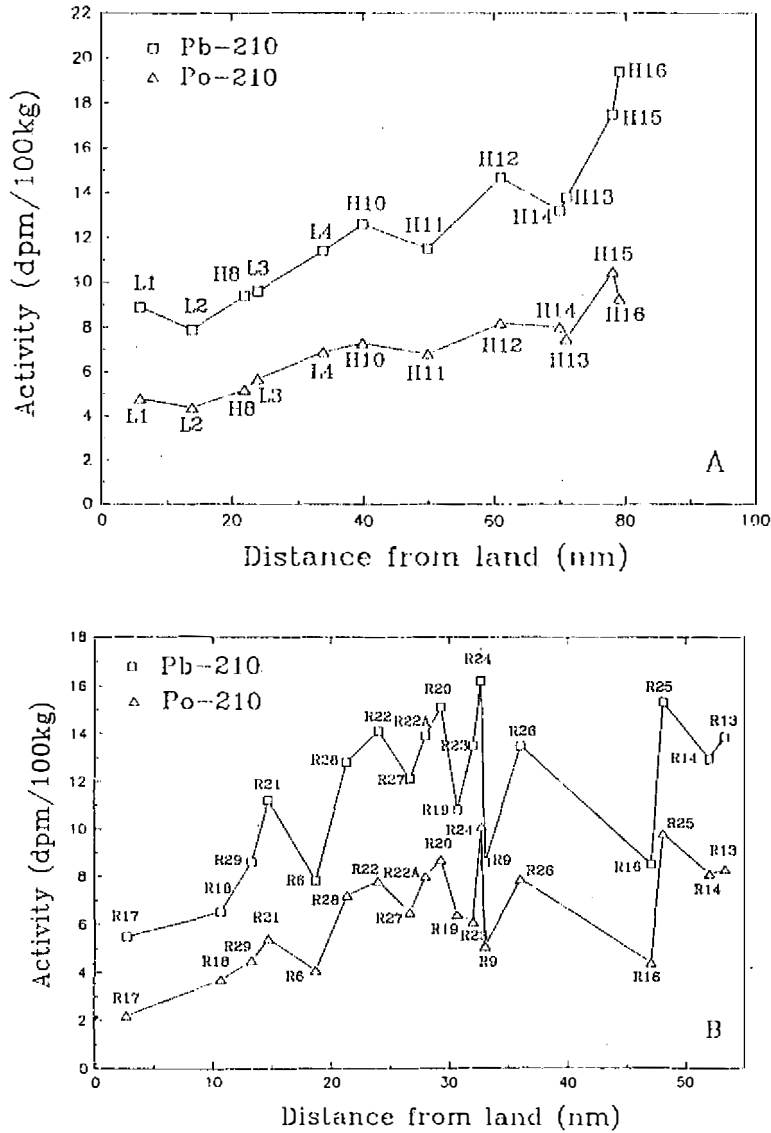


Fig. 6. Surface water Pb-210 and Po-210 distributions as a function of distance from land: (A) off northeastern Taiwan from OR#1, Cruises 249 and 259, showing a systematic increase for both nuclides toward the deep sea; (B) off eastern Taiwan from OR#1, Cruise 255, showing a more complicated variation for both nuclides across the Kuroshio.

uniform with a Po-210/Pb-210 activity ratio of about 0.5 to 0.6.

Figure 8 shows the Po-210/Pb-210 activity ratio plotted against distance from the nearest land in nautical miles (nm). The data from all three cruises show a fairly narrow range of variation with a mean of 0.56 and a standard deviation of 0.05. This is a very interesting feature and implies a mean residence time of about 0.7 years for Po-210 in surface water under a steady-state

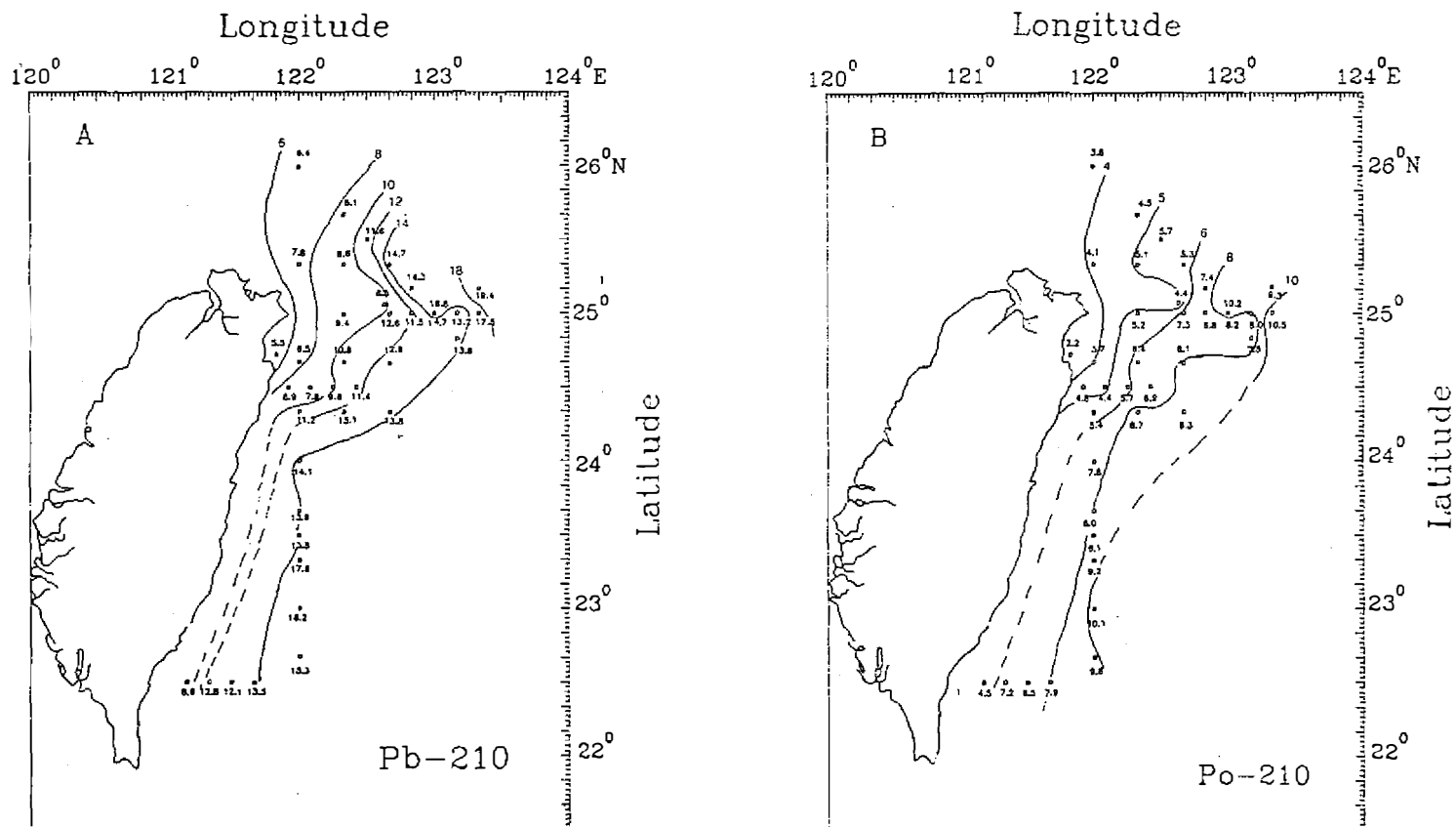


Fig. 7. Surface water Pb-210 (A) and Po-210 (B) distributions off eastern and northeastern Taiwan. The pattern for both contours is similar but the Po-210 values are much lower than the Pb-210 values.

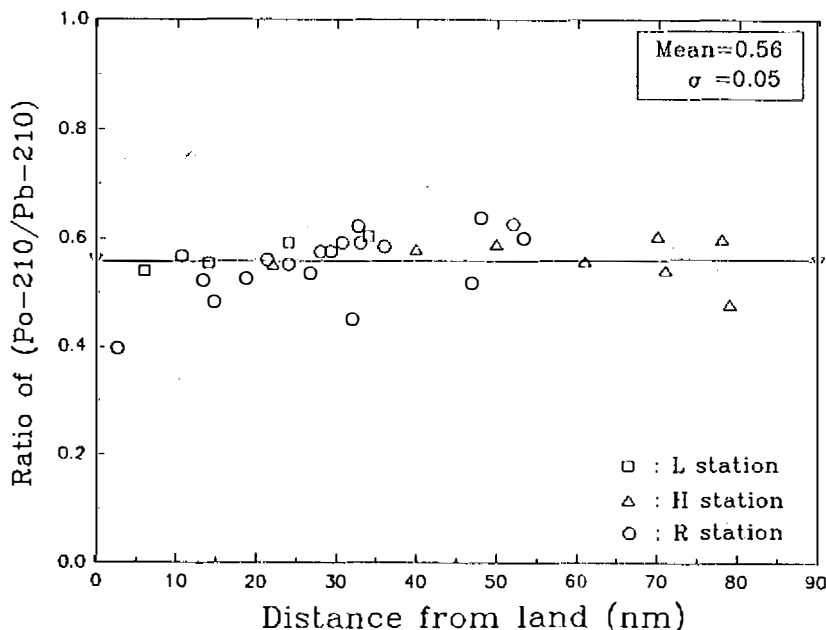


Fig. 8. Plots of surface water Pb-210/Pb-210 activity ratio against distance from land. Cruise 249 (L), 255 (R) and 259 (H) data all indicate a fairly constant ratio of about 0.5 to 0.6 (0.56 ± 0.05), corresponding to a mean residence time of about 0.7 years for Po-210 under a steady-state assumption.

assumption. This is quite consistent with the results obtained by others as well as with our own model calculation for the upper layer.

8. SUMMARY AND CONCLUSIONS

A water column in the Kuroshio may be divided into three layers based on the activity difference or ratio between Po-210 and Pb-210 measured in a profile. In the upper layer (0 ~ 150 m), Po-210 is deficient, suggesting a preferential removal of Po-210. In the middle layer (150 ~ 500 m), Po-210 is enriched, indicating a net input of Po-210 most likely due to desorption or dissolution of the nuclide from sinking particles. In the lower layer (below 500 m), a radioactive equilibrium between the two nuclides is reached, indicating that the mean residence time of Po-210 with respect to removal is over 2.5 years at least. These results are consistent with previous observations in the open oceans. Box model calculations with a material balance under the steady-state assumption suggest the following:

- (a) The mean residence time for Po-210 and Pb-210 in the upper layer is 0.9 and 1.5 years respectively. Both nuclides are highly reactive, especially Po-210, and so are rapidly removed from the upper layer by particle scavenging.

- (b) At least 40% of the Po-210 removed from the upper layer is recycled within the middle layer, as manifested by a Po-210 maximum there. A much lower recycling efficiency ($< 5\%$) is obtained for Pb-210, indicating that the Pb-210 adsorbed by particles may reach the bottom without being recycled through its settling course.
- (c) The Pb-210 minimum exists in the lower layer, because the influence of Pb-210 delivered from the atmosphere diminishes with depth. The mean residence time for Pb-210 at station H16 (12 years) is one tenth that at station R23 (120 years), suggesting the additional effect of bottom boundary scavenging by resuspended particulates at H16.

If the particulate Pb-210 fraction (R) in the upper layer is increased, then the mean residence time for both nuclides in the dissolved phase will decrease, but the mean residence time for particulate Po-210 as well as Pb-210 will increase. The effect is more sensitive for the particulate phase. The mean residence time for particulate Po-210 and dissolved Pb-210 will increase proportionally with the activity ratio of Po-210 to Pb-210 in the particulate phase or the T value. However, it is unlikely that the T value can be much higher than the reported value of 1.05.

The water column over the East China Sea shelf zone has Po-210 in excess of Pb-210 below the 25 m depth and the excess extends to the bottom. This is quite different from the Po-210 excess observed in the Kuroshio water where the Po-210 excess occurs below the 150 m depth. Intrusion and upwelling of the Kuroshio subsurface (middle layer) water which has a Po-210 excess may be responsible for the Po-210 excess observed in the subsurface shelf water. This interpretation is consistent with many previous observations as well as the KEEP hydrographic observations.

In the surface water, Pb-210 activities are always higher than Po-210 activities, due to a rapid removal of Po-210 and an excess of Pb-210 maintained by the atmospheric flux. The removal of Pb-210 and Po-210 in the surface water from the shelf area and the area near the land mass is more effective than from the Kuroshio region and the deep sea area. Such a distribution pattern may reflect the combined effects of particle scavenging and boundary scavenging. Both Po-210 and Pb-210 along the Kuroshio are fairly uniform and the contours coincide with the Kuroshio path that turns northeastward near $25^\circ N$ latitude. Despite the fact that both Po-210 and Pb-210 in the surface water increase toward the Kuroshio and the deep sea, their activity ratio remains fairly uniform at about 0.5 to 0.6 (0.56 ± 0.05), corresponding to a mean residence time of about 0.5 to 0.8 years for Po-210 in all the surface waters. These results are consistent with those obtained by others and those calculated from a box model. The effect of the Kuroshio on the behavior of these nuclides remains unclear at present.

Acknowledgements. The first author wishes to thank the staffs of the Radioisotope Laboratory of the Institute of Marine Geology, National Sun Yat-sen University, for their assistance during the years when he was conducting his M.S. thesis research. This paper is a condensed form of this thesis. Both authors acknowledge the generous help received from the technical staffs and crews of the R/V Ocean Researcher I and Haifu. This work has been supported by grants (NSC 79-0209-M110-02 and NSC 80-0209-M110-06) from the National Science Council to the Institute of Marine Geology, National Sun Yat-sen University.

REFERENCES

- Bacon, M. P., D. W. Spencer and P. G. Brewer, 1976: Pb-210 Ra-226 and Po-210/Pb-210 disequilibrium in seawater and suspended particulate matter, *Earth and Planetary Science Letters*, **32**, 277-296.
- Burton, W. M. and N. G. Stewart, 1960: Use of long-lived natural radioactivity as an atmospheric tracer, *Nature*, **186**, 584-589.
- Chern, C. -S., J. Wang and D. -P. Wang, 1990: The exchange of Kuroshio and East China Sea shelf water, *Journal Geophysical Research*, **95**, 16017-16023.
- Chung, Y. and H. Craig, 1980: Ra-226 in the Pacific Ocean, *Earth and Planetary Science Letters*, **49**, 267-292.
- Chung, Y., R. C. Finkel and K. Kim, 1982: Ra-226, Pb-210 and Po-210 in the Red sea, *Earth and Planetary Science Letters*, **58**, 213-224.
- Chung, Y. and H. Craig, 1983: Pb-210 in the Pacific: the GEOSECS measurements of particulate and dissolved concentrations, *Earth and Planetary Science Letters*, **65**, 406-432.
- Chung, Y., R. Finkel, M. P. Bacon, J. K. Cochran and S. Krishnaswami, 1983: Intercomparison of Pb-210 measurements at GEOSECS station 500 in the northeast Pacific, *Earth and Planetary Science Letters*, **65**, 393-405.
- Chung, Y. and R. Finkel, 1988: Po-210 in the western Indian ocean: distributions, disequilibria and partitioning between the dissolved and particulate phase, *Earth and Planetary Science Letters*, **88**, 232-240.
- Craig, H., S. Krishnaswami and B. L. K. Somayajulu, 1973: Pb-210-Ra-226: radioactive disequilibrium in the deep sea, *Earth and Planetary Science Letters*, **17**, 295-305.
- Fan, K. L., 1980: On upwelling off northeastern shore of Taiwan, *Acta Oceanographica Taiwanica*, **11**, 105-117.
- Liu, C. -T. and S. -C. Pai, 1987: As Kuroshio turns, II, the oceanic front north of Taiwan, *Acta Oceanographica Taiwanica*, **18**, 49-61.
- Liu, K. -K., S. -C. Pai and C. -T. Liu, 1988: Temperature-nutrient relationships in the Kuroshio and adjacent water near Taiwan, *Acta Oceanographica Taiwanica*, **21**, 1-17.
- Nozaki, Y. and S. Tsunogai, 1973: Lead-210 in the North Pacific and the transport of terrestrial material through the atmosphere, *Earth and Planetary Science Letters*, **20**, 88-92.
- Nozaki, Y. and S. Tsunogai; 1976: Ra-226, Pb-210 and Po-210 disequilibria in the western North Pacific, *Earth and Planetary Science Letters*, **32**, 313-321.
- Nozaki, Y., J. Thomson and K. K. Turekian, 1976: The distribution of Pb-210 and Po-210 in the surface waters of the Pacific Ocean, *Earth and Planetary Science Letters*, **32**, 304-312.

- Nozaki, Y., N. Ikuta and M. Yashima, 1990: Unusually large Po-210 deficiencies relative to Pb-210 in the Kuroshio Current of the East China and Philippine Seas, *Journal of Geophysical Research*, 5321-5329.
- Poet, S. E., H. E. Moore and E. A. Martell, 1972: Lead-210, bismuth-210 and polonium-210 in the atmosphere: accurate ratio measurement and application to aerosol residence time determination, *Journal of Geophysical Research*, 77,6515-6527.
- Shannon, L. V., R. D. Cherry and M. J. Orrer, 1970: Polonium-210 and lead-210 in the marine environment, *Geochimica et Cosmochimica Acta*, 34, 701-711.
- Somayajulu, B. L. K. and H. Craig, 1976: Particulate and soluble Pb-210 activities in the deep sea, *Earth and Planetary Science Letters*, 32, 268-276.
- Turekian, K. K., D. P. Kharkar and J. Thomson, 1974: The fate of Pb-210 and Po-210 in the ocean surface, *Journal of Atmospheric Research*, 8, 639-657.
- Wong, G. T. F., S. C. Pai, K. K. Liu, C. T. Liu and C. T. A. Chen, 1991: Variability of the chemical hydrography at the frontal region between the East China Sea and the Kuroshio northeast of Taiwan, Estuarine, *Coastal and Shelf Science*, 33, 105-120.

台灣東方及東北海域黑潮水中鉛-210 及釷-210之分佈及其放射性不平衡狀態

林育農 鍾玉嘉

國立中山大學海洋地質研究所

摘要

本文主要探討台灣東部外海及東北海域，黑潮與東海海水混合區域中之 Pb-210 和 Po-210 的分佈與其放射性不平衡狀態。在黑潮及其邊緣水體之水域中，四個垂直剖面及三十四個表水測站皆完成 Pb-210 和 Po-210 總活性之偵測。

沿著黑潮北流路徑的三個剖面測站具有一些共同的特徵：Pb-210 活性最大值皆在深度 25 ~ 100m 間；Po-210 的最大值則在甚接近 200m 深度處；二核種在 200m 以下皆隨深度遞減且在 500m 以下達活性平衡。因此每一水柱可依據此二核種之活性比區分成三層。在表層水（0 ~ 150m）中，Po-210/Pb-210 比值小於 1，顯示 Po-210 可能較先由水中移走。於中層水（150 ~ 500m）中，此比值大於 1，顯示 Po-210 可能因溶解或脫附作用而再循環。在深層水（500m 以下）中，此比值大約等於 1，顯示此二核種幾乎已達活性平衡。

利用一簡單的盒子模式（box model）並假設其在穩定狀態下（steady-state），可計算出表層水中 Po-210 和 Pb-210 之駐留時間各為 0.9 和 1.5 年。由於此二核種對水中顆粒物質的反應性極高，致使其在表層水

中快速地被顆粒態物質所清除。然而由表層水去除之 Po-210 至少有 40% 在中層水中溶解而再循環。而只有少於 5% 的 Pb-210 以此路徑再循環。這些特徵在開放的大洋中是常見的，這表示黑潮本身並沒有影響到此二核種的分佈。在測站 H16 的深層水中 Pb-210 的平均駐留時間是 12 年，只有另一測站 R23 的十分之一，顯示在測站 H16 中，顆粒物質的邊界和現場清除作用可能較為有效。

在東海陸棚水中，Po-210 於表水以下 25m 處即呈超量狀態且延伸至海底（120m）。這與黑潮水中所觀察到的現象截然不同，在黑潮水中超量 Po-210 出現在 150m 以下。東海陸棚水中之超量 Po-210 可能是由於因再循環作用而富含 Po-210 之黑潮中層水的侵入和湧昇所造成。此現象和由水文調查，觀察到之黑潮次表層水被抬升約 130m 的結果一致。

在表水測站中 Po-210 和 Pb-210 之活性濃度由近陸水域或陸棚區朝深海增加，反應了顆粒物質濃度和邊界清除作用的影響。然而所有的表水樣品 Po-210/Pb-210 之活性比皆相當接近，為 0.5~0.6，顯示 Po-210 的平均駐留時間約為 0.5 至 0.8 年。此結果和由盒子模式（box model）在穩定狀態（steady-state）下進行顆粒清除作用時，計算所得之表層水（0~150m）的平均駐留時間 0.9 年大致吻合。