Behavior of ^7Be in Funka Bay, Japan, with reference to those of insoluble nuclides, ^{234}Th , ^{210}Po and ^{210}Pb

Noriyuki TANAKA* and Shizuo TSUNOGAI

Department of Chemistry, Graduate School of Fisheries, Hokkaido University, Hakodate 041, JAPAN

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^7Be in seawater and sediment trap samples in Funka Bay, Japan was determined together with ^{210}Pb , ^{210}Po and Th isotopes. The sampling was carried out in summer in 1977 and 1979, when the Bay constituted a closed system for mixing with water outside the Bay. The concentration of ^7Be of atmospheric origin in seawater did not monotonously decrease with depth, but showed a weak maximum in the intermediate layer. The layer also contained larger amounts of ^{234}Th , ^{210}Po and ^{210}Pb in summer, 1979. The results obtained from sediment trap samples suggest that significant portions of these nuclides and some other heavy metals are transported downward by particulate matter. The residence time of Be in Funka Bay (mean depth 60m) was calculated to be 90–160 days from the activity ratio of $^7\text{Be}/^{234}\text{Th}$ of the trapped material and the removal rate of ^{234}Th from seawater obtained from the disequilibrium with ^{238}U .

These facts also support the TSUNOGAI and MINAGAWA's settling model for the removal of insoluble metals from seawater rather than the irreversible removal from seawater and the rapid transport of these metals in particles.

INTRODUCTION

A cosmic ray produced radioisotope, ^7Be (half-life, 53 days), may be useful to monitor various processes occurring in the hydrosphere on a time scale from a few tens to 200 days. LAL (1963) calculated the mean concentration of ^7Be in the surface seawater (depth 75 m) to be 0.18 dpm/l from its production rate in the atmosphere and reported it to be 0.01 to 0.07 dpm/l in the surface water of the Indian Ocean. YOUNG and SILKER (1974) measured ^7Be in the surface seawater and estimated the deposition rate of ^7Be from the atmosphere and the vertical diffusion coefficient of the surface seawater of 0.02 to 20 $\text{cm}^2 \text{ sec}$.

Be is one of the elements having the shortest residence times around 100 years in the oceans such as Al, Fe, Ti and Th discussed by TSUNOGAI and MINAGAWA (1978). This fact implies that Be is also related to the particulate

removal in the oceans.

Therefore, ^7Be can be used as a tracer for the study of the particulate transport process in the surface layer of the oceans. The advantage of using ^7Be as a tracer is that its source in the oceans is confined to the surface where it is supplied largely from wet precipitation and that the transport rate from the surface to the deeper layers might be estimated directly from its vertical profile and its half life. BISHOP *et al.* (1977) collected a large amount of particulate matter from the subsurface water by using a large volume *in situ* filtration system (LVFS) and suggested that the particulate removal of ^7Be is due to the surface adsorption rather than the active biological uptake, since the fraction of particles larger than 58 microns contained no measurable activity of ^7Be . SILKER (1972) have suggested that the particulate transport of ^7Be is not important in the oceans, since particulate fraction of ^7Be activity in seawater was only about 10% or less.

*Present address: Department of Geology & Geophysics Yale University, New Haven, Conn. 06520 U.S.A.

The behavior of ^7Be in seawater is worthwhile to be compared with those of other insoluble nuclides such as Th isotopes, ^{210}Po and ^{210}Pb because they are also removed rapidly from seawater (SHANNON *et al.*, 1970; BROECKER *et al.*, 1973; BHAT *et al.*, 1969; MATSUMOTO, 1975; ALLER and COCHRAN, 1976; NOZAKI and TSUNOGAI, 1976; BENNINGER, 1976; KHARKAR *et al.*, 1976; TUREKIAN, 1977; SANTSCHI *et al.*, 1979). SPENCER *et al.* (1978) and BREWER *et al.* (1980) have asserted that the removal of such elements by rapidly sinking particles, "zooplankton fecal pellets", is more important than that by aggregates of suspended matter. BEASLEY *et al.* (1978) have also stated fecal pellets to be an important conveyer for the vertical transport of ^{210}Po and ^{210}Pb . On the other hand, TSUNOGAI and MINAGAWA (1978) have concluded that particles conveying these radionuclides should be similar to suspended matter and thus large aggregates, "marine snow", may be responsible for their downward transport, because marine snow grows up from

particles of colloidal size and break down to small particles while sinking in seawater.

We determined ^7Be together with ^{234}Th , ^{210}Po and ^{210}Pb in seawater and in settling particles collected with sediment traps in Funka Bay in 1977 and 1979. This paper is intended to show that the distribution of ^7Be strongly depends on the particulate transport and that ^7Be has an intimate relation with other insoluble radioisotopes.

MATERIALS AND METHOD

Figure 1 shows the locations of observed stations in Funka Bay. Sediment trap experiments were carried out at St. P. The hydrographic surveys have been done since 1974 by OHTANI and KIDO (1980) and others. The cold water of the Oyashio flows into this Bay in early spring. In summer, the water in the Bay is strongly stratified and stays without significant mixing with water outside the Bay. In early autumn, the Tsugaru Warm Current flows into the Bay and mixes vertically well by cooling in

Fig. 1. Locations of sampling stations. The water depths at St. P ($42^{\circ}16.3'\text{N}$, $140^{\circ}35'\text{E}$) and St. 4 ($42^{\circ}00'\text{N}$, $141^{\circ}28'\text{E}$) were 92 and 515 m, respectively.

winter. The winter water mass stays in the Bay till the water of Oyashio flows in.

The sediment trap employed consists of twin rectangular P.V.C. boxes and its opening area and height are 0.17 m^2 and 25 cm , respectively. Under each box, a pyramidal P.V.C. funnel (height 30 cm) with a plastic bottle was attached. The seawater for the determination of ^7Be was pumped up into a 500-liter polyethylene container on the vessel and the seawater for the determinations of ^{210}Po , ^{210}Pb and ^{234}Th was collected with 50-liter Van Dorn samplers.

^{234}Th , ^{210}Po and ^{210}Pb were co-precipitated with ferric hydroxide and calcium carbonate from 50-liter of seawater after the solution acidified with nitric acid containing ^{232}Th , ^{208}Po and stable Pb stood for about 6 hrs. After Po was deposited on a silver disk, Th was separated from the solution by an ion exchange technique. The fraction containing Pb was stored for about a year for the growth of ^{210}Po , a daughter of ^{210}Pb . The ^{210}Pb activity was calculated from the ^{210}Po activity grown during the storage. Th was electrodeposited on a silver disk and its beta and alpha activities were counted with a low background GM counter of an anticoincidence type and a multi-channel pulse height analyzer with a silicon surface barrier detector, respectively. The detail of this method will be reported elsewhere by HARADA and TSUNOGAI.

Seawater pumped up into a 500-liter polyethylene container on the vessel was analyzed for ^7Be . The seawater was acidified with 1.5-liters of conc. H_2SO_4 containing 25 mg of stable Be. ^7Be was co-precipitated with ferric hydroxide and calcium carbonate. The precipitates obtained were dissolved in 7.5N HNO_3 and the solution was heated to remove dissolved carbon dioxide. Ammonia water was then added to the solution and the precipitate of hydroxide formed was filtered. The precipitate was dissolved in 200 ml of 6 N HCl and the solution was shaken with 100 ml of methyl isobutyl ketone to extract almost all Fe. The water phase was completely dried

up on a hot plate and 10 ml of 0.5 N HCl and 200 ml of 20% NaOH solution were added. Be was left in the solution, while Fe remaining after the extraction was precipitated as hydroxide, which was filtered off with a glass fiber filter. The filtrate was acidified with 6N HCl and pH was adjusted with ammonia water to 9.6 with TB, as an indicator to precipitate beryllium hydroxide. The precipitate collected was dissolved in 50 ml of 0.5N HCl , and 1 g of EDTA-2Na salt was added and pH was adjusted to 5.3 with dilute ammonia water. After addition of an acetate buffer (pH 5.3), a mixture of 25 ml of acetyl acetone and 100 ml of benzene, the solution was shaken for 10 minutes. The organic phase was separated and washed twice with 20 ml of the acetate buffer (pH 5.3). Be was then back extracted into 50 ml of 6N HCl . The volume of the solution was adjusted to 100 ml and 1 ml of its aliquot was used for chemical yield determination of Be by atomic absorption spectrometry. The remaining fraction was heated to remove benzene completely. Beryllium hydroxide was precipitated with ammonia water, filtered, and ignited at 750°C to convert to BeO in an electric furnace. The γ -activity of BeO in a polyethylene tube was counted by a well-type scintillation counter with a NaI(Tl) scintillator. The counting efficiency of the counter was $17.7 \pm 1.35\%$ and the overall chemical yield was 24 to 75% (51% on an average).

The material obtained with sediment traps was filtered through a glass fiber filter (Whatman GF/C) or a Nucleopore filter ($0.6\text{ }\mu\text{m}$ in pore size). Followed by the digestion of a sample on a filter with a mixture of HF , HNO_3 and HClO_4 , one twentieth fraction of the solution was analyzed for ^{234}Th and the remainder for ^7Be , ^{228}Th , ^{230}Th and ^{232}Th . The subsequent procedures for ^{234}Th and ^7Be determinations were the same as those described above. The long lived Th isotopes were separated from the solution by an ion exchange technique and electrodeposited on a silver disk. Its alpha and beta activities were counted.

Fig. 2. Vertical profiles of ^7Be and σ_t observed on May 1, July 23 and Aug. 22, 1977.

RESULTS AND DISCUSSION

^7Be , ^{234}Th , ^{210}Po and ^{210}Pb in seawater The vertical profiles of ^7Be and the density of the water (σ_t) in Funka Bay are plotted in Fig. 2. The profiles of σ_t show that the seasonal thermocline well developed in July and August in the surface 40m layer, but it was not obvious in May. The overall analytical error for ^7Be seems to be not larger than the counting error of about 11% for the duplicate analysis of the same sample. The concentration of ^7Be in May is relatively constant with depth, showing 240 dpm/1000l in the surface water and 184 dpm/1000l in the deep water of 70m depth. The concentration of ^7Be in the intermediate layer, however, increased during summer from July to August, and a maximum value was always found in the layer. The inventory of ^7Be (dpm/unit area) in the three profiles differs by a factor of 3. This may be due to the intense fluctuations of the input rate of ^7Be from the atmosphere and the removal rate of ^7Be from seawater as well as its short residence time in seawater.

The data on ^{234}Th , ^{210}Po and ^{210}Pb in seawater at the same site on Aug. 20, 1979 are given in Fig. 3 together with the density of seawater (σ_t). The profile of σ_t at St. P on Aug. 20, 1979 is almost the same as that on Aug. 22, 1977. ^{210}Po and ^{210}Pb are slightly concentrated in the intermediate layer, and ^{234}Th also appears to be concentrated in the layer, although its values are much scattered. The previous studies on these nuclides in coastal seawater have not clearly shown their maximum layers or convexed vertical distributions. (BHAT *et al.*, 1969; MATSUMOTO, 1975) The sampling locations studied may have offered too shallow water depths or the sampling depth intervals may have been too large to reveal the maximum layer. However, we can find that ^{234}Th was concentrated in the intermediate layer in the report of MINAGAWA and TSUNOGAI (1980) who determined ^{234}Th in seawater at the station near St. P in this study on Aug. 12, 1974.

^{234}Th is a fully radiogenic nuclide in seawater, but a half of ^{210}Po , a substantial part of ^{210}Pb and all of ^7Be in seawater are airborne.

Fig. 3. Vertical profiles of σ_t , ^{234}Th , ^{210}Pb and ^{210}Pb observed at St. P on Aug. 20, 1979.

In spite of their different source functions, the similarity of their convex profiles for 30 to 60 m depths suggests that the behaviors of nuclides are controlled by the similar factor in the layer. It seems likely that these nuclides are incorporated into organic particles and sink with them. However, the decrease in the

dissolved oxygen in the layer is not so obvious (Fig. 4) and regenerated amounts of the nuclides calculated from these nutrients and their ratios to nutrient in settling matter in seawater are insufficient to explain the surplus in the layer. The radionuclides, therefore, are necessary to be generated in a dissolved form from sinking

Fig. 4. Dissolved oxygen and phosphate observed at St. P in the summer of 1977.

Table 1 The concentrations of ^7Be , ^{234}Th , ^{228}Th , ^{230}Th and ^{232}Th in the settling matter collected with sediment traps at St. P in Funka Bay.

Sampling depth (m)	Specific activity (dpm/gram of dry weight)				
	$^7\text{Be}^{*1}$	$^{234}\text{Th}^{*1}$	^{228}Th	^{230}Th	^{232}Th
June 27 - July 23, 1977					
40 m	130	590	1.9 ± 0.1	0.6 ± 0.1	0.4 ± 0.2
80 m	140	1,020	3.0 ± 0.2	1.0 ± 0.1	0.6 ± 0.2
July 26 - Aug. 22, 1977					
40 m	180	610	2.0 ± 0.1	0.4 ± 0.03	0.2 ± 0.02
80 m	150	790	2.7 ± 0.1	0.8 ± 0.03	0.7 ± 0.02
Sept. 14 - Nov. 21, 1977 ^{*2}					
40 m F	70	1,770	2.8 ± 0.2	0.5 ± 0.01	1.8 ± 0.2
B	190	2,540	0.4 ± 0.1	0.03 ± 0.02	0.1 ± 0.03
80 m F	120	2,280	12.6 ± 0.5	1.1 ± 0.2	1.9 ± 0.2
B	220	1,320	4.8 ± 0.4	0.4 ± 0.1	0.5 ± 0.1

*1 The radioactivities of these short-lived isotopes have been corrected for the decay during the sampling period, assuming constant fluxes with time. Errors for the determinations of ^7Be and ^{234}Th are 20–30 dpm/g and 40–70 dpm/g, respectively.

*2 Cited from TSUNOGAI et al. (1980). Fraction F is of light particles suspending in the funnel and collected by filtering and fraction B is of dense particles settled in the bottle containing saturated NaCl soln. attached to the end.

particles without decomposition of organic fraction. The chemical nature of the nuclides suggests that regeneration is extremely difficult. Or otherwise this may suggest that the settling

particles are broken up to small particles, which can pass through the pore ($0.4 \mu\text{m}$) of membrane filter and stop settling down or slow down their sinking velocity in the layer.

Table 2 The activity ratios of $^7\text{Be}/^{234}\text{Th}$, $^{228}\text{Th}/^{234}\text{Th}$, $^{230}\text{Th}/^{234}\text{Th}$ and $^{232}\text{Th}/^{234}\text{Th}$ in settling matter collected with sediment traps.

Sampling depth (m)	$^7\text{Be}/^{234}\text{Th}$	$^{228}\text{Th}/^{234}\text{Th}$	$^{230}\text{Th}/^{234}\text{Th}$	$^{232}\text{Th}/^{234}\text{Th}$
June 27 - July 23, 1977				
40 m	0.22	3.2×10^{-3}	9.3×10^{-4}	6.2×10^{-4}
80 m	0.14	9.2×10^{-3}	9.3×10^{-4}	5.9×10^{-4}
July 26 - Aug. 22, 1977				
40 m	0.30	3.3×10^{-3}	6.8×10^{-4}	3.4×10^{-4}
80 m	0.19	3.4×10^{-3}	10.0×10^{-4}	8.9×10^{-4}
Sept. 14 - Nov. 1, 1977				
40 m	0.05	1.2×10^{-3}	2.1×10^{-4}	7.3×10^{-4}
80 m	0.07	5.2×10^{-3}	4.3×10^{-4}	7.7×10^{-4}

*1 These values are wighted means of the F and B fractions given in Table 1.

^7Be and other nuclides in settling matter To find the particulate flux of ^7Be , sediment traps were deployed three times in the water of 40 and 80m depths at St. P in 1977. The specific activities of ^7Be , ^{234}Th , ^{228}Th , ^{230}Th and ^{232}Th in settling matter collected are given in Table 2. The specific activities of ^7Be are almost the same for all the samples. This fact is to be noted, because ^7Be of the surface origin could be expected to decrease with increasing depth, if ^7Be would be irreversibly incorporated into particles and sink at a definite velocity. ^{230}Th and ^{228}Th were more concentrated in settling particles collected at 80m depth than in those at 40m depth. The specific activities of ^{232}Th observed at 80m depth were nearly equal to those observed at 40m depth, except for those observed during the period from July 26 to Aug. 22, 1977.

For further discussion on the mechanism of the uptake of these nuclides into particulate matter, we calculated the activity ratios of $^7\text{Be}/^{234}\text{Th}$, $^{228}\text{Th}/^{234}\text{Th}$, $^{230}\text{Th}/^{234}\text{Th}$ and $^{232}\text{Th}/^{234}\text{Th}$ of the sediment trap samples (Table 2). The $^7\text{Be}/^{234}\text{Th}$ ratios at 40m depth are higher than those at 80m depth in the summer of 1977. This fact can be explained by a mechanism that sinking particles continuously pick up ^{234}Th or these nuclides are exchanged between the sinking particles and seawater. If the above mechanisms does not work, $^7\text{Be}/^{234}\text{Th}$ in sinking particles should increase with depth, because the half-life of ^7Be is longer than that of ^{234}Th . This denies the removal of these nuclides only from the surface seawater into sinking particles (e.g. fecal pellets) which are produced in the surface water and sink without any exchange of those nuclides with seawater. The small $^7\text{Be}/^{234}\text{Th}$ ratio observed at 40m depth in autumn may be due to the warm Tsugaru current flowing into the surface of the Bay in that season.

Net fluxes of ^{234}Th and ^7Be

The net removal of ^{234}Th estimated from its deficiency from U-238 in seawater and the data obtained by MINAGAWA and TSUNOGAI (1980) and the flux observed with sediment traps are compared in

Table 3. The observed fluxes of ^{234}Th for sediment traps were about ten times smaller than those estimated. This small trapping efficiency may have been caused by the shape of the traps and the current velocity of seawater as discussed by GARDNER (1980). Assuming the $^7\text{Be}/^{234}\text{Th}$ ratio (R) of material collected in the sediment traps is equal to that of whole settling matter and the particles sink downward unilaterally, we can obtain the net downward flux (F_{Be}) and the residence time (τ_{Be}) of Be in seawater as follows;

$$F_{\text{Be}} = \frac{1}{\tau_{\text{Th}}} \times A_{\text{Th}} \times R, \tau_{\text{Be}} = \frac{A_{\text{Be}}}{F_{\text{Be}}}$$

where A_{Be} and A_{Th} are the inventories of ^7Be and ^{234}Th (dpm/m²), respectively, and τ_{Th} is the residence time of ^{234}Th (days) which has been obtained by MINAGAWA and TSUNOGAI (1980). The results calculated are also given in Table 3. The fluxes of ^7Be obtained from the sediment trap experiment were also about ten times smaller than calculated ones as those of ^{234}Th . The residence time of Be in a 80 m water column is estimated to be 120 – 160 days, which is somewhat longer than that of ^{234}Th . However, the above estimation includes rather large uncertainties and so the difference between the residence times of the two nuclides may not be significant. Therefore, we can conclude that Be is one of elements like ^{234}Th which are actively removed from seawater. Recently, AABOE *et al.* (1981) have also reported the residence time of Be in Long Island Sound to be around 4 days, where the residence time of Th is obtained to be 1 day (ALLER and COCHRAN, 1976).

The active removal of ^7Be in coastal seawater also suggests us its usefulness in studying the mechanisms of particulate removal of metals in seawater and of the downward transport of particles from the surface to the deep. In the previous studies (SILKER, 1972; YOUNG and SILKER, 1974), the eddy diffusivity of the surface seawater was estimated with ^7Be as a radioactive tracer like tritium. The vertical diffusivity obtained, however, may not be real

Table 3 Observed inventories of ^{234}Th and ^7Be in seawater and their observed and calculated particulate fluxes at the bottom of each water column.

A. ^{234}Th

Depth (m)	$^{234}\text{Th}/^{238}\text{U}^{*1}$ in seawater	Residence time of Th (days)	Calculated flux (dpm/m ² day)	Observed flux (dpm/m ² day)
0-40	0.64	61	9.4×10^2	0.8×10^2
0-80	0.60	53	2.1×10^3	2.7×10^2

*1 Cited from MINAGAWA and TSUNOGAI (1980).

B. ^7Be

Depth (m)	$^7\text{Be}/^{234}\text{Th}^{*1}$ in settling matter	Residence time of Be (days)	Calculated flux (dpm/m ² day)	Observed flux (dpm/m ² day)
40	0.22-0.30	90-120	$(2.1-2.8) \times 10^2$	0.2×10^2
80	0.14-0.19	120-160	$(2.9-4.0) \times 10^2$	0.5×10^2

*1 Given in Table 2.

*2 See the text for the way of calculation.

unless the effect of particulate transport is taken into consideration at least in coastal sea, where the active particulate transport of ^7Be takes place.

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